NEW ASPECTS OF COLLABORATIVE RESEARCH ON SOIL POLLUTION, FOOD SAFETY AND SOIL REMEDIATION TECHNIQUES IN ASIA

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ABSTRACT

Soil pollution and food safety issues are major challenges for discussion in Asia resulting from previous industrial development without suitable regulations. Over the years, a nationwide comprehensive survey to understand the distribution of heavy metal contaminated soil was conducted in many Asian countries. Included in this survey are descriptions of Taiwan’s experiences and related databases. Rice is the staple food in many Asian countries. Many paddy soils were contaminated by illegal discharges of wastewater from local factories. Results from field studies in Taiwan showed that rice variety and soil Cd concentration are major factors affecting edible rice safety. The Indica type rice absorbed more Cd from contaminated soil than Japonica type. The most contaminated were brown rice and polished rice when the soil Cd concentration extracted by HCl was higher than 2 mg/kg. No relationship could be found between soil Cd and brown rice Cd. The Cu and Zn concentrations in brown rice were in a fixed range under a wide range of soil Cu and Zn. These relationships coincide the plateau theory. The concentration of Cu and Zn in rice is not proportional to its concentration in the soil. When the total Cr and Ni concentration of rural soils reached 250 and 600 mg/kg, rice yield will decrease. But the Cr and Ni concentration in polished rice were less than 4 and 14 mg/kg, respectively. In paddy soil and rice uptake system, Pb solubility and mobility is very low, however rice yield will decrease by 20% as soil total Pb approached 2,000 mg/kg. When the total As concentration of rural soil reached 60 mg/kg, rice production will be significantly reduced. To explain the relationship between heavy metal concentration in soil and plants, six hypotheses (i.e., single-metal abrupt toxicity threshold, soil-plant barrier, clean sludge concept, uptake plateau concept, aging effect, and absence of evidence) were proposed from previous research findings. The regulations of heavy metal in contaminated soils, the main reasons of the development of legislations, and the strategies for monitoring and remedial actions in USA, UK, Netherlands, Germany, Japan, China, and Taiwan were also compared in this article. Finally, concerning the high toxicity and mobility of Cd, its transmission in food chain and human intake of Cd from all pathways were also analyzed.

Key words: Agro-environmental quality, soil quality, heavy metal contamination, soil remediation, regulations of heavy metals, health risk assessment system, human health, food safety
INTRODUCTION

Main Concerns in Soil Pollution

In Eastern Asia, issues concerning agro-environmental quality, soil quality, food security, heavy metal contamination and remediation, and human health are more popularly discussed in recent years. On the other hand due to rice market liberalization, land use conversion from paddy soils to non-waterlogged cropping system, posed some problems on soil properties and soil system. Some irrigation water for rice production in Asia has been contaminated by with waste water from illegal discharges of industrial parks or livestock. The contamination affected the paddy soil qualities and food safety due to presence of heavy metals.

More than 90% of the potential contamination areas of rural soils in Taiwan were from wastewater (Chen 1991). The main concern of the Taiwan EPA, Council of Agriculture, and Department of Health is the Cd pollution in brown rice since 1988. Also it was found out that more than 300 ha of rural soils in Central Taiwan were contaminated by multiple elements (Cu, Cr, Ni, Pb, and Zn). The wastewater was discharged from illegal plants located in central Taiwan (Taiwan EPA website, http://sgw.epa.gov.tw/public/en/index.htm).

The Environment Agency of Japan entrusted the National Resources Information Center (NRIC) to conduct a soil survey of heavy metals (Cu, Zn, Cd, Pb, Cr, Mn, Ni, and As) in its arable land and forest soil from 1978-1982 (Japanese Soil Association 1984). Soil samples were collected from 687 sites. The results obtained from this survey could be use to determine the natural abundance level of heavy metals in cultivated soils of Japan.

In Korea an extensive monitoring project of the distribution of heavy metals in soil and crops was conducted a few years ago. It was concluded that the concentrations of Cd, Cu, Pb and Zn in paddy soils without an evident anthropogenic source of contaminants were mostly below the threshold levels of arable land pollution designated in the Soil Environment Conservation Law in Korea (Yang et al. 2007).

A National Soil Pollution Survey Program will be conducted in China for two years (2006-2008) It will be conducted by the State Environmental Protection Administration of China (SEPA) (http://www.zhb.gov.cn/natu/yjsp/qgtrzdc/index.htm, In Chinese). The main objectives of this project are: 1) to understand the soil environmental quality by systematic and corrective methods; 2) to investigate the types, degree and main reasons of soil pollution; 3) to assess the health risk of the contamination sites; (4) to select the best soil remediation techniques to be applied; and 5) to establish the soil pollution protection and remediation act. Based on the draft report within the last decade, the potential contaminated soil in China is about 10% of total rural soils.

Soil-crop Inventory Database of Heavy Metals in Asian Countries

In Eastern Asia such as China, Japan, Korea and Taiwan, a complete database of soil-crop inventory on heavy metals has been developed in the last two decades. From this national monitoring and study programs, they have developed the relationships between concentration of heavy metals in rice, other crops and soils to evaluate the food safety.

Soil inventory of metals. Kato et al. (2000) investigated the elemental compositions of alluvial soils in Japan. They collected soil samples from 366 different sites and determined 19 elements (Al, Ba, Ca, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Sr, Ti, V, Zn, and Si). Yamazaki et al. (2001), and the trace and ultra-trace elements of 540 soil samples selected from 78 different soil profiles. They also estimated the natural abundance levels of 57 elements in these soils.

In Korea, major sources of metal pollution in paddy soils were related with mining activities such as tailings erosion to paddy fields and irrigation use of mine drainages. Soil contamination with Cd and Cu exhibited a detrimental effect on soil nutrient availability. Concentrations of Cd and Pb in rice grains in these areas often exceeded the CODEX safety guideline of 0.2 mg/kg. The hazard quotients for metals through rice consumption were significantly higher than other exposure pathways. They need to develop the protocols for pollution monitoring, risk assessment and remediation techniques for paddy soil environment (Yang et al. 2007).

In Taiwan, heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) contaminations have been
found in the paddy soils due to illegal wastewater discharged from industrial plants (i.e., chemical, electroplate, pigment) and livestock (i.e., swine). Since 1982, the Environmental Protection Administration of Taiwan (Taiwan EPA 2007) design four stages of soil survey project on contaminated soils. The database of heavy metals survey, ensure a better understanding of its pollution status in rural soils and different crops. The major results of these stages are illustrated as the following (Taiwan EPA 2007):

- **Stage 1** (1983-1987): The total survey area was about 1,160,000. Each representative survey unit is 1,600 ha. The final report was published by Taiwan EPA in 1987.
- **Stage 2** (1987-1991): Stage 2 was conducted for 300,000 ha of rural soils selected from stage 1 which have higher concentrations of metals and each representative survey unit was 100 or 25 ha. The draft regulations of heavy metals were set up as: As 60, Cr 16, Cd 10, Cu 100, Pb 120, Hg 20, Ni 100, Zn 80 mg/kg. However, the metals in soils were extracted by 0.1M HCl, except As and Hg which were extracted by aqua regia.
- **Stage 3** (1992-2000): The survey lands were selected according to the results of stage 2 which have relatively high potential contamination of metals in the rural soil. The total survey area was about 50,000 ha and each representative survey unit was 1 ha.
- **Stage 4** (2000 to now): This stage was conducted following the Soil and Groundwater Pollution Remediation Act (SGWPR Act) announced by Taiwan EPA in 2000. The contaminated control sites and remedial sites were announced based on the regulations of pollutants listed in the SGWPR Act (As 60, Cr 250, Cd 5, Cu 200, Pb 500, Hg 5, Ni 200, and Zn 600 mg/kg). This regulation is based on the total contents of heavy metals in the soils digested by aqua regia.

