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Review of the Current Situation of Cd Contamination in Agricultural Field in the Mae Sot District, Tak Province, Northwestern Thailand

Pensiri Akkajit

Faculty of Technology and Environment, Prince of Songkla University Phuket Campus
80 Kathu, Phuket 83120, Thailand
* Corresponding author: Email: pensiri.a@phuket.psu.ac.th; Phone: (+66)-7627-6488

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Abstract

This paper reviews contamination status, sources and remediation of Cd in agricultural land in Mae Sot District, Tak Province, Northwestern Thailand. The Cd contamination became an environmental issue because mining of a Zn deposit area located uphill of the Mae Tao creeks caused movement of Cd and Zn along the creek to contaminate agricultural soils downstream. Blood Cd levels of residents in this contaminated area were found to exceed the national average of 0.5 μg g^{-1}creatinine. The main route of Cd exposure in this area is the consumption of rice locally grown in this Cd contaminated area. Remediation of Cd-contaminated soils is therefore necessary to reduce public health risks. Remediation technologies considered include chemical remediation and phytoremediation.

Keywords: Cd; contamination; Mae Sot; mining; phytoremediation; soil; Tak province; Zn

Introduction

Heavy metal contamination caused by mining and ore processing is of major concern among the range of environmental impacts associated with Thailand's economic development [1]. For the past 30 years, zinc ore (Zn) has been exploited from the Padaeng deposit in Mae Sot District, Tak Province, Northwestern Thailand [2]. Two Zn mines were operated; the first is no longer active, while the second is owned and operated by the Padaeng Industry Public Company Limited. It is the largest Zn mine in Southeast Asia [3], with an overall production capacity of 110,000 metric tons of Zn metal per year [4].

The mining activities generated a large amount of waste and tailings, resulting in heavy metal contamination of the soils. Cadmium (Cd) contamination in agricultural soils and rice in the vicinity of the mine was first reported in 1998

(Figure 1). Since 2003 it was found that paddy fields receiving irrigation from the Mae Tao and Mae Ku creeks passing through the Zn deposit area contained elevated Cd and Zn levels [5]. It was reported that 1,600 m^2 of paddy fields at the Mae Sot District were contaminated with Cd and Zn [3].

The objective of this study was to assess the contamination status of Cd in soils, sediments and rice plants in the vicinity, and propose technological options for remediation, including chemical remediation and phytoremediation.

Cadmium contamination in an environment

Cd is a particularly hazardous heavy metal because it can be accumulated by plants to levels toxic to humans and animals when consumed even in small amounts [6, 7]. The transfer of Cd to agricultural areas in the vicinity of the mine therefore poses a major human health risk and also impacts on the environment [7, 8].

1) Soil and sediment

Several studies have been conducted to determine Cd levels in soils and identify the origin of Cd in the vicinity of the mine [9, 10, 11]. Soil total Cd and Zn in Thailand ranges from 0.01 to 1.3 mg Cd kg^{-1} [12] and 5 to 158 mg Zn kg^{-1} [13] with a mean value of 0.03 mg Cd kg^{-1} and 45 mg Zn kg^{-1}, respectively [12, 13]. However, soil samples from agricultural areas around the Pha Te village, the Mae Sot District, have total soil Cd and Zn concentrations ranging from 0.63 to 30.4 mg Cd kg^{-1} and 14.4 to 594 mg Zn kg^{-1}, respectively. The upper-paddy soils that receive irrigation water through a canal from the Mae Tao creek and that flow into the lower-paddy soils showed high Cd and Zn concentrations (5.93 to 30.4 mg Cd kg^{-1} and 286 to 594 mg Zn kg^{-1}, respectively) [10]. Mae Tao creek originates in the mountains of Northwestern Thailand and is directly influenced by mining activities. The Mae Tao creek passes through the mine area, Pha Te and Mae Tao Mai Villages, then Mae Sot city, in turn [13]. Soil samples from the Mae Tao creek were found to have low Cd levels upstream (8.45 mg Cd $kg^{-1)}$, increasing to 22.5 mg Cd kg^{-1} at Mae Tao Mai Village. Mae Ku creek, on the other side of the mountain with Zn mining, showed high Cd levels (7.55 to 34.95 mg Cd kg^{-1}). The Mae Tao Ngae Sai and Nong Khiao creeks in the northeastern and southwestern highlands of the mine area had Cd levels of 3.05 mg Cd kg^{-1} and 1.1 mg Cd kg^{-1}, respectively [14]. The data indicate that these soils are contaminated with Cd and Zn, and the source of contaminant is located upstream from the Mae Tao creek [14]. Figure 2 illustrates the location of the creeks in relation to the mine.

