Heavy Metal Contamination and Remediation in Asian Agricultural Land

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Abstract
This paper reviews contamination status, sources and remediation of heavy metals in agricultural land of most Asian countries, with an emphasis to China, which are undergoing rapid economic development. Some farmland soil in suburbs of most cities and sewage irrigation district in China are polluted to some extend with heavy metals such as Cd, As, Zn, Cu, and Hg, consequently resulting in metal contamination of agricultural products and pose a potential risk to human health. It has been reported that foodstuff such as vegetable, grain and domestic animal feed are severely contaminated with heavy metals in the Asian countries. The sources of heavy metals in arable lands in most Asian countries include natural source, mining, smelting, agrochemicals and sewage sludge applications, and livestock manure uses. Systematic remediation technologies for contaminated soils are addressed as well, which include physical/chemical remediation, phytoremediation, microbial remediation and integrated remediation.
Key words: soil contamination, heavy metal, risk assessment, remediation, Asia

1. Soil heavy metal contamination in China and other Asian countries

(1) Accumulation and impacts of heavy metals in agricultural soils

The South and Southeast Asian countries, such as Peninsular Malaysia, Vietnam, India, Thailand, Philippines, Indonesia, Bangladesh, and Pakistan have paid much attention to contamination of agricultural soils and crops by heavy metals due to their potential effects on human health and long-term sustainability of food production in the contaminated areas. It was reported that in Korea the average concentrations of Cd, Cu, Pb, and Zn in surface layer of the rice paddy soils (0-15cm) were 0.11 mg kg^{-1} (ranged from 0 to 1.01), 0.47mg kg^{-1} (0-41.6), 4.84 mg kg^{-1} (0-66.4), and 4.47 mg kg^{-1} (0-96.7), respectively. In orchard fields, the average contents of Cd, Cu, Pb, Zn, As, and Hg in surface soils (0-20cm) were 0.11 mg kg^{-1} (ranged from 0-0.49), 3.62 mg kg^{-1} (0.03-45.3), 2.30 mg kg^{-1} (0-27.8), 16.60 mg kg^{-1} (0.33-106), 0.44mg kg^{-1} (0-4.14), and 0.05 mg kg^{-1} (0.01-0.54), respectively (Jo and Koh, 2004). In Japan the estimated average levels of Cd, Cu, and Zn in rice were 75.9 mg kg^{-1}, 3.71 mg kg^{-1}, and 22.9 mg kg^{-1}, respectively. The average levels of Cd, Cu, and Zn in rice-fields were 446 mg kg^{-1}, 19.5 mg kg^{-1}, and 96.4 mg kg^{-1}, respectively (Herawati *et al.*, 2000). China is facing soil contamination problems, especially heavy metal pollution (Luo and Teng, 2006; Brus *et al.*, 2009). It was estimated that nearly 20 million hectares arable soils (approximately one fifth of the total areas of farmland) were contaminated by heavy metals in China, which was assumed to result in a reduction of more than 10 millions tons of food supplies in China annually (Wei and Chen, 2001). The proportion of exchangeable fractions of Cd in soil of Zhangshi irrigation area in Shenyang, Liaoning province, with the history of sewage irrigation over 45 years, was much higher than that of Cu and Pb. It was suggested that Cd would be the most mobile element in the soil and more available to crop, with great risk of moving into the food chain. As a consequence, Cd contamination in the arable soils became the most serious problem in this region (Xiong *et al.*, 2003). Other areas including Taihu Lake plain and Pearl River Delta region, in which have been under rapid economic development, were all found moderate to serious contamination by heavy metals recently (Huang *et al.*, 2007; Hang et al., 2009).