The database of heavy metal content in rural soils of Taiwan was first conducted from 1992 to 1997. The upper level of background total concentration of heavy metals in surface soil (0-15cm depth) were estimated as (mg/kg soil): As 18, Cd 2, Cr 50, Cu 35, Pb 50, Ni 50, and Zn 120.

Based on the results of 319 ha rural soil survey in 2002, Taiwan EPA reported that the areas of contaminated rural soil higher than the regulations were; 1) Ni-contaminated 159 ha, 2) Cu-contaminated 148 ha, 3) Cr-contaminated 127 ha, 4) Zn-contaminated 113 ha, 5) Cd-contaminated 17 ha, 6) Pb-contaminated 4 ha, and 7) Hg-contaminated 0.3 ha. The total contaminated rural soil was about 251 ha.

### Crop inventory of metals

In Taiwan, Lin (1991) made a detailed survey on the distribution of heavy metals (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in different crops of Taiwan. The results are listed as follows:

- The mean values of heavy metal concentration in brown rice (n=431, mg/kg dry) are As 0.17, Cd 0.07, Cr 0.16, Cu 2.48, Hg 0.001, Ni 0.54, Pb 0.43, and Zn 39.2.
- The mean values of heavy metal concentration in leaf-vegetables (n=144, mg/kg dry) are As 0.12, Cd 0.24, Cr 0.02, Cu 4.64, Hg 0.04, Ni 2.14, Pb 3.69, and Zn 38.1.
- The mean values of heavy metal concentration in root-vegetables (n=112, mg/kg dry) are As 0.05, Cd 0.21, Cr 0.03, Cu 3.00, Hg 0.03, Ni 1.63, Pb 2.58, and Zn 27.4.
- The mean values of heavy metals of fruit (n=90, mg/kg dry) are As 0.05, Cd 0.11, Cr 0.26, Cu 3.52, Hg 0.02, Ni 0.95, Pb 2.11, and Zn 27.7.

In Japan, many case studies and databases were established in many river basins where heavy metals were discharged from different mines in the last two to three decades (Chino, 1973; Morishita, 1975, 1981, 1982; Kitagishi et al., 1976; Yoshikawa et al., 1977; Kitagishi and Yamane, 1981; Asami, 1982).
4

The distribution of heavy metals in the different parts of brown rice was investigated and the differences of heavy metal concentration between different species of rice were also shown.

Contamination of heavy metals in paddy soils of Taiwan. According to the database of rural soils with potential contamination conducted by Taiwan EPA in 2002, more than 300 ha of rural soils were regarded as seriously contaminated sites and at least about 3,000 ha were regarded as slightly or moderately contaminated sites in Taiwan. Hence crop quality and food safety issues have become increasing concerns after 2000. The regulation criteria of total contents of heavy metals in the SGWPR Act are listed as follows (mg/kg): Cd 5.0, Cr 250, Cu 200, Ni 200, Pb 500, and Zn 600, based on total digestion method by aqua regia. The maximum tolerance of heavy metals in rice were: Cd 0.5 mg/kg dry weight (DW), Hg 0.05 mg/kg DW, and Pb 0.2 mg/kg DW (Taiwan DOH, 2007). The contaminated area by different metals exceeding

Fig. 1. The relative percentage and accumulative percentage of the concentration of a) Cd, b) Cu, and c) Zn in the surface soil (0-15 cm depth) of database collected from the whole soil survey project from 1992 to 1997 (Chen et al. 1998)
the regulation criteria of pollutants in SGWPR Act of paddy soils for Ni, Cu, Cr, Zn, Cd, Pb and Hg were 159, 148, 127, 113, 17, 4 and 0.3 ha respectively. Most of those (184 ha) were in Changhua county located in central Taiwan. They were mainly contaminated with Cu, Zn, Ni, and Cr.

According to the AGWPR Act, the crops grown in the contaminated soils should be collected and destroyed by the governmental agency to avoid human health risk through food chain. Although the heavy metal contents of the contaminated paddy soils were higher than the recommended, the effects of this total concentration of the metals on the crop quality and human health require further examinations.

**RELATIONSHIP BETWEEN HEAVY METALS CONCENTRATION IN RICE AND IN SOILS**

These relationships were approaches to predict the distribution of heavy metals in rice and crops by total or bioavailable concentration of heavy metals in soils, specifically for Cd, Cu, Zn, and Pb within the last decade. Many bioavailable extraction techniques (specifically for Cd) were also tested and evaluated for the prediction of heavy metal distribution in rice. Plateau theory, time bomb theory, and other concepts were approaches to predict the heavy metals distribution in rice, vegetables and other grains after irrigation with wastewater discharge or sewage sludge and livestock compost amendment. There is a need for long term experiments to share this knowledge.

In Asian countries, the major concerns were how to approach and assess the food safety of heavy metals in brown rice and how to examine and revise the regulation of heavy metals in contaminated soils, rice, and vegetables.

**Cadmium (Cd) in Rice in Japan**

Morrishita (1975) reported that the relationship between Cd concentration in soils and rice in four districts of Japan namely Annaka, Kurobe, Bandai and Fuchu located in Junma, Fukusgima Fukushima and Toyama county respectively (Fig. 2). Relationships showed that the empirical critical Cd values in soils correspond to 0.4 or 1.0 mg/kg in unpolished rice grain. Allowable levels in soils could potentially be determined in each district of Japan. Results also indicated that the differences of Cd concentration in brown rice were due to diversity in soil properties (specifically in Fuchu district).

Herawati *et al.* (2000) collected 178 paired soil and brown rice samples from Japan (n=111), China (n=22) and Indonesia (n=45) to compare levels of Cd, Cu, and Zn.
concentrations in these countries. The result showed that Japanese brown rice and soil have the highest levels of Cd and Cu among three countries (Table 1). Considering the different soil types in Japan, the highest soil Cd were in Cambisols, but the highest brown rice Cd were in Fluvisols (Table 2). This implies the influence of soil properties on Cd bioavailability. The correlations between these three metals in soil and in brown rice were not significant. Many studies also found similar results (Suzuki and Iwao 1982; Rivia et al. 1990; and Schumacher et al. 1994). Factors such as soil pH, soil texture, metal absorption and assimilation by plants may affect the relations.

In order to investigate the relationship between soil properties and Cd concentration in wheat grain, Adams et al. (2004) collected and analyzed 162 paired soil and wheat grain samples from commercial farms in Britain during 1998-2000. From two long-term sewage sludge experimental sites in English, eighty-four paired samples were also collected and analyzed. All data were combined and used in multiple regressions. Soil properties (e.g. soil organic matter, contents of Fe and Al oxides, total Zn concentration, etc.) tested in the multiple

Table 1. Cd, Cu, and Zn concentrations in rice and soils of Japan, China, and Indonesia

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*Extracted by HCl (Herawati et al. 2000)

Table 2. Cd, Cu, and Zn concentrations in soil and rice in different soil types of Japan, China, and Indonesia
regressions. Only soil total Cd concentration and pH were found to be significantly (P<0.05) correlated. Resulting in the following equation:

\[
\log \text{grain Cd (mg/kg)} = 0.12 + 0.43 (\pm 0.029) \log \text{soil Cd (mg/kg)} - 0.16 (\pm 0.016) \text{soil pH}; n = 246, R^2 = 0.53, SE = 0.20.\]

This equation is useful in predicting Cd concentration in wheat grain under different soil conditions. Fig. 3 shows the influence of soil pH and soil total Cd on the predicted grain Cd. The predicted grain Cd decreases with soil pH and increases with soil total Cd.