Cadmium concentrations in sediments in Mae Sot District have been studied extensively, and have been found to exceed the Thai standard for Soil Quality for Habitats and Agriculture of 37 mg Cd kg^{-1} [15]. The highest concentrations of Cd and Zn (73.1 mg Cd kg^{-1} and 1,330 mg Zn kg^{-1}, respectively) were detected in the creek sediment collected from Mae Tao creek [10]. Thailand's Pollution Control Department [16] reported Cd concentrations in sediments along Mae Tao creek and in the Zn mine area ranging from 44 to 63 mg Cd kg^{-1} and 82 to 326 mg Cd kg^{-1}, respectively. During April 2011 and February 2012, Cd concentrations in sediments upstream and downstream of Mae Tao creek ranged between 0.84 to 7.86 mg Cd kg^{-1}, exceeding the European maximum level of 3.0 mg Cd kg^{-1} for agricultural soils [17]. The highest total Zn and Cd concentrations in Mae Tao creek were found in stream sediments (1,231 mg Zn $kg^{-1}$and 37.11 mg Cd kg^{-1}) and suspended solids (7,767 mg Zn $kg^{-1}$and 18.27 mg Cd kg^{-1}). In the Mae Ku creek stream sediments contained 316.55 mg Zn kg^{-1} and 7.99 mg Cd kg^{-1}, whilst suspended solids contained 7,723 mg Zn kg^{-1} and 7.75 mg Cd kg^{-1}, respectively) [18].

Since soil is an extremely heterogeneous system, the chemistry of metals has been shown to vary from place to place. Metals exist as a variety of chemical species and exhibit different behavior in terms of chemical interaction, mobility, biological availability and potential toxicity. Bioavailability can be defined as the fraction of the total metal that is readily available for uptake by living organisms. It is therefore important to understand the processes of distribution and transformation of metals under the prevailing soil environments, in order to understand the migration and movement of Cd into uncontaminated soils. Assessment of the changes in the Cd forms and measurement of soil parameters would allow more insight into mechanisms that might be responsible for Cd immobilization and/or metal movement. The three-step BCR sequential extraction proposed by the Standards, Measurements and Testing Programme of the European Union [19] has been used extensively to determine the bioavailability of metal in this particular area. The extraction procedures are useful under defined conditions for predicting metal transformation with respect to their extraction capacity. Sequential extraction techniques estimate the amounts of metals in various solid fractions which can be operationally categorized as follows: easily soluble (exchangeable-BCR1), Fe-Mn oxide bound (reducible-BCR2), organic (oxidizable-BCR3), and organic and silicate bound (residual-BCR4). This procedure has been standardized and applied to a variety of matrices including sediments, soils; sewage sludge, mining wastes, with some modifications [20]. Various studies have been conducted to determine element behavior in order to estimate the risk associated with Cd movement [10, 11, 21].

In 2007, Cd in soils from the Mae Tao and Mae Ku sub-catchments showed the highest mobility with the highest content in the first (exchangeable-BCR1) (25 to 30%) and the second (reducible-BCR2) fractions of the three-step BCR sequential extraction [21]. The major proportion of Cd and Zn in soils collected from the Pha Te village, the Mae Sot District, was dominantly associated with the exchangeable fraction (40 to 70% of total Cd and 37 to 46% of total Zn, respectively) [10]. In the stream sediments from Mae Tao creek, Cd is distributed mostly in extractable forms (BCR1 and BCR2); on the other hand, Cd from Mae Ku creek are dominated by the less extractable forms of BCR2 and BCR4 [20]. Significantly, 70 to 90% of Cd in the paddy fields was found to be present in the exchangeable fraction (BCR1) [11]. The exchangeable fraction is only weakly absorbed, is easily solubilized and thus is readily bioavailable for plant uptake. This poses significant risks to the ecosystem and has significant potential to affect the environment via transfer of Cd through the food chain.