Accumulation of heavy metals in crops grown in metal-polluted soil may easily cause damage effect on human health through food chain. Fu *et al.*, (2008) contacted an investigation on heavy metal contents in rice sampled from Taizhou city in Zhejiang province, China and found that the geometric mean of Pb in polished rice reached 0.69 mg kg^{-1}, which was 3.5-folds higher than the maximum allowable concentration (MAC) (0.20 mg kg^{-1}) of the safety criteria for milled rice. Cd contents in 31% of the rice samples exceeded the national MAC (0.20 mg kg^{-1}), and the arithmetic mean also slightly exceeded national MAC. In Dabaoshan Mined area of Guangdong Province, China, surrounding farmland has been seriously contaminated by Cd and other toxic metals as a result of long-term mining (mainly iron and copper) as well as discharge of untreated wastewater. The average concentration of cadmium in rice exceeded 150 times of the State Food and Health Standards (Lin *et al.*, 2005), which has caused a health risk to local residents. Norra *et al.* (2005) reported the concentration of As in the winter wheat grain could reach 0.7 mg kg^{-1} cultivated in agricultural area of west Bengal Delta Plain, irrigated with As rich groundwater.

2. Sources of heavy metals in agricultural soils

It is important to identify the sources and status of soil contamination by toxic metals so as to take proper treatments to reduce soil contamination and to keep sustainable agricultural development.

(1) Natural source

The initial sources of heavy metals in soils are the parent materials from which the soils were derived, but the influence of parent materials on the total concentrations and forms of metals in soils is modified to varying degrees by pedogenetic processes (Herawati *et al.*, 2000). In areas affected lightly by human activities, heavy metals in the soils derived mainly from pedogenetic parent materials, and metals accumulation status was affected by several factors such as soil moisture and management patterns. A research conducted in Gansu province, China, by Li (2008) concluded that the main factor for heavy metals accumulation was lithological factor in three arid agricultural areas. It is reported that soil *aqua regia* soluble fraction of Co, Ni, Pb, and Zn were highly correlated with soil Al and Fe. These elements were associated with indigenous clay minerals in the soil high in Al and Fe.

(2) Mining

There are different sources of metal contamination in mining areas, including grinding, concentrating ores and tailings disposal (Wang *et al.*, 2004; Adriano, 1986). Inappropriate treatment of these tailings and acid mine drainage could pollute the agricultural fields surrounding the mining areas (Willians *et al.*, 2009). Take Tongling copper mine in Anhui province in China as an example, metal mining had been an important economic base in this area from ancient time. The major mining areas have been concentrated in a narrow star-shaped basin called Fenghuang Mountain. Long-term mining activities in this area had caused widespread metal pollution. The soil concentration of average total Cu was 618 mg kg^{-1}, with a wide range of 78-2830 mg kg^{-1}. Lead concentration in soil also showed a large variability with a mean of 161 mg kg^{-1}. The total Zn concentration varied from 78 to 1280 mg kg^{-1}, with an average of 354 mg kg^{-1} (Wang *et al.*, 2004). It was reported that the majority of the agricultural soils were contaminated with As. High As concentration in these soils may be attributed to arsenopyrite which is known to occur in many areas of Southeast Asia, especially in tin mining regions (Patel *et al.*, 2005).

(3) Smelting and flying ash

It was reported that atmospheric deposition was responsible for 43%-85% of the total As, Cr, Hg, Ni and Pb inputs to agriculture soils in China (Luo *et al.*, 2009). Actually, most of heavy metallic pollutants in air derive from flying ash coursed by highly anthropogenic activities (Liu *et al.*, 2005), such as electric power, mining, metal smelting and chemical plants. Total trace element deposition (wet and dry) to agricultural soils was calculated by Luo (2009) from the average deposition fluxes of each element and the total agricultural land area (1.22×10^8 hectares in 2005) in China. In this study, it was reported that the deposition from atmosphere in China were generally higher than those in New Zealand except for Zn and comparable to the area of Tokyo Bay. The most common elements from atmospheric deposition were Hg, Pb, As, Cd and Zn, and non-ferrous metal smelting and coal combustion were two of the most important ways contributing to metal pollutants in the air. Streets *et al.* (2005) have pointed out that, among the Hg emissions in China, approximately 38% of Hg comes from coal combustion, 45% from non-ferrous metal smelting, and 17% from miscellaneous activities, of which battery and fluorescent lamp production and cement production are of most importance. Zn was the metal deposited in agricultural soils in the largest amount from the atmosphere in China, and Pb and Cu followed.

