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Article

Agricultural Methods for Toxicity Alleviation in Metal Contaminated Soils: A Review

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Due to the fact that possible risk associated with soil-crop-food chain transfer, metal contamination in croplands has become a major topic of wide concern. Accumulation of toxic metals in edible parts of crops grown in contaminated soils has been reported from number of crops including rice, soybean, wheat, maize, and vegetables. Therefore, in order to ensure food safety, measures are needed to be taken in mitigating metal pollution and subsequent uptake by crop plants. Present paper critically reviewed some of the cost effective remediation techniques used in minimizing metal uptake by crops grown in contaminated soils. Liming with different materials such as limestone ($CaCO_3$), burnt lime (CaO), slaked lime [$Ca(OH)_2$], dolomite [$CaMg(CO_3)_2$], and slag ($CaSiO_3$) has been widely used because they could elevate soil pH rendering metals less-bioavailable for plant uptake. Zn fertilization, use of organic amendments, crop rotation and water management are among the other techniques successfully employed in reducing metal uptake by crop plants. However, irrespectively the mitigating measure used, heterogeneous accumulation of metals in different crop species is often reported. The inconsistency might be attributed to the genetic makeup of the crops for selective uptake, their morphological characteristics, position of edible parts on the plants in respect of their distance from roots, crop management practices, the season and to the soil characteristics. However, a sound conclusion in this regard can only be made when more scientific evidence is available on case-specific researches, in particular from long-term field trials which included risks and benefits analysis also for various remediation practices.

Key words: Toxic metals, Land pollution, Accumulation in Crops, Mitigating measures

Introduction

Trace metals contamination of agricultural soils has become one of the most significant environmental problems today (Chand et al., 2012). Metal uptake by crop plants can have strong adverse impacts on human health through the food chain (Sadon et al., 2012). In fact, in addition to the crop yields get contaminated, excessive metals in the plant can result in decreased crop yield too due to the inhibition of plant metabolic processes (Singh and Aggarwal, 2006). As illustrated in Table 1, accumulation of toxic metals in edible parts of crops grown in contaminated soils has been reported for number of crops including rice (Simmons et al., 2006; Singh et al., 2011), soybean (Salazar et al., 2012), wheat (Brunetti et al., 2012), maize (Mohammad and Zahra, 2012) and vegetables (Singh et al., 2012). Furthermore, in many cases, the metal content in edible parts of the crop exceeds the maximum permissible limits imposed by Codex Alimentarius Commission (Table 2). It has been reported that over 80% of the intake of metals such as Cd and Pb could be from crop-based foods (Marzec and Schlegel-Zawadzka, 2002). However, the accumulation of metals in agricultural crops and thresholds of dietary toxicity in soil-crop system may vary with several factors (Islam et al., 2007; Cooper et al., 2011). In this context, Singh et al. (2012) reported that accumulation amount depends on crop species and variety. While Tangahu et al. (2011) and Islam et al. (2007) reported that nature of the soil (soil pH, organic matter content, clay mineral and other soil physical and biochemical properties) and climatic conditions are also important in determining the metal content in crop plants. According to Cooper et al. (2011), season and crop management practices too are involved in determining the metal content in crops. As reported by Islam et al. (2007), the interactions of soil-plant roots-microbes play important roles in regulating heavy metal movement from soil to the edible parts of the crops.

Apart from the metals with unknown biological

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functions (Cd, Cr, Pb, Co, Ag, Se, and Hg), essential elements (Fe, Mn, Zn, Cu, Mg, Mo, and Ni) also keep accumulating in soils by means of wastewater irrigation, animal manures and sewage sludge application, use of fertilizer and agrochemicals (Thomas et al., 2012). In the toxicological point of view, the essential elements are also important, because, at higher concentrations they too can be toxic to plants as well as to dietary intake levels (Karavoltsos et al., 2002). With the continuous addition of undesirable metals into the environment, remediation of contaminated soils receives increasing attention (Yeh and Pan, 2012). Depending on the resource availability, severity of the problem, nature of the metals and contaminated soil etc., different remediation methods have been employed in cleaning up contaminated lands (Chen, 2000). In this context, systematic remediation technologies such as bioremediation, physical/chemical remediation and integrated remediation are among the widely used techniques (Luo, 2009). In the present manuscript, we critically review some of the cost effective remediation techniques used in minimizing metal uptake by crops grown in contaminated soils.