Figure 1 Location of the Zn mine at the Mae Sot District, Tak Province, Thailand

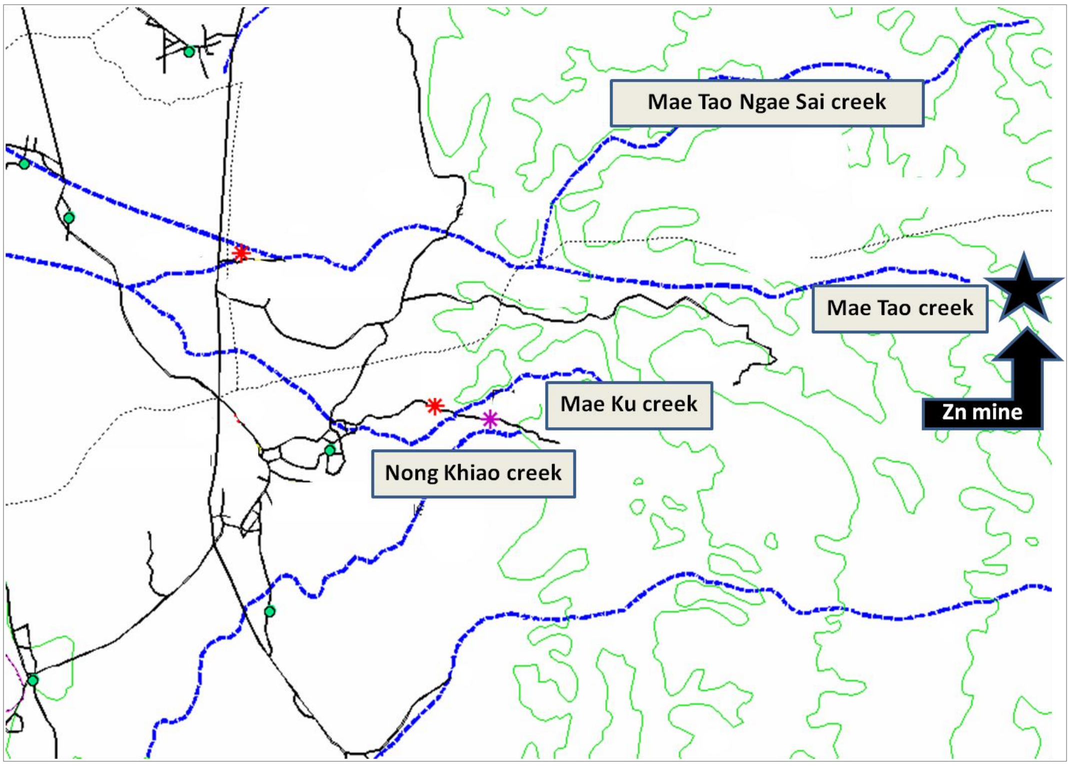


Figure 2 The location of the Mae Tao, Mae Tao Ngae Sai, Mae Ku, and Mae Nong Khiao creeks. Source: modified from Unhalekhaka and Kositanont, 2008

2) Water

The quality of surface water around the Mae Sot District was investigated in addition to analyses of soils and sediments. Mae Tao creek is located between latitude 16º 39' 90" to 15º 40' 28" N and longitude 098º 36' 66" to 098º 42' 26" E in Mae Sot District, Tak Province, in a mountainous area on the border between Thailand and Myanmar [22]. Records of monthly mean rainfall (mm) for a 10-year period (2000-2009) in this area showed that the highest monthly rainfall occurred during June to August; with the highest monthly rainfall of 370 mm recorded in July [13]. Cd and Zn concentrations in surface water from Mae Tao creek (which passed through the Zn mining area), were higher during the rainy season (0.028 to 0.032 mg Cd L^{-1} and 0.049 to 0.378 mg Zn L^{-1}, respectively) than in the dry season (0.005 to 0.006 mg Cd L^{-1} and 0.049 to 0.091 mg Zn L^{-1}, respectively) [23]. A temporal distribution of Cd concentrations in surface water between the two seasons was clearly observed [13, 24], suggesting that the paddy fields receive Cd mainly via suspended sediment transport from Mae Tao creek. However, turbidity and suspended solid levels were found to vary according to the season. Downstream of Mae Tao creek, the accumulated bed sediment transport during the wet season was 133,200 kg, with peak bed sediment transport occurring immediately after heavy storms [25]. Information on sediment transport will help in determining water quality and contamination in the area as well as the impact on water utilization and the aquatic ecosystem.