**Heavy Metals in Rice in Taiwan**

*Field studies in 2005 and 2006.* To understand more about the relationship between heavy metals in rice and in soils under different soil properties and rice varieties and ensure the safety of agricultural products, Guo *et al.* (2006; 2007) conducted the field trials in Changhua (2005), Taoyuan and Hsinchu (2006) county. Twelve most widely cultivated rice varieties in Taiwan including japonica, indica, and sticky type were selected as trial crops. The pH of Cu, Zn, Ni and Cr contaminated soil in Changhu and Cd contaminated soil in Taoyuan were around 7.2 and 5.5 respectively. To assess the bioavailability of heavy metals (Cd, Cr, Cu, Zn, Ni, and Pb) in soils, four extractants were used as follows; 1) 0.1M HCl, 2) 0.43M H3NO4, 3)0.05M EDTA and 4) 0.01M CaCl2. The results from Changhua and Taoyuan on Cd, Cu, and Zn concentration in brown rice of Tainung No. 71 (Japonica type), Tainung Sen no. 20 (Indica) and in the soil extracted by HCl were presented in Figs. 4, 5 and 6.

As shown in Fig. 4, no relationship could be found between soil Cd and brown rice Cd in both rice varieties. Regardless of Cd contamination levels, the Indica type absorbed about 2-3 more Cd from soil than the Japonica type. Some studies conducted in Japan as pot or field experiments also showed similar results (Arao and Ae, 2003; Morishita *et al.* 1987). When HCl extractable Cd in soil is higher than 2.5 mg/kg, all Cd concentrations in brown rice of both varieties were higher than 0.5 mg/kg, reaching the regulated limit of edible rice Cd concentration in Taiwan. It is clear that rice variety and soil Cd concentration are major factors affecting edible rice safety.

Although the soil properties and Cu concentration varied in Changhua and Taoyuan, the Cu concentration in brown rice in both varieties ranges between 3 to 8 (Fig. 5). This range is lower than the maximum permissible limit of 10 mg/kg regulated in China (Yan *et al.* 2006). Xu *et al.* in 2006 reported that under the pot experiments, soils amended with CuCl2 · 3H2O to different Cu contamination levels, Cu concentrations in rice grain increased with soil total Cu levels below 150–200 mg/kg, but decreased with soil total Cu levels above 150–200 mg/kg. However, these results showed that the rice uptake of Cu was not affected

![Fig. 3. The influence of soil pH and soil total Cd on the predicted wheat grain Cd. Vertical bars represent 95% confidence intervals. Dotted line represents the current European Union foodstuff regulations on the maximum permissible concentration (MPC) of Cd (0.235 mg/kg DW) from Adams et al. 2004.](image-url)
Fig. 4. The relationship between Cd concentrations in soils extracted by 0.1M HCl and in brown rice of two varieties in two counties of Taiwan (Guo et al. 2006; 2007).

Fig. 5. The relationship between Cu concentrations in soils extracted by 0.1M HCl and brown rice of two varieties in two counties of Taiwan (Guo et al. 2006; 2007).
apparently by soil Cu and rice varieties under the field conditions.

Cultivated in the field with similar concentration range of Zn in soil, the Tainung No. 71 absorbed more Zn in Taoyuan than in Changhua (Fig. 6). The lower soil pH in Taoyuan may increase the Zn solubility in soil and enhance the Zn uptake of rice, whereas this phenomenon was not shown in Tainung Sen No. 20. From this research and other related studies, it is doubtless that factors affecting heavy metal uptake of plants are various and complicated. It seems that no universal methods were existed to predict heavy metal concentration in plants. In this versatile world with complicated soils, there are always some exceptions reported in the literatures. Although some principles are useful guidelines to assess heavy metal bioavailability, the case-specific field trials are best methods to handle the real situations.

**Field Research in 1990s**

**Cadmium.** The regulation levels of total contents of Cd in soils in the world ranged from 1 to 5 mg/kg. The upper limit of background Cd contents of representative rural soil was 3 mg/kg in Taiwan. The study in Taiwan and Japan suggested that the Cd content in brown rice and in soil was not significantly related as shown in Fig. 7. Results were based on the research conducted in Cd contaminated fields in Taiwan (Li et al. 1988; and Chen 1991).

The field survey in central Taiwan (Fig. 8) suggested that most of the contaminated brown rice and polished rice were found in soils where total Cd concentration was higher than 2 mg/kg in soils extracted with 0.1 M HCl (Liu et al. 1998). The database in Taiwan and Japan indicated that the Cd-contaminated rice could be found in areas with different soil properties and soil management regardless if the total content of Cd in soil was less than 4 mg/kg.

**Copper.** The regulation levels of the total content of Cu (Fig. 9) in soils around the world ranged from 100 to 1500 mg/kg. The upper limit of background Cu content of representative rural soil was 35 mg/kg in Taiwan. The Cu concentration in polished rice
grains ranged from 4 to 8 mg/kg and it does not increase together with the increasing Cu concentration in soils (Liu et al. 1998). When the total Cu concentration in soil reaches 320 mg/kg, the rice production will be significantly reduced by 30%. The rice production will have a 50% reduction when the total Cu concentration in rural soil reaches 600 mg/kg; however the Cu concentration in brown rice was less than 17 mg/kg in this contaminated site (Liu et al. 1998). The relationships hereby presented satisfy the plateau theory which states that the Cu concentration in rice is not proportionate to the increasing Cu concentration in soil (Corey et al. 1987; and Chang et al. 1997).

**Zinc.** The regulation levels of the total Zn content in soils all over the world ranged from 200 to 3000 mg/kg. The upper limit of background Zn concentration in representative rural soil was 120 mg/kg in Taiwan. The Zn concentration in polished rice ranged from 20 to 80 mg/kg when the soil’s total Zn ranged from 60 to 960 mg/kg in this contaminated site. If the total Zn concentration in soil reaches 500 mg/kg, the rice production will significantly be reduced by 30% and the Zn concentration of polished rice will range from 50 to 80 mg/
kg. Fig. 10 shows that rice production will decrease by 50% when the total Zn concentration in soil reaches 800 mg/kg, but the question of whether the food was safe or not can be further discussed as the Zn content in polished rice was less than 30 mg/kg (Liu et al. 1998). The relationships satisfy the plateau theory wherein the Zn concentration in rice is not proportionate to the increase of Zn concentration in the soil (Corey et al. 1987; and Chang et al. 1997).

Chromium. The regulation levels of total content of Zn in soils in the world ranged from 100 to 1500 mg/kg. The upper limit of background Cr contents of representative rural soil was 100 mg/kg in Taiwan. The mean content of Cr in brown rice (Fig. 11) is 0.14 mg/kg in Taiwan. The rice production will be reduced when the total Cr concentration in rural soil reaches 250 mg/kg, but the rice should be edible as the Cr concentration in polished rice was lower than 4 mg/kg (Liu et al. 1998). The regulation standard of Cr in soil of Taiwan is proposed not to be changed (250 mg/kg). The relationships indicated that Cr concentration of rice is not higher than 6 mg/kg owing to the increase of Cr concentration in the soil.