Metal uptake by crops could effectively be minimized through alteration of some physical and chemical characteristics of soil such as pH, redox potential, cation exchange capacity, composition and concentration of humic substances etc., which have influence on immobilization of toxic elements in soil (Tlustoš et al., 2006). It has been frequently reported that lime, phosphates, or organic matter residues could be employed in altering the

Table 1. Heavy metal accumulation in edible parts of different crops ($\mu g\ g^{-1}$ dry wt.) grown on metal contaminated soils.

Crop	Zn	Cu	Pb	Cd	Ni	Reference/s
Radish	67	29	03	06	10	Singh et al. (2012)
Carrot	28	13	15	02	12	
Potato	96	26	43	30	27	
Onion	49	17	13	04	28	
Spinach	86	29	23	20	11	
Amaranthus	87	26	35	28	16	
Fenugreek	64	88	36	04	05	
Mustard	70	32	27	20	26	
Cauliflower	53	13	29	03	40	
Cabbage	32	12	34	08	29	
Soybean	71	51	18	02	16	
Bean	53	48	11	02	17	
Tomato	29	24	6	03	17	
Brinjal	29	43	2	02	01	
Peas	71	17	19	01	15	
Okra	65	71	33	07	07	
Rice	21-48	3.7-4.8	0.1-0.3	0.1-0.4	2.3	Machiwa (2010); Luo et al. (2012)
Wheat	28-48	4.6-6.0	0.1-0.6	0.03-0.1	0.03-0.08	Brunetti et al. (2012)

Table 2. Maximum allowable limits of Cd in different crops adopted by Codex Alimentarius Commission (extracted from Makino et al., 2010).

Crop	Cd concentration ($\mu g\ g^{-1}$ dry wt.)	Reference/s
Polished rice	0.4	Codex Alimentarius Commission (2005; 2006)
Wheat grains	0.2	
Edible root and stem vegetables	0.1	
Potato (peeled)	0.1	
Beans (except soybeans)	0.1	
Leafy vegetables	0.2	
Other vegetables	0.05	

metal immobilization in soils subsequently reducing the plant uptake (Bolan and Duraisamy, 2003). Crop rotation with hyper-accumulator species as a component before being establishment of main crop is also recognized as being viable technique (Murakami et al., 2009) and cultivation of non-food crops or proven lesser absorptive crops on the contaminated land have also received considerable attention (Takeda et al., 2007). Though the success is vastly dependent upon the availability of reliable water source, management of water is considered as being effective for some crops such as rice (Makino et al., 2010).

Commonly Used Methods

Use of inorganic amendments The efficacy of inorganic amendments in decreasing the metal bio-availability is reported to be higher than that of organic amendments mainly due to the fact that creation of additional binding sites for heavy metals and alteration of pH in the soil solution (Grabowska, 2011). Some of the inorganic amendments available for use include; Al-montmorillonite, clinoptilolite, diammonium phosphate, ferrous sulfate, hydroxyapatite, lime, manganese oxides, red mud, synthetic zeolites, and water treatment sludge. The use of them has been encouraged by their effectiveness, affordability at low cost and availability in large scale being many of them are industrial by-products.