(4) Fertilizers and agrochemicals

Heavy metals input to arable soils through fertilizers courses increasing concern for their potential risk to environmental health. Lu *et al.* (1992) reported that the phosphate fertilizers were generally the major source of trace metals among all inorganic fertilizers, and much attention had also been paid to the concentration of Cd in phosphate fertilizers. However the concentration of Cd in both phosphate rocks and phosphate fertilizers from China was in general much lower than those from the USA and European countries. It should be concerned that although the contents of toxic metals in most of the fertilisers in China were lower than the maximum limits, the trace elements input to agricultural land were still worth concern, since the annual consumption of fertilizers accounted to 22.2, 7.4 and 4.7×10^6 tons for N, P and K fertilizers (in pure nutrient), respectively (Luo *et al.*, 2009; NBSC, 2006). Traditionally, agriculture has been the main base of the economies in this region. In some of the countries mentioned above, phosphatic fertilisers have been used for long periods. For instance, the great majority of agricultural soils in Malaysia are heavily fertilized by this kind of fertilizers, which was reported by Zarcinas *et al.* (2004). Regression analysis resulted in that log *aqua regia* soluble As, Cu, Cd, and Zn in soil are significantly correlated with log *aqua regia* soluble P. Soils in these southern Asian countries have P requirements, so that histories of P fertilizers addition, with associated with impurities (Cd, Cu, As, and Zn), seem to be greater on these countries (Zarcinas *et al.*, 2004).

Agricultural use of pesticides was another source of heavy metals in arable soils from non-point source contamination. Although pesticides containing Cd, Hg and Pb had been prohibited in 2002, there were still other trace elements containing pesticides in existence, especially copper and zinc. It was estimated that a total input of 5000 tons of Cu and 1200 tons of

Zn were applied as agrochemical products to agricultural land in China annually (Luo *et al.*, 2009; Wu, 2005).

Coca, groundnut, mustard and rice had elevated concentrations of heavy metals (especially Cu and Zn) assessed when compared to the other plants (cabbage, oil palm, aubergine, lady's fingers). This may be contributed by the widespread use of Cu and Zn pesticides on these crops. A survey also showed that heavy metal concentration in surface horizon and in edible parts of vegetables increased over time. Pandey *et al.* (2000) reported that the metal concentration in soil increased from 8.00 to 12.0 mg kg^{-1} for Cd, and for Zn from 278 to 394 mg kg^{-1}. They also suggested that if the trend of atmospheric deposition is continued, it would lead to a destabilizing effect on sustainable agricultural practice and increase the dietary intake of toxic metals. Sinha *et al.* (2006) concluded that the vegetables and crops growing in such area constitute risk due to accumulation of metals in India. The researchers also studied the effect of municipal wastewater irrigation on the accumulation of heavy metals in soil and vegetables in the agricultural soils in India. The mean concentrations of Pb, Zn, Cd, Cr, Cu and Ni in waste water-irrigated soil around Titagarh region were 130, 217, 30.7, 148, 90.0 and 104 mg kg^{-1}, respectively. And also, the concentrations of Pb, Zn, Cd, Cr and Ni in all vegetables (pudina, cauliflower, celery, spinach, coriander, parsley, Chinese onion and radish) were over the safe limits.

The industrial effluents often contain many heavy metals. In industrial areas, many agricultural fields are inundated by mixed industrial effluent or irrigated with treated industrial waster water. The plant available metal content in soil showed the highest level of Fe, from 529 to 2615 mg kg^{-1}, and lowest level of Ni, from 3.12 to 10.5 mg kg^{-1}. The results also suggested that the accumulation of Cr in leafy vegetables was found more than fruit bearing vegetables and crops (Sinha *et al.*, 2006).