3) Crops

Rice (*Oryza sativa L.*) is Thailand's main staple food crop [26, 27] accounting for 66% of the country's total agricultural land [27]. The Codex Committee on Food Additives and Contaminants (CCFAC) has proposed a maximum permissible level for Cd in rice grains of 0.2 mg Cd kg^{-1} [28]. Thai rice variety Khao Dawk

Mali 105 or Jasmine rice, grown in the contaminated paddy fields in Mae Sot District, showed the highest value of Cd (82%) as compared to other metals, with manganese (Mn) and Zn also accumulating in above-ground parts (stems) of the rice plants, at levels of up to 34 and 29%, respectively [29]. In 2008, Cd levels in home-consumed rice grains collected from the Cd-polluted paddy fields around Pha Te village, ranged from 0.12 to 1.27 mg Cd kg^{-1}, whilst soybean seeds (*Glycine max L.*) contained Cd at levels ranging from 0.07 to 0.80 mg Cd kg^{-1} [13]. Research undertaken over the past 5 years has identified serious health risks associated with prolonged consumption of rice grown in this Cd contaminated area [30, 31]. Several hundred villagers in these areas were found to have high levels of Cd in their blood, with an average of 8.2 µg g^{-1} creatinine. This was far in excess of the national average of 0.5 µg g^{-1} creatinine, and was attributed to consumption of contaminated rice and other local crops [31] as the main route of exposure.

4) Human impact

Regardless of the route of exposure, Cd is retained and accumulated mainly in the human kidney [32, 13]. Therefore, urinary excretion is a good indicator of excessive Cd exposure and body burden [32, 33]. The World Health Organization (WHO) has established a urinary Cd level of 5.24 µg g^{-1}creatinine as a threshold to protect against kidney damage, whereas the European Food Safety Authority (EFSA) has set a urinary Cd threshold of 1 µg g^{-1}creatinine [34]. A population survey in 2004 for Cd exposure using urinary Cd measurement among exposed residents aged 15 years and older in contaminated areas of Mae Sot District showed that individuals who consumed rice (*Oryza sativa L.*) grown locally in the contaminated areas had higher urinary Cd than those consuming rice grown in other areas. Of the 7,697 persons surveyed, 45.6% had urinary cadmium levels < 2 µg g^{-1}creatinine, 4.9% were between 5 to 10 µg g^{-1}creatinine and 2.3% had Cd concentrations > 10 µg g^{-1}creatinine [9]. Renal dysfunction among the exposed population was evident in an increase in incidences of tubular and glomerular dysfunctions [8]. In addition, a 2013 survey of primary school children in the Cd-contaminated villages who consumed locally grown rice also showed high urinary Cd of ≥ 1 µg g^{-1} creatinine. Such high urinary Cd levels are likely to impact adversely on renal function in these children [35]. Another survey of the population aged above 40 in Mae Sot District (230 men and 370 women) showed the Benchmark Dose Lower Confidence Limit (BMDLs) of urinary Cd and blood Cd in the range of 4.4 to 8.1 µg g^{-1} creatinine and 4.4 to 6.2 µg L^{-1} creatinine, respectively. This exposure level corresponds to a specific increase in the probability of an adverse response of Cd exposure, compared with the response at zero background exposure. Moreover, women are much more likely to get osteoporosis (bones becoming weak and brittle) and suffer more often than men. Cd-exposed persons aged 40 and older showed a rate of osteoporosis in women (21.5%) significantly higher than that for men (14.7%). Increasing urinary Cd levels appeared to correlate with reduced bone density in women and also renal dysfunction in this female population [30]. This suggested that the inhabitants at the Mae Sot District were at risk of adverse renal effects induced by Cd exposure [36].