Liming Liming has long been used as a proven inorganic soil amendment which can elevate soil pH rendering metals less-bioavailable for plant uptake (Ciecko et al., 2004, Puschenreiter et al., 2005). Depending on the acid-neutralizing capacity, different liming materials such as limestone ($CaCO_3$), burnt lime (CaO), slaked lime [$Ca(OH)_2$], dolomite [$CaMg(CO_3)_2$], and slag ($CaSiO_3$) etc., are available for use (Bolan and Duraisamy, 2003). Despite the effectiveness of liming has repeatedly been reported using field as well as pot experiments, the effect of liming on metal uptake may vary with the crop, the metal of concern and with soil characteristics (Puschenreiter et al., 2005). Not like in acidic soils, lime application in saline soils may however be less-effective in reducing metal uptake. Cattani et al. (2008) reported that application of lime decreased the Cd concentration in rice by 25%. Uptake of metals such as Cd, Zn and Pb by wheat has also been reported to be decreased by the addition of lime (Tlustoš et al., 2006). However, reports are also available on little or no effect of liming on Cd uptake by crops (Shaha et al., 2012). Aguilar et al. (2004) studied the relationships between physicochemical soil parameters and Pb mobility in soils and reported that $CaCO_3$ is predominantly responsible for Pb retention in soils. Elevated pH due to liming leads Cd^{2+} to form Cd $(OH)^+$ which comparatively has a strong affinity to soil adsorption sites than with Cd^{2+}. However, making Cd less-available through elevated pH is not always effective in reducing plant uptake of the metal. Among the liming induced biological alterations, enhanced activity of soil C (Delorme et al., 2001) and changes in bacterial diversity (Da Silva and Nahas, 2002) have been reported. However, Blake and Goulding (2002) stressed the need for conducting long term investigations on the effect of liming strategies on element mobility in soil, their uptake by plants and associated soil characteristics such as soil pH, sorption capacity and organic matter content. Further stepping to this, Hough et al. (2003) derived some predictive models of element uptake by selected crops based upon a database of soil characteristics to optimize liming strategies. As stated by Knox et al. (2001), lime application needs to be repeated over several years in order to ensure satisfactory metal immobilization, thus, required quantity may be much higher than most of the other inorganic amendments. Furthermore, incorporation of much lime into the soil may result in immobilization of essential nutrients also (Conyers, 2002).

Putwattana et al. (2010) reported that apart from lime, other inorganic amendments such as zeolite and manganese oxide can reduce Cd uptake in rice and wheat. According to Shi et al. (2005), available silica arising from the zeolite plays a significant role in decreasing Cd uptake in plants, which was in line with Chen et al. (2000) who too reported that zeolite was more effective in suppressing Cd uptake by rice and wheat. Red mud, another inorganic amendment has been employed in assessing the reduction in metal uptake by *Festuca rubra* and *Amaranthus hybridus* grown on contaminated soils (Friesl et al., 2003). Their findings showed that reduction in plant uptake of Cd, Pb and Zn by two plant species ranged 38-87%, 50-81% and 66-87%, respectively. The effectiveness of red mud in reducing heavy metal uptake by oilseed rape, pea, and wheat was also proved by Lombi et al. (2002). Red mud may however contain Cr and As, thus attention must be paid when large scale application is planned (Friesl et al., 2004). Commercial MgO has also been effectively employed in reducing Cd uptake by paddy plant grown on Cd-contaminated soils (Kikuchi et al., 2008).