Nickel. The regulation levels of the total content of Ni (Fig. 12) in soils in the world....
ranged from 20 to 400 mg/kg. The upper limit of background Ni content in representative rural soil was 60 mg/kg in Taiwan. The Ni content in brown rice grains ranged from 4 to 30 mg/kg when the total Ni content in rural soil ranged from 40 to 320 mg/kg in Taiwan. When the total Ni concentration of rural soils reached 600 mg/kg, the Ni content in polished rice grains was less than 14 mg/kg (Liu et al. 1998). There are no relationships found between Ni concentrations in brown rice and in soils.

**Arsenic.** The regulation levels of the total content of As in soils in the world ranged from 20 to 75 mg/kg. The upper limit of background As contents of representative rural soil was 18 mg/kg in Taiwan. When the total As content in the soils of Taiwan was less than 20 mg/kg, As content of the brown rice (fresh weight) was 0.15 mg/kg in the average and that of the other crops was 0.01 mg/kg in average. When the total As concentration in rural soil reached 60 mg/kg, the rice production will be significantly reduced.

**Lead.** The regulation levels of the total content of Pb in soils around the world ranged from 100 to 1000 mg/kg. The upper limit of background Pb contents of representative rural soil (Fig. 13) was 50 mg/kg in Taiwan. The Pb content in brown rice grains and polished rice grains was almost less than 1 mg/kg when the total soil Pb ranged from 50 to 1350 mg/kg in Taiwan (Liu et al. 1998). We estimated that the rice production will be reduced by 20% if the total concentration of Pb reaches 2,000 mg/kg. This relationship indicated the solubility and mobility of Pb in paddy soil and rice uptake system.

**HYPOTHESIS TO EXPLAIN THE RELATIONSHIPS BETWEEN METALS IN CROPS AND METALS IN SOILS**

National regulations on heavy metals in rice, crops and rural soils have been developed in the Soil Remediation Act of some Eastern Asian countries. Health risk assessment system is also proposed and included into the new Soil Remediation Act to evaluate the food safety and human health after the soil and crops were contaminated. Unfortunately, there are only limited and unreliable databases to be developed in the Southeastern Asian counties.

Land application of manure compost, municipal, commercial and even industrial wastes that is considered beneficial for agriculture is gradually being popular worldwide. However, heavy metals in the compost and wastes accumulated in soils and plants may pose health risks to human and ecosystems. It is important to understand the key assumptions related to the metal behavior in soils and crops whenever the authorities plan to establish or revise the regulations for heavy metal contaminated soil. Because many Asian countries may enact new regulations

![Fig. 11. The relationship between Cr concentration in brown rice grains and in soil extracted by 0.1M HCl for the database of central Taiwan's rice cropping system conducted twice a year from 1994 to 1996. (Database of Liu et al. 1998)](image-url)
following the metal loading limits in Part 503 established by US EPA (2007), the six hypotheses underlying the development of Part 503 are necessary to be introduced and reviewed in this article. Although the emphasis of Part 503 is on the conditions of disposal of sewage sludge, the principles involved could be applied to other metal-containing materials deposited on agricultural land. After which, regulation criteria of heavy metals in soil and the approaches under the development of regulations in Japan, UK, Netherlands, and Germany will be briefly introduced as references.

The title of the six hypotheses with the corresponding proposed authors in parentheses are listed as follows:

- Hypothesis 1: Single-metal abrupt toxicity threshold (Chang 1992)
- Hypothesis 2: Soil-plant barrier (Chaney and Giordano 1977)
- Hypothesis 3: Clean sludge concept (Chaney 1989)
- Hypothesis 4: Uptake plateau concept (Corey et al. 1987)
- Hypothesis 5: Aging effect (Rundle et al. 1982)
- Hypothesis 6: Absence of evidence (USEPA 1995)

_Hypothesis on single-metal abrupt toxicity threshold_. The loading limit for phytotoxic metals on agricultural soils calculated by USEPA is based on this
hypothesis which implicitly assumes that phytotoxic response in crops is an abrupt function of metal concentration in plant tissue (Chang et al. 1992). The schematic diagram of abrupt and gradual toxicity responses of a plant for Zn is shown in Fig. 14 (McBride 2003). It is clearly seen that the threshold concentration of small but economically significant crop yield loss (i.e. 10% reduction) is apparently smaller for gradual than that for abrupt toxicity response. The Zn concentration in maize and other monocots at 50% growth reduction occurrence is in the range of 600-1000 mg/kg (Boawn and Rasmussen 1971), far lower than the value (1975 mg/kg) calculated by the abrupt toxicity responses in the report of Chang et al. (1992). Davis-Carter et al. (1991) reported that the severity of Zn toxicity increased as the plants matured. From the results above, it is doubtful that the simplistic USEPA approach of using short-term assays with young plants to calculate the threshold concentrations is suitable for general application to field conditions and mature crops in all soil types.

From the growers’ point of view, setting phytotoxic threshold at 50% crop yield reduction is unacceptable. The better way to determine phytotoxic threshold is to reassess and discuss under the consideration of site-specific conditions, experimental variability, and other key factors affecting metal toxicity to crops. Taking into account a wide range of yield reductions and the probabilities of reaching those reductions at various metal accumulations in different soil properties may be a better approach to determine phytotoxic threshold (McBride 2003). Besides, the effects of metal interactions in crops on toxicity threshold may also be significant and needed to be assessed.

**Hypothesis on soil-plant barrier.** This hypothesis assumes that the soil-plant barrier limits transmission of many toxic metals through the food chain, as shown in Fig. 15, so the human health risk from food chain is negligible (Chaney and Giordano 1977). For Pb and Hg, their very low solubility in soils demonstrated the soil barrier. Some metals such as Zn, Cu, Ni, Mn, and As will cause plant senescence to prevent metal transmission in food chain (e.g. plant barrier) (Basta et al. 2005). Although the evidences of soil-plant barrier exist, it is doubtful to conclude that the barrier provides protection for the human food chain in all cases, especially for Cd, Zn, Mo, and Se.

Some studies showed that the transfer of Cd from soil to seeds of wheat, corn, sunflower, flax, and palak tissue can be high enough to exceed health standards in some countries, even in slightly or moderately Cd contaminated land (Nan et al. 2002; Sharma et al. 2007; Li et al. 1997). For some toxic metals, the effectiveness of the soil-plant barrier primary depends on plant physiology and associated rhizosphere effects rather than soil properties. The plant physiological processes varied remarkably among plant species, and it is difficult to include the

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**Fig. 14.** Schematic diagram of gradual and abrupt phytotoxic responses in a crop. (Cited from McBride 2003)
species-specific effect on crop uptake of metals in risk assessment of heavy metal contaminated soil. More comprehensive and case-specific research is required to help authorities establish reliable regulations to prevent adverse agricultural impact from toxic metal contaminated land.

**Hypothesis on clean sludge concept.** According to the study of Cd accumulation in lettuce (Fig. 16) with an effect of pH and sludge, this hypothesis assumes that crop uptake of soil Cd is determined by sludge Cd total concentration, when Cd input rate were fixed (Chaney 1990; Chaney 1989). The hypothesis is accepted by USEPA to determine heavy metal limits (exceptional quality, EQ) at or blow which no restrictions were required on the use of sewage sludge applied to land. Although the result of the study conducted by Jing and Logan (1992) showed that plant uptake of Cd was positively correlated with total Cd in sludge, the relationship is weak ($r^2=0.33$). The evidence of the hypothesis is not strong in this study. Other researches have also failed to support the concept of “Clean Sludge”. Zarcinas et al. (2001) found that Cd concentration in wheat grain has no relationship with total Cd in sludge, but depends on sludge properties. Results of some longer-term field studies were also inconsistent to the hypothesis. For example, equal application rate of Cd from sludge with low and high Cd to acid soils after 8 years loadings did not affect plant uptake of Cd (Whatmuff 1999).