Remediation methods

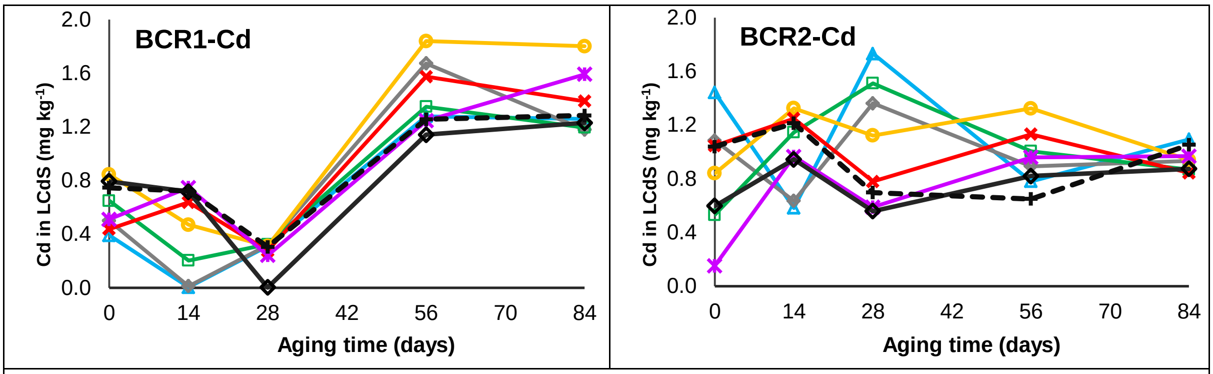
1) Chemical technologies

A range of approaches have been tested in these contaminated areas to identify appropriate technologies for reducing Cd toxicity. The performance of three fertilizers including triple superphosphate (TSP), diammonium phosphate (DAP), and phosphate rock (PR) as stabilizing agents was studied, and was shown to be effective in this locality [37]. The leachable concentrations of Cd in PR, DAP and TSP treated soils were reduced from 306 mg Cd kg^{-1} in the control to 140, 34, and 12 mg Cd kg^{-1}, respectively, corresponding with an increase in the more stable forms of Cd and the phosphate dose that based on the molar ratio of PO_4 of 1:2, 2:3, 1:1, and 2:1, respectively [37]. Moreover, TSP proved an effective stabilizing agent to reduce Cd leachability in the locality's three different soil types (loamy-sand, clay loam, and clay soils) with a 94% reduction of Cd leachability when TSP was applied at a ratio of 2:1 for 30 days. The partitioning of Cd was changed from the potentially available phase to the more stable phase in all TSP treatments [38]. The effects of organic fertilizer on phytoavailability and distribution of Cd and Zn in rice plants were also examined and it was found that organic fertilizer application is likely to reduce uptake of both Cd and Zn by rice, by transforming metals from the soluble to the more stable phase [39].

The efficiency of soil amendments in reducing metal uptake by plants has been extensively studied, with the aim to convert and/or transform metals from the bioavailable pool to the insoluble fraction [2, 40, 41]. Assessment of Cd and Zn uptake by *Ocimum gratissimum* or African basil was studied with the addition of hydroxyapatite (HA) and cow manure. African basil showed a decrease in leaf Cd concentration from 1.5 to 0.3 mg Cd kg^{-1} and a decrease in Zn concentration from 69.3 to 34 mg Zn kg^{-1} by addition of cow manure and HA, respectively [40]. Addition of sugarcane waste-products (boiler ash, filter cake and vinasse) as a soil amendment at an application rate of 3% (w/w) stimulated sugarcane growth and increased plant biomass production (6 and 3-fold higher for the above-ground parts and roots, respectively) and resulted in a decrease of up to 54.5% in concentrations of the most bioavailable Cd fractions (exchangeable-BCR1 and reducible-BCR2 fractions) in treated soils [41]. Additionally, a separate study of fractionation of Cd and Zn in Cd-contaminated soils amended by sugarcane waste-products confirmed the observed reduction in exchangeable fraction (BCR1) of Cd in the treated soils during the first 28 days of the experiment [21]. The rate of metal transformation differed according to the ratio of incorporation of soil amendments (Figure 3) [21]. The efficacy of biochar in reducing bioavailability of Cd and uptake by *Jatropha curcas L.* plants was also tested [42]. Addition of biochar resulted in a significant increase in the soil's growth potential and a significant decrease in Cd uptake ($p<0.05$).