(5) Wastewater irrigation

Farmland irrigated by wastewater in China accounted to 36, 180, 000 hectares, occupying approximately 7.3% of total irrigation area (Bulletin of Environmental Status in China, 1998). Sewage irrigation can alleviate the water shortage to some extent, but it can also bring some toxic materials, especially heavy metals, to agricultural soils, and cause serious environmental problems. This is particularly a problem in densely populated developing countries where pressure on irrigation water resources is extremely great, especially northern dry land in China. The amount of wastewater released had reached 5.25×10^{10} tons in 2005, of which industrial wastewater accounted for 2.43×10^{10} tons (Luo *et al.*, 2009; SEPAC, 2006a). The most important wastewater irrigation areas contain Zhangshi wastewater irrigation area in Shenyang Liaoning province, Xi'an wastewater irrigation area in Shaanxi province, Beijing sewage irrigation area, Shanghai wastewater irrigation area. In Chhattisgarh, central India, soil was irrigated with As polluted ground water. People in this region were suffering from arsenic borne diseases. The arsenic concentration ranged from 15 to 825 μg L^{-1} in the polluted water, exceeding the permissible limit, 10 μg L^{-1}. The contaminated soil had the median level of 9.5 mg kg^{-1} (Patel *et al.*, 2005). Many industrial plants in this region operate without any, or minimal, wastewater treatment and routinely discharge their waster into drains, which either contaminate rivers and streams or add to the contaminant load of biosolids (sewage sludge). Biosolids are increasingly being used as soil ameliorants and streams and rivers are the primary source of water for irrigation.

(6) Sewage sludge application

Although the contents of toxic metals in sewage sludge had also been markedly reduced, and most of them were below the national discharge standard of pollutants for municipal wastewater treatment plants, due to the huge increase in the amount of wastewater treated, the sewage sludge produced increased rapidly. According to data from SEPAC (2006), approximately 4.6×10^{6} tons (dry weight) of municipal sewage sludge was produced in China in 2005. It was estimated that the direct agricultural land application of sewage sludge in China had a ratio about 10% (Luo *et al.*, 2009). Copper is strongly attached to organic material and may be added as a contaminant with organic soil amendments. There is also now a considerable body of evidence documenting long-term exposure to high concentrations of heavy metals (e.g. Cu) as a result of past applications of sewage sludge (McGrath, 1994), Cu and Zn from animal manures (Christie and Beattie, 1989), and past applications of Cu-containing fungicides (Zelles *et al.*, 1994).

In the agriculture areas of Hyderabad, Pakistan, researches studying the effect of long time applied wastewater sludge on the concentrations of heavy metals in soil irrigated with fresh canal water (SIFW) and soil irrigated with waste water (SIDWS) are as follow: the total mean concentrations of Cu, Zn, Pb and Cd are 11.2, 105, 21.1 and 1.6 mg kg^{-1}, respectively in soil of SIFW and 32.2, 209, 67.4 and 4.3 mg kg^{-1}, respectively in soil of SIDWS. The concentration of metals in the soil of SIDWS is at large higher than that in SIFW. The high percentage of Cd and Cr in SIDWS, the author attributed it to waste effluent from small industries (tanneries and batteries) situated in domestic area (Jamali *et al.*, 2007).

(7) Livestock manures

People's demand for meat, eggs and dairy raised extremely over the past decades, due to their living standard rising continuously. Heavy metals are presented in livestock fodders as additives for health and beneficial reasons. Take As for example, it had been used as feed additive for pig and poultry diseases control and growth improvement (Li and Chen, 2005). Unfortunately, As was still in use in some countries such as the USA and China, although it as an animal feed additive had been prohibited in Europe (Li and Chen, 2005). According many reports, the concentrations of heavy metals in poultry manures correspondingly increased with the usage of the feed additives. Livestock manures accounted for approximately 55%, 69% and 51% of the total Cd, Cu and Zn inputs, respectively. Among the metals concerned, Cd was a top priority in agricultural soils in China, with an average input rate of 0.004 mg/kg/yr in the plough layer (0-20 cm) (Luo *et al.*, 2009).