On the other hand, the strongly adsorptive ability of sludge is believed to prevent unacceptable solubility or bioavailability of metals in soil after long-term application of sludge with metal concentration below EQ value. Many studies failed to support this “sludge protection” effect on heavy metals in soil. Broos et al. (2001) showed that available Cd and Zn in soil were not apparently different among various sources of contamination (e.g. metal salts, sewage sludge, air pollution, and mining waste). It is clear that the chemical forms of metals in sludge and soil determines the solubility and toxicity of metals, but most regulations of contaminated land or sewage sludge are solely on the basis of total metal concentrations in soil or sludge. More researches for metal bioavailability with key factors in consideration were needed to provide reliable evidence for better and reasonable regulations.

**Hypothesis of uptake plateau concept.** According to the famous “uptake plateau” concept proposed by Corey et al. (1987), the metal concentrations in plant tends to reach a maximum as the cumulative sludge application increases (Fig. 17). Based on this concept some scientists asserted that the risks were overestimated using a linear uptake as a function of metal accumulation. Although the Part 503 for toxic metals is based on a linear extrapolation rather than plateau concept, the
plateau concept supports the strategy of no upper limits on metal loading in Part 503 if metal concentration in the sludge are lower than the calculated limits (Chaney and Ryan 1993). Although the uptake plateau is observed in some studies (Sukkariyah et al. 2005; Chang et al. 1987; and Brown et al. 1998), the evidence of plateau phenomena under field conditions over longer periods is more reliable but rare.

In the long-term field sludge experiments, some scientists found that Cu appeared to show a plateau effect in some crops, particularly maize (Hinesly and Hansen 1983; Hinesly et al. 1984; and Soon et al. 1980). The strong sorption ability of sludge and soil organic matter to Cu and the root barrier to Cu translocation in plants make the plateau phenomena (Poschenrieder et al. 2001). However, results from many long-term field studies showed no plateau phenomena, especially for Cd, Zn, and Ni. (de Melo et al. 2007; McBride et al. 2000; and Chaudri et al. 2001). Furthermore, the concentration of Cd and Zn in the plant tissue is frequently proportional to total metal concentration in the soil under long-term field experiments (McGrath et al. 2000). The short-term sludge experiments with plateau effect may be caused by temporary soil pH increases, growth dilution, ion competition effects, or root toxicity effects that limit the crop’s metal uptake ability (McBride 2003). Further studies and reviews are required to sort out the conditions that the plateau effect really occurs.

**Hypothesis of aging effect.** The “time bomb” theory espouses that upon slow degradation of soil organic matter, with subsequent acidification of the soil under the mineralization reactions involving N, S and toxic metals in soil will become more soluble (McBride 1995). Mehler et al. (1987) added different levels of Cd (as CdSO₄) to 12 soils with relatively long histories of sludge application (5-30 years). The corn tops were grown and harvested after 5 weeks and analyzed for Cd content. Fig. 18 shows the evidence that the sludge decomposition in Burkhardt soil will enhance Cd uptake by corn, comparing with the sludge protection effect on Congaree soil. In contrast, this hypothesis of aging effect assumed that the permanent immobilization ability of soil or sludge can reduce the opportunity of “time bomb” effect. McGrath and Cegarra (1992) conducted a long-term field trial and found the aging effect. The metal-contaminated sewage sludge was applied to the field plot at 16.4 ton/ha/yr from 1942 to 1961. After 1961, the sludge application was replaced by inorganic fertilizers. Soil samples were taken at irregular intervals since the trial began (1942, 1951, 1960, 1967, 1972, 1980, and 1983). Fig. 19 shows that metals extracted by CaCl₂ (Cd, Zn, Ni), NaOH (Zn and Ni), and EDTA (all metals) increased within the first 10 years of sludge addition and then no further change. The applied metals may be adsorbed by oxides or clay minerals in soils, as the hypothesis stands, or leached out in field soils. However, some studies found the sludge-bound

![Fig. 17. Relationship between cadmium concentration in plant leaf and soil, with the data plotted by non-linear regression. (Chang et al. 1996)](image)
metals in soils may become more soluble or bioavailable after the long termination of sludge application (Walter et al. 2002; Antoniadis et al. 2007). In the complicated field conditions, it is difficult to predict the toxic metal behavior after the cessation of sludge application. Monitoring regularly may be a safer way to prevent any unacceptable risks.

**Hypothesis of absence of evidence.** Because of scarce evidence, this hypothesis assumed that the unregulated toxic metals in sewage sludge will not cause significant risks to human and ecosystem. There is no loading limit for Cr present in the USEPA Part 503 rule, because the field study data did not show retardation in the growth of a young plant even at the highest Cr loading (i.e. 3,000 kg/ha). After reviewing new information concerning Cr and the land application of sewage, USEPA decided to delete Cr limit from the land applied sewage sludge regulation (USEPA 1995). It is generally believed that as long as soil organic matter is high, the probability of Cr^{6+} production will be low, as organic matter can reduce Cr^{6+} (James et al. 1997; Banks et al. 2006). However, in tropical area the organic matter in soil decomposes quickly, the risks will be higher in Cr.

Fig. 18. Concentration of cadmium in corn tops harvested in the Congaree and Burkhartd soils with and without sludge (+S, -S) and lime (+L, -L), and with increasing levels of soluble Cd (CdSO_{4}) added. (cited from Mahler et al. 1987)

Fig. 19. Percentage extraction of six metals in four sequential extracts (▲=CaCl_{2}, as water-soluble and exchangeable fraction; ●=NaOH, as organic-bound fraction; * = EDTA, as carbonate-bound fraction; □=aqua regia, as residual fraction) The years referred to are 1940 to 1985. (extracted from McGrath and Cegarra, 1992)
contaminated soil. In addition, soil organic matter shows weak capability to reduce Cr\(^{6+}\) under high pH condition (Bartlett and Kimble 1976). For soils with unregulated metals higher than normal soils, more attention was needed on their potential risks.

Without detailed knowledge of the soil properties, plant physiologic processes, and metal behaviors in site-specific conditions, a comprehensive and reliable regulation is impossible to establish. More studies should be supported by governments to collect situation-specific data as powerful evidence for legislation.

REGULATIONS ON TOTAL CONTENT AND BIOAVAILABILITY OF HEAVY METALS IN SOILS

The updated regulations of heavy metal in United States of America, United Kingdom, Netherlands, Germany, Taiwan, China, and Japan will be introduced briefly in this section. Table 3 summarizes the basis of regulation establishment in these countries. Some regulation criteria for agricultural land and reference websites in these countries are presented in Table 4 and Table 5.

USA (2000)

The cumulative pollutant loading rate (i.e., maximum permissible pollutant loading level, MPPLL) was established by USEPA in 40 CFR Part 503 to regulate soils contaminated with heavy metals from sewage sludge application (USEPA 2007). However, if the applied sewage sludge with heavy metal concentration were lower than the monthly average concentration in Table 3, there is no cumulative pollutant loading restrictions. Different requirements were set for different land uses (i.e., agricultural land, forest, public contact site, lawn, and home garden). Fig. 20 shows how USEPA regulate land application of sewage sludge. The MPPLL of a pollutant was determined by modeling transfer of the pollutants through 12 exposure pathways, assuming that the well-being of the most exposed individual is reasonably protected (USEPA 1989). The MPPLL of heavy metals are listed as follows (kg/ha): As 41, Cd 39, Cu 1500, Pb 300, Hg 17, Ni 420, Se 100, and Zn 2800. More detailed information can be found in the website: http://www.epa.gov/epacfr40/chapt-I.info/chi-toc.htm.