Fertilization Fertilization could effectively be used in two different ways to minimize metal accumulation in crops. The first is the direct approach for example, use of Zn fertilization to reduce Cd uptake by plants. As the indirect approach, once hyper-accumulated plants as a component of crop rotation are grown on metal polluted sites, fertilization could enhance the metal uptake resulting low or no metals remain to be uptake by subsequent crops. Following the direct approach, Cu (2008) reported a significant reduction in Cu accumulation in *Brassica juncea* due to phosphorous fertilization. According to them, at the application rate of 80 Kg P_2O_5 ha^{-1}, the height and biomass of the plant were increased by 30% and 31% respectively, while reducing Cu content by 14% compared to the control. Rojas-Cifuentes et al. (2012) studied the effect of Zn fertilizer ($ZnSO_4$) on Cd accumulation in the seeds of nine crop species. Their results revealed that Zn fertilizer lead to lower the Cd content in flax, sunflower, soybean, and durum wheat, however, no consistently was found in the reductions among different crops. Furthermore, they reported that reductions may not have any significant impact on the marketability of those seeds. According to Chaney et al. (2009), liming along with Zn fertilizer strongly reduced Cd uptake by lettuce. Dar et al. (2012) conducted a study with wheat grown on cadmium polluted soil and reported that Zn application could reduce Cd content in grains. Among different explanations, Moustakas et al. (2011) suggested that competitive transport and absorption interaction between these two ions as the reason behind the reduction in Cd uptake caused by Zn fertilization. According to Harris and Taylor (2001), reduction in Cd accumulation in durum wheat seeds by Zn might be due to alleviation of Zn stress through application of Zn fertilizer. Cooper et al. (2011) too have conducted a study with wheat and based on the results, they stressed a potential risk of higher levels of Cd accumulation if P-fertilizers with a higher Cd content are used. In accordance with their view, Thomas et al. (2012) reported that heavy metal accumulation in the soil does not depend on levels of phosphate fertilizer but on the levels of heavy metal impurities in the phosphate fertilizer. Due to the fact that both antagonist and synergistic interactions between Zn and Cd have been reported, uptake, translocation, and remobilization of Cd under Zn enriched soils are still needed further elucidation (Rojas-Cifuentes et al., 2012**)**. The ionic structures and electronegativities of Cd and Zn are found to be much similar to each other though have different ionic radii (Zn^{2+} = 0.074 nm, Cd^{2+} = 0.097 nm), which might be at least partly the reason for species-defendant manner of response.

On the other hand, use of K fertilizers could increase Cd uptake by plants (Zhao et al., 2003), which is useful when phytoremediation is practiced with hyper-accumulated plants. However, various potassium forms differed in terms of their effects on Cd accumulation in plants. If K is added as KCl, soil Cd can form relatively stable complexes such as $CdCl^+$ and $CdCl_2$ (Norvell et al., 2000), which tends to shift Cd from solid to soil solution enhancing Cd solubility and subsequent bioavailability. In particular, in calcareous soils where metal availability is normally be limited by high pH, chloride plays a key role in Cd uptake by crops, (Makino et al., 2006). However, if K is added as K_2SO_4, similar results are not expected because sulphate complexes with Cd are found to be less-stable. If fertilizers with high NH_4 are used, pH of the soil solution decreases leading to increase in metal bioavailability.

Use of organic amendments Generally, organic amendments are recognized as being beneficial in several different ways to enhance crop production (Chiu et al., 2006). Addition of compost, farmyard manure (FYM) and organic wastes etc., is often practiced in immobilization of heavy metals and soil amelioration of contaminated soils (Clemente et al., 2005). However, the degree of success depends on many factors including the nature of the soil (soil type, salt content, pH and redox potential), nature of the organic matter and their microbial degradability, and on metals of concerned (Walker et al., 2004). Angelova et al. (2010) investigated the impact of organic amendments (peat, compost and vermicompost) on uptake of heavy metals (Pb, Zn, Cd, and Cu) by potato *(Solanum tuberosum L.)* plants. They found a correlation between the quantity of the mobile forms and the uptake of metals by potato and reported decreased heavy metal content in potato peel and tubers due to successful immobilization of Pb, Cu, Zn, and Cd by organic amendments. Furthermore, compost as well as vermicompost was found to be the best amendments, which expressed the highest reduction (10%) in metal uptake.

Addition of farmyard manure (FYM), a natural organic amendment could improve the physical characteristics and nutrient availability of soil while decreasing metal uptake by plants (Putwattana et al., 2010). Alamgir et al. (2011) from their study on effect of FYM on Cd and Pb accumulation by Amaranth observed that application of FYM could decrease Cd and Pb uptake. Furthermore, the contents of both Cd and Pb in the shoot and root of Amaranth showed a significantly negative correlation ($r = -0.84$ to -0.87) with the rates of FYM applied to the soil. Their findings are in line with Yassen et al. (2007), who observed reduction in Cd concentration (from 41% to 31%) in spinach plant grown on contaminated soil as a result of application of FYM. Kibria et al. (2011) reported 27% and 62% reduction in the grain Cd content of rice by the addition of FYM and lime mixed with FYM respectively. However, contrary to this, Kos et al. (2004) reported increased uptake of Zn and Cd by *Amaranthus* spp. after the incorporation of FYM into the soil. According to Li et al. (2006), phytoavailability of Cd in cherry-red radish (*Raphanus sativus*) was reduced by the addition of chicken and pig manure composts. Different responses of FYM to phytoavailability of metals might be due to the variation in source of FYM and plant species of concern (Putwattana et al., 2010).