2) Phyoremediation and alternative crops

Phytoremediation uses plants to remove, accumulate, contain and/or stabilize heavy metals in contaminated soils, sludge, sediments, surface water and groundwater, and has been widely tested in Mae Sot Distict to determine site specific conditions to achieve the desired goal of reducing the risk associated with the Cd contamination. The efficacy of phytoremediation is species-dependent; a hyperaccumulator is a plant that is capable of accumulating high amounts of heavy metals in above-ground parts without suffering phytotoxic effects [8]. *Gynura pseudochina* (L.), *Chromolaena odorata* (L.), *Crassocephalum crepidioides* (Benth.), and *Conyza sumatrensis* (Retz.) are commonly found around the Zn mine area in Mae Sot District and are classified as hyperacumulating plants under greenhouse conditions [3]. These four species were also tested under field conditions in contaminated soils in Mae Sot District and other areas with similar site characteristics. In addition, other species growing naturally in the area were also tested to find Zn phytoaccumulative species [43]. The results indicated that vetiver grass (*Vetiveria zizanioides Nash)*, *Cyperus rotundus Linn*, *Kaempferia* sp., *Gynura pseudochina* (L.) DC., and *Cyanotis tuberosa* showed potential for remediation of Zn-contaminated soils, with the highest Zn accumulation of 73 g kg^{-1} ash found in the roots of *Cyperus rotundus Linn* [43].



Where
L1, L2, L3, L4, L5, L6, and L7 = sugarcane waste-product amended soils;
LC = the control (no amendment)

Figure 3 Distribution of Cd between exchangeable (BCR1) and reducible (BCR2) due to sugarcane waste application

Due to its high tolerance and adaptation to the local environment, vetiver grass, with its long and dense root system, has been extensively tested for its effectiveness in remediating contaminated land in Mae Sot District [44,45, 46]. Vetiver can absorb heavy metals and reduce soil concentrations, thus mitigating transport due to leaching and runoff [44, 45]. There are two species of vetiver in Thailand, namely *Chrysopogon nemoralis* (Balansa) Holttum and *Chrysopogon zizanioides* (L.) Roberty, each with a number of ecotypes [44]. Two ecotypes of *Chrysopogon nemoralis* (Nakhon Sawan and Prachuap Khiri Khan) and *Chrysopogon zizanioides* (Kamphaeng Phet 2 and Surat Thani) were studied and grown in the Zn mine area at the Padaeng Industry Public Company Limited in 2006 to determine growth performance under field conditions in the contaminated area [44]. These four ecotypes of vetiver are commonly found in Thailand and grow naturally in a wide range of environments [46]. Two *Chrysopogon zizanioides* ecotypes showed good growth performance with high biomass production, indicating its potential for phytoremediation in this area [44, 45].

In addition to vetiver, *Chromolaena odorata* or Siam weed has also been widely studied. *Chromolaena odorata* grows well in highly contaminated soils, with high biomass production and no visual symptoms of phytotoxicity [47]. Cd and Zn accumulated mainly in the shoots (14.1 mg Cd kg^{-1} and 278.0 mg Zn kg^{-1}), indicating its capability to translocate heavy metals from soil to plant biomass. In comparative studies, both species showed high levels of metal accumulation in the roots, with metal concentrations in plant tissue increasing with time [48].

Recently, water hyacinth (*Eichhornia crassipes)* was also found to be effective in removing heavy metals in Cd-contaminated soils, with the addition of chelating agents (EDTA and DTPA). Again, soil Cd levels decreased over time from the begining until 100 days after planting [49].

Conclusions

Cd contamination in Mae Sot District has arisen from downstream transport via the two Mae Tao creeks passing through a Zn deposit and mine area. Cd and Zn are transported along the creek to surrounding agricultural soils. Moreover, Cd contamination in paddy fields was found to be dominated by the exchangeable fraction

(BCR1), whose high bioavailability exacerbates health, environment and ecosystem risks through bioaccumulation along the food chain.

Urinary Cd levels of the local residents are much higher than the national average of 0.5 μg g^{-1} creatinine due to consumption of contaminated rice and other locally grown crops. Since prolonged consumption carries risks of adverse renal and other health impacts, the Thai government has prohibited rice cultivation in this area since 2004 in an effort to minimize further exposure. The multidisciplinary clean-up actions for site remediation used in this area included the following: encourage planting of non-edible crops; promote phytoremediation; remove contaminated soils; and stabilize soil Cd using chemical treatments. Understanding the factors controlling Cd and Zn movement in the paddy and agricultural soils is crucial to contain contamination and mitigate soil dispersion, especially during the rainy season in order to prevent Cd accumulation via the food chain. A multidisciplinary approach is needed to address the challenges of environmental health risk management in this locality.

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