UK (2002)

The Soil Guideline Values (SGVs) for three land uses [i.e. residential (with and without vegetables), allotments, and commercial/industrial] were published in 2002 to understand whether the soil concentration of contaminant X poses a risk to human health or the environment (UK Environment Agency 2007a). The SGVs were derived from the Contaminated Land Exposure Assessment (CLEA) model, a scientific framework developed by government-supported research for assessing the risks to human health from chronic exposure to contaminated soil (UK Environment Agency, 2007b). Since the soil types, site conditions, contaminant behaviors, human activity patterns, and contaminant toxicology are important factors affecting the risks caused by contaminated land, they were considered in the studies for the development of SGVs. The scientific approach applied by UK for establishing the SGVs is a valuable reference (Environment Agency, UK 2007c). The SGVs for the residential with vegetable growing are listed as follows (mg/kg): As 20, Cr 130, Hg 8, Ni 50, Pb 450, and Cd 1(pH 6) 2(pH 7) 8 (pH 8).

More detailed information can be found in the website: http://www.environment-agency.gov.uk/subjects/landquality/113813/672771/?lang=e

Netherlands (1999)

The regulations of heavy metals in soils established by the Netherlands have been very internationally influential. Three types of standards were set in Dutch soil remediation policy; soil remediation intervention values, indicative levels for serious contamination, and target values (VROM 2007a). Those standards varied with soil organic matter and clay content, under the consideration of the fact that contaminant in different soil types cause different risks. Both human and ecotoxicological effects of soil contaminants were taken into account in the extensive studies of the National Institute for Public Health and Environmental Protection for the standards determining. The intervention values for the standard soil (10% organic matter and 25% clay) are listed as follows (mg/kg): As 55, Cd 12, Cr 380, Cu 190, Hg 10, Pb 530, Ni 210, and Zn 720.
Table 3. Summary of the basis for heavy metal contaminated soil regulation in different countries

<table>
<thead>
<tr>
<th>Country</th>
<th>Main considerations to develop the regulation for heavy metals in soils</th>
</tr>
</thead>
</table>
| USA       | 1. A number of risk-based assessments to identify the potential pollutants in the soil system.  
           | 2. Scientific consideration for 12 exposure pathways of pollutants in the food chain.  
           | Reference website: [http://www.epa.gov](http://www.epa.gov) |
| UK        | 1. The human health risk-based scientific framework (i.e. CLEA model)  
           | 2. Scientific consideration for site-specific conditions.  
           | 3. Potential health risks estimated from different land use.  
             | 2. Potential risks calculated from soils with different soil organic matter and clay content  
             | 3. Potential health risks estimated from different land use.  
             | Reference website: [http://international.vrom.nl](http://international.vrom.nl) |
| Germany   | 1. The pathway-based research projects for assessing health risks  
           | 2. Potential health risks estimated from different land use.  
           | Reference website: [http://www.bmu.de/english/](http://www.bmu.de/english/) |
| China     | 1. Potential health risks estimated from different land use.  
           | 2. Heavy metal behaviors at different soil pH levels (pH < 6.5, 6.5-7.5, and > 7.5)  
| Taiwan    | 1. National survey of background and pollution sites and professional recommendations  
           | 2. Regulation criteria of foreign countries  
| Japan     | 1. The human health risk-based studies and pathway considerations  

Fig. 20. Schematic representation of sewage sludge application in land regulated by USEPA.
More detailed information can be found in the website: http://international.vrom.nl/pagina.html?id=7445

**Germany (1999)**

Trigger and action values established in Germany under the consideration of three pathways (i.e. soil-human health pathway, soil-plant pathway, and soil-groundwater pathway) and different land uses (e.g. playgrounds, residential areas, parks and recreational facilities, commercial and industrial areas, agricultural land, vegetable garden, grassland) were reported in the Federal Soil Protection and Contaminated Sites Ordinance (BbodSchV) (Germany Federal Environment Ministry 2007). In addition, precaution values were established to obviate new soil pollution. The results of many research projects supported by the government were the basis of those regulating values. The trigger values for the soil-plant pathway in agricultural land (only Cd with action value but no trigger value, other metals with trigger values but no action values) are listed as follows (mg/kg): As 200 (extracted by aqua regia), Cd 0.04/0.1 (strong Cd-accumulative plant/general plant) (extracted by ammonium nitrate), Pb 0.1 (extracted by ammonium nitrate), and Hg 5 (extracted by aqua regia).

### Table 4. The regulation of heavy metals in soils in USA, UK, Netherlands, and Germany.

<table>
<thead>
<tr>
<th></th>
<th>As</th>
<th>Cd</th>
<th>Cr</th>
<th>Hg</th>
<th>Cu</th>
<th>Ni</th>
<th>Pb</th>
<th>Zn</th>
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<tr>
<td><strong>Cumulative loading rate (kg/ha)</strong></td>
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<tr>
<td>USA (2000)</td>
<td>41</td>
<td>39</td>
<td>17</td>
<td>1500</td>
<td>420</td>
<td>300</td>
<td>2800</td>
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<tr>
<td><strong>Soil guideline value (mg/kg)</strong></td>
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<tr>
<td>Residential with plant uptake</td>
<td>20</td>
<td>2</td>
<td>130</td>
<td>8</td>
<td>--</td>
<td>50</td>
<td>450</td>
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<tr>
<td>Residential without plant uptake</td>
<td>20</td>
<td>30</td>
<td>200</td>
<td>8</td>
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<td>50</td>
<td>450</td>
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<td>Allotments</td>
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<td>75</td>
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<tr>
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<td>1400</td>
<td>5000</td>
<td>480</td>
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<td>5000</td>
<td>700</td>
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<tr>
<td><strong>Intervention value (mg/kg)</strong></td>
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<tr>
<td>Standard soil</td>
<td>55</td>
<td>12</td>
<td>380</td>
<td>10</td>
<td>190</td>
<td>210</td>
<td>530</td>
<td>720</td>
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<tr>
<td>(10% O.M. and 25% clay)</td>
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<td>(10% O.M. and 25% clay)</td>
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<td>Germany (1999)</td>
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**Soil-human health pathway**

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<tr>
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<th>Cr</th>
<th>Hg</th>
<th>Cu</th>
<th>Ni</th>
<th>Pb</th>
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<tbody>
<tr>
<td><strong>Trigger value (mg/kg)</strong></td>
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<td>Playgrounds</td>
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<td>20</td>
<td>--</td>
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<td>50</td>
<td>--</td>
<td>350</td>
<td>1000</td>
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<tr>
<td>Commercial/Industrial</td>
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<td>1000</td>
<td>80</td>
<td>--</td>
<td>900</td>
<td>2000</td>
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**Soil-plant pathway**

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<th>Cu</th>
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<td><strong>Trigger value (mg/kg)</strong></td>
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<tr>
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<td>--</td>
<td>5</td>
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<td>1.5</td>
<td>0.1</td>
<td>2</td>
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<tr>
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<tr>
<td>Agriculture &amp; vegetable garden</td>
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<td>0.04/0.1</td>
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<tr>
<td>Grassland</td>
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<td>2</td>
<td>1300</td>
<td>1900</td>
<td>1200</td>
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**Soil-groundwater pathway**