In the case of acidic soils, increased soil pH by the addition of FYM could decrease the availability of metals through increased adsorption. In fact, high pH could result in more adsorption sites through increased negative charges on the soil surface. Furthermore, addition of organic matter increases CEC of soil, thus hydroxyl, phenoxyl, and carboxyl groups of the soil with added organic materials could alter the adsorption and complex of heavy metals in soil. Accordingly, Kabata-Pendias (2001) reported that after the addition organic matter, Cu, Zn, Pb and Cd could accumulate in organic horizons of soil as stable forms. However, as reported by Cooper et al. (2011), regular application of organic matter may reduce plant uptake of metals because addition of organic compounds to the soil could lead in increased chelation of metals reducing plant availability of them.

Other Agronomic Practices

Crop rotation As stated by Islam et al. (2007), crop rotation can also be effective in altering the bioavailability and crop accumulation of heavy metals. In this context, several options are available for selecting crops to be cultivated on the contaminated sites. Of course, the land can be used in producing non-food crops/industrial crops by which bio-magnification of metals through food chain is minimized. In this regard, depending on the climatic factors and other requirements, fiber crops such as flax, cotton and hemp (Yanchev et al., 2000: Angelova et al., 2004), energy crops such as corn, jatropha and gliricidia or other industrial crop could be selected for the cultivation.

As another option, phytoremediation, a cost-effective green technology which uses living green plants to uptake and concentrate metals from contaminated soil into the aboveground shoots, could be used before being establishment of food crops. In fact, phytoremediation, due to its inherited merits such as ability to selective uptake, active translocation and bioaccumulation in the entire plant body, has gained widespread attention (Zhuang et al., 2007; Tangahu et al., 2011). However, as recognized by Li et al. (2012), the technique is better suited for agricultural lands with low to moderate metal contamination. Phytoextraction can be done with hyper-accumulator species, whose above ground parts generally accumulate relatively high levels of metals. As reported by Teofilo et al. (2010), potential hyper-accumulators should have high bioconcentration factor (BCF) and high translocation factor (TF) values. Generally they are to achieve a shoot-to-root metal-concentration ratio greater than one, while nonaccumulating plants typically have a shoot-to-root ratio considerably less than one. Ideally, hyperaccumulators thrive well in toxic environments, require little maintenance and produce high biomass (Salido et al., 2003). As assessed with these important traits, many plant species could be recognized as successful hyper-accumulators of metals and metalloids (Tangahu et al., 2011). However, majority of the identified species is known to be slow growers, further their mechanical harvesting is difficult also, thus the technique is not recommended for large scale phytoextraction (Li et al., 2012).

Water management Makino et al. (2010) reported that water management is effective in particular for reducing Cd uptake by rice plants. In this regard, flooding the entire paddy field during the period between tilling to head formation is recommended (Arao et al., 2009), though it may result in operational difficulties for machineries. Flooding may also lead to release of methane as well as As from the paddy soil due to possible reducing conditions (Arao et al., 2009). Contrary to this, Angle et al. (2003) reported that both growth and metal uptake of *Thlaspi*, *Alyssum*, and *Berkheya* were increased with increasing soil moisture contents. Therefore, they encouraged irrigation for hyper-accumulator plants to enhance metal uptake.

Conclusion

It is clearly evident from the published literature that a number of crop species have the ability to uptake and accumulate metals when they grow on contaminated soils. Due to the possible risk associated with transfer of metals through soil-crop-food chain, increased attention has been paid on mitigating measures adoptable for diverse conditions. However, marked variations in metal uptake and their distribution to various plant parts have been often reported, thus need for further elucidations on as to how different crop species respond to different mitigating measures is stressed. Such investigations must accompany an analysis between risks and benefits of various cultivation and remediation practices also.

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