3. Soil pollution control and remediation

Conventional approaches employed for control and remediation of metals from contaminated sites include: (1) land filling, the excavation transport and deposition of contaminated soils in a permitted hazardous waste land; (2) fixation, the chemical processing of soil to immobilize the metals, usually followed by treatment of the soil surface to eliminate penetration by water, and (3) leaching, using acid solutions as proprietary leaching agent to leach metals from soil followed by the return of clean soil residue to site (Krishnamuthy, 2000). Conventional methods used for metal detoxification produce large quantities of toxic products and are cost effective. The advent of bioremediation technology has provided an alternative to conventional methods for remediating the metal-polluted soils (Khan *et al.*, 2009).

Systematic remediation technologies for contaminated soil have been developed, which included bioremediation, physical/chemical remediation and integrated remediation. Six research and development trends in soil remediation are summarized as follows: green and environmentally-friendly bioremediation, combined and hybride remediation, *in situ* remediation, environmentally functional material based remediation, equipment based site remediation, remediation decision supporting system and post-remediation assessment (Luo, 2009).

Phytoremediation is another emerging low-cost *in situ* technology employed to remove pollutants from the contaminated soils. Much work in metal phytoremediaton based on laboratory, glasshouse and field experiments has been carried out in China during last decade. The efficiency of phytoremediation can be enhanced by the judicious and careful application of appropriate heavy-metal tolerant, plant growth promoting rhizobacteria including sysbiotic nitrogen-fixing organisms (Khan *et al.*, 2009). Vegetables, especially mint, from SIDWS (soil amended with and irrigated with waste water) contained high levels of Zn, Cd and Pb than vegetables grown in the same site, suggesting that the cultivation of leafy vegetables should be avoided. (Jamali *et al.*, 2007). Mani *et al.* (2007) investigated the interaction between Cd and Ca, Zn and organic matter for Cd-phytoremediation in sunflower. The results suggested that phytoremediation of Cd contaminated soil through soil-plant-rhizospheric processes. The *Bacillus sphaericus* could be tolerant to 800 mg L^{-1} Cr (VI) and reduced > 80% during growth (Pal and Paul, 2004). A study revealed the relationship between adsorption of Cd by soil and the property of soil, and the influence on the uptake by plant roots. The results indicated that the adsorption capacity of the soils for Cd increased with the increase in the pH or alkalinity of the soil. However, the adsorption rate decreased with the increased in pH. The results also indicated that Cd adsorption capacity of tropical vertisols was higher than those of temperate vertisols (Ramachandran and Dsouza, 1999). Adhikari and Singh (2008) studied the effect of city compost, lime, gypsum, and phosphate on cadmium mobility by columns. All the treatments, lime application reduced the movement of Cd from surface soil to lower depth of soil to a large extent. And combined application of lime and city compost reduced the movement of Cd in soil profile. The results show that the high soil pH may reduce the mobility of Cd and the organic matter control the sorption of Cd in soil.

It is imperative to develop wide-use, safe, and cost-effective *in situ* bioremediation and physical/chemical stabilization technologies for moderately or slightly contaminated farmland, to develop safe, land reusable, site-specific physical/chemical and engineering remediation technologies for heavily polluted industrial site, and to develop phytostabilisation and eco-engineering remediation technologies for control of soil erosion and pollutants diffusion in mined areas. Besides, it also needs to develop guidelines, standards and policies for management remediation of contaminated soil (Luo, 2009). The Asian countries should take more efforts in promoting international exchanges and regional cooperation in soil environmental protection and in enhancing capacities of the management and technologies innovation.

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