<table>
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<tr>
<th></th>
<th>As</th>
<th>Cd</th>
<th>Cr</th>
<th>Hg</th>
<th>Cu</th>
<th>Ni</th>
<th>Pb</th>
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<tr>
<td><strong>Trigger value (mg/kg)</strong></td>
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<td>50</td>
<td>1</td>
<td>50</td>
<td>50</td>
<td>25</td>
<td>500</td>
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</tbody>
</table>

*Extracted by ammonium nitrate
*Extracted by aqua regia
*0.04 for Cd-accumulative plants; 0.1 for general plants
More detailed information can be found in the website:
http://www.bmu.de/english
soil_conservation_contaminated_sites/current/aktuell/3874.php

China (2007)
The State Environmental Protection Administration of China (SEPA) has established a regulation of heavy metals for maintaining the soil environmental quality in different land uses in 1996 and 2007. The categories of land uses are classified into 4 groups, including exhibition sites, greenhouse for vegetable production, soils for edible agricultural product, and acceptable levels of residual radionuclides in soil of site considered for release. Different regulation criteria for different pH levels were established in the land uses of greenhouse for vegetable production and soils for edible agricultural product. The regulation criteria of heavy metals in soils for edible agricultural product (soil pH ranged from 6.5-7.5) are listed as follows (mg/kg, total content): Cd 0.3, Hg 0.5, As (paddy soil) 25, As (upland) 30, Cu (rural soil) 100, Cu (vegetables) 200, Pb 80, Cr (paddy soil) 300, Cr (upland) 200, Zn 250, and Ni 50.

More detailed information can be found in the website:
http://www.zhb.gov.cn/tech/hjbz/bzwb/trhj/trhjzlbz/

Taiwan (2000)
In 2000, Taiwan EPA has announced the Soil and Groundwater Pollution and Remediation Act. The regulations of most inorganic and organic pollutants are also developed for rural soils and non-rural soils including industrial park. Soil Pollution Monitoring Standards and the Soil Pollution Control Standards were determined after collecting the results of national survey studies, regulation criteria from foreign countries, and the opinions of professionals and officials. Based on the 7 years of experiences, activities for revising the regulations for different land uses and risk levels are now in consideration. The Soil Pollution Control Standards for rural soils established in 2000 are listed as follows (mg/kg, extracted by aqua regia except As and Hg): Cd 5.0, Cr 250, Cu 200, Ni 200, Pb 500, Zn 600, As 60, and Hg 5.

More detailed information can be found in the website:

Japan (2003)
The Soil Contamination Countermeasure Law was enforced in February 2003 to protect the public health and establish measures to prevent human health hazard caused by the contaminated soil. The most serious contaminant exposure pathways for human were direct ingestion of contaminated soil and ingestion of groundwater contaminated with heavy metals eluted from contaminated soil. Thus, the Soil Concentration Standard and the Soil Leachate Standard were approved to control and assess the risks caused by these two pathways, respectively (Ministry of the Environment, Japan 2007). The Soil Leachate Standards for heavy metals are listed as follows (mg/L, extracted by water): Cd 0.01, Cr^6+ 0.05, Hg 0.0005, Se 0.01, Pb 0.01, As 0.01.

More detailed information can be found in the website: http://www.env.go.jp/en/water/

Brief summary
Each country has its specific environmental and legal conditions, so that simple comparisons of quantitative standard used in different countries can be misleading. Different assumptions have been used to reflect different contaminant behavior, local soil types, and other technical factors in different countries. Therefore, it is usually inappropriate to apply quantitative standard directly from other countries. A full understanding of the contexts within which the standard have been made is required to review the regulations and compare with others. The websites presented in this section provide useful reference sources to understand these countries’ regulation more in detail.

STRATEGIES FOR MONITORING AND REMEDIATION PROJECT

Many soil remediation techniques have been developed and applied in the field scale of contaminated sites of rural soils, including excavation, attenuation by mixing, chemical stabilization, soil washing, phytoremediation, and thermal desorption. The most popular remediation techniques used in Taiwan and
some cases experiences will be briefly described here. On the other hand, what are the new aspects to be considered on the clean up level of soil remediation project? What are the new strategies to be developed for action on the monitoring soil quality and food safety for human health? Netherlands and UK have changed their objectives and policies for soil remediation in recent years. It is worth to know and will also be briefly introduced in this section.

Dilution technique. If heavy metal concentration is lower in the subsurface soil than that of surface soil, deep plow and consequently mixing the two layers can significantly decrease the metal levels to meet the regulation of pollutants in the SGWPR Act of Taiwan. The depth of subsoil should be enough to dilute the total metal concentration of the surface soil (0-20 cm). This turnover and dilution method is thus suitable for “slightly or moderately” contaminated paddy soil, especially for relatively low metal concentrations. This method can significantly decrease the Cd concentration of contaminated soils.

For most of the metals-contaminated paddy soils in Taiwan, soil turnover and dilution method were the most popular remediation methods to be applied in the site because it has the low costs and low risk advantages compared with other soil remediation techniques such as acid washing or applying chelating agent to extract the pollutants from the soils (Taiwan EPA 2007). This dilution method produces lower levels of organic carbon and fertile soil after remediation. Fertilization should be managed for soil cleanup by applying composts and chemical fertilizers to increase the soil fertility and to promote the crop productivity. For paddy soils of Taiwan, plow layer should be rebuilt for storage of the irrigation water.

Chemical stabilization or chemical washing techniques. This technique is to apply the chemical amendments to decrease the mobility or solubility of metals in the contaminated paddy soil and thus to decrease the metal uptake of plants. The reliable reclamation materials were successfully developed in many contaminated sites, including apply lime materials, organic residues, compost, hydrous Fe oxides, hydrous Mn oxides, and zeolites (Chen et al. 2000b).

In Japan, calcium chloride and iron chloride were selected as washing chemicals to restore Cd-contaminated paddy soils in-situ condition. Washing with calcium chloride led to the formation of cadmium chloride complexes,

| Table 5. The regulation of heavy metals in soils in Taiwan, China, and Japan. |
|-------------------|---|---|---|---|---|---|---|---|
|                  | As | Cd | Cr | Hg | Cu | Ni | Pb | Zn |
| **Taiwan EPA regulation (2000)** | | | | | | | | |
| Total content (mg/kg)                      | | | | | | | | |
| Rural soil                                  | 60 | 5 | 250 | 5 | 200 | 200 | 500 | 600 |
| General land use                            | 60 | 20 | 250 | 20 | 400 | 200 | 2000 | 2000 |
| Background content                          | 18 | 3 | 50 | 0.5 | 35 | 60 | 60 | 120 |
| **China regulation (2007)**                | | | | | | | | |
| Total content (mg/kg)                       | | | | | | | | |
| Exhibition site                             | 80 | 22 | 610 | 50 | 600 | 2400 | 600 | 1500 |
| Agricultural land (pH 6.5-7.5)              | 25 | 0.3 | 200 | 0.3 | 100 | 50 | 50 | 250 |
| Greenhouse soil (pH 6.5-7.5)                | 30 | 0.3 | 300 | 0.5 | 200 | 50 | 80 | 250 |
| **Japan regulation (2003)**                | | | | | | | | |
| Soil concentration standardf                | 150 | 150 | 250 | 15 | -- | -- | 150 | -- |
| Soil leachate standard (mg/L)               | 0.01 | 0.01 | 0.05 | 0.005 | -- | -- | 0.01 | -- |

* For Cr⁶⁺ only.
# Unit: mg/kg
enhancing Cd extraction from the soil. This washing also substantially decreased soil level of exchangeable and acid-soluble Cd (Makino et al. 2003; 2006, 2007; Maejima et al. 2007).

**Phytoremediation.** Chen and Lee (1997) reported that rainbow pink was grown in a Cd-contaminated site in northern Taiwan for five weeks, the Cd concentration in the plant shoots significantly increased to 115 mg/kg (74-fold). Lai and Chen (2004, 2005 and 2006) reported that applying EDTA can significantly increase the metal concentration in the soil solution and in the shoots of rainbow pink when it was growing in single or in combined-metals contaminated soils. To reuse these metal-contaminated soils, a large area for phytoremediation experiment was conducted in Central Taiwan during 2005 to 2006, supporting from Taiwan EPA.

Lai et al. (2007) selected 12 plant species from 33 plant species based on the result of small area-experiment (0.1ha) before the beginning of this large area study. Twelve high potential phytoextraction species were selected as: Rainbow pink (*Dianthus chinensis*), Serissa (*Serissa japonica*), French marigold (*Tagetes patula*), Chinese ixora (*Ixora chinensis Lam*), Sunflower (*Helianthus annuus*), Croton (*Codialum variegatum*), Kalanchoe (*Kalanchoe blossfeldiana*), Garden canna (*Canna generalis spp.*), Garden verbena (*Verbena hybrida*), Purslane (*Portulaca olerlua* Linn), Scandent Scheffera umbrella tree (*Schefflera arboricola Hayata*), and Bojers spurge (*Euphorbia splendens*), respectively. A total area of 1.3ha was divided into 12 blocks and one plant species is planted in one block (0.1ha). A phytoremediation experiment using a large area shows that the selected 12 plant species were grown well and there were no toxic symptoms after planting in combined metals (Cu, Zn, Ni, Cr) contaminated site in Central Taiwan. For most of plant species, the final Cr, Cu, Ni, and Zn concentration in their shoots showed significant increase compared to the initial concentration.

Although many hyperaccumulators can accumulate high heavy metal concentration in biomass, their susceptibility to disease, low growth rate, or low biomass will reduce the efficiency of phytoremediation. Therefore, selecting metal-accumulating plants which are also compatible with local environments and even with mechanized cultivation technique can optimize phytoremediation. Since rice, soybean, and maize adapt to the weather and soil conditions in most part of Japan very well and the cultivation system for them were also well established and highly mechanized, Murakami et al. (2007) conducted a pot experiment using the low level of Cd contaminated paddy soils (0.83-4.29 mg/kg) under the upland conditions. The study was done to compare the phytoextraction ability of these crops. They found that Milyang 23 rice (an Indica-Japonica hybrid) accumulated 10 to 15% of the total soil Cd in its shoot after growing for 60 days. Under similar experimental conditions, Ishikawa et al. (2006) reported that the Cd-extracting ability of three rice cultivars were significantly higher than that of *Brassica juncea* (L.), a well-known metal hyperaccumulator. Further research is needed to investigate its accumulating ability in the field. According to the studies mentioned above, phytoremediation with rice cultivars may be a more cost-effective and practical technique, comparing with the dilution technique for the remediation of low Cd contaminated soils.

In China, many phytoremediation projects were also conducted by pot and pilot experiments in slightly and moderately Cu-, Zn-, Cd-, and Pb-contaminated soils (Luo et al. 2005, 2007; Li et al. 2006, 2007; Wu et al. 2006, 2007).

**New aspects and strategies of soil remediation.** There are some fundamental principles under the development of remediation strategies in most European and North American countries (Nathanail and Bardos, 2004):

- The precautionary principle;
- The risk-based philosophy for identifying, prioritizing and assessing the need for remedial action;
- The necessary for preventing future pollution;
- The ‘polluter pays’ principle, with a mechanism for helping innocent landowners.

**Netherlands**

Previously, the soil remediation goal of Netherlands was to make the soil fit for multifunctional use. It requires that all contamination from the soil should be removed. Although under some site-specific situations, the all-removed policy can be replaced by isolating,
controlling and monitoring the contamination, the goal is still too strict and the real risks of soil contamination are not taken into account. The effect of these policies is that the authorities has found it hard to fund and execute all the clean-ups, the required clean-up level is difficult to attain technically, and the progress in remediation operations is little. To solve these problems, the government change the remediation goal as ‘function-oriented and cost-effective’. The new goal focused on making a site suitable for current or planned use. The standard approach or customised approach can be applied depending on site-specific situation (VROM 2007b).

**United Kingdom**
Under the goal of sustainable development, the ‘suitable for use’ approach is used in UK for contaminated land (UK Environment Agency 2007d). The approach recognizes that the risks caused by land contamination will vary greatly depending on the land use and many other factors like the hydrology and geology of the site. Therefore, a site-by-site based risk assessment is recognized as more practicable and reliable approach.

**TOTAL INTAKE OF CD IN THE FOOD CHAIN**

_Cd concentration in the edible aquatic life_. Lee _et al._ (1978b) studied the Cd concentration in 153 edible aquatic life. Samples were collected from 30 stations of 10 rivers in western Taiwan. They indicated that the mean concentration of Cd in different edible aquatic life variable and ranged from 0.54 to 1.80 mg/kg dry weight (DW). According to the World Health Organization’s recommendation and the production of fish as well as shellfish in those rivers of western Taiwan, the intake of Cd via digestive tract does not threaten people health. After that, 115 samples of grass shrimp were collected from 13 counties or cities in Taiwan which had higher production of grass shrimp from November 1984 to July 1985. The mean content of Cd was 210 g/kg fresh weight (FW) (Chang _et al._ 1987).

The Cd concentrations of aquaculture fish including tilapia, milk fish, common carp, eel, and trout for a total of 272 samples were investigated from fifteen major aquaculture counties and cities in Taiwan between July 1986 and June 1987. The Cd content of edible portion of fish are Tilapia 27, milk fish 35, common carp 31, eel 33, and trout 17 g/kg FW, respectively (Loh _et al._ 1990). The survey of heavy metals in cultured shellfish conducted in 1986 with a total 350 samples, were collected from shellfish ponds and markets in Taiwan. The samples included 57 purple clams, 76 short-neck clams, 101 clams and 116 oysters.

The Cd content of purple clams, short-neck clams, clams, and oysters were 128, 209, 164, and 274 g/kg FW respectively (Tsai _et al._ 1990). Fresh imported fish contained < 0.4 to 11 g/kg and canned or salted fish contained from 9 to 42 g/kg. The Cd concentration in fish, meat, and fruit generally contain similar levels and values of 5-10 g/kg FW are representative for those food classes (WHO 1992).

_Cd in different crops (potato, cereal, rice, fruit, vegetables)._ Cd concentration in fruit generally contained 5-10 g/kg FW. Most plant-based foodstuffs containing higher Cd concentration and a value of 25 g/kg FW is considered representative for the staple items, cereals and root vegetables (WHO, 1992). Values in excess of 50-100 g/kg FW are also considered normal distribution surveyed in United Kingdom, Finland, Sweden, Denmark, and the Netherlands.

Some studies indicated that the accumulation of Cd at different rate and the final concentration of Cd in plant tissues are different between food crop species grown on same soils, range from 50 to 500 g/kg DW(Jackson and Alloway, 1992). Some studies also showed same effects that is Cd concentration in crops growing in soils amended with sewage sludge for a long term experiments (Juste and Mench 1992).

Jackson (1990) indicated that the mean concentration of Cd in potato of different countries is almost less than 150 g/kg DW, values ranging from 10 to 90 g/kg DW in regular soils or 80 to 510 g/kg DW in sludge-amended soils. Jackson (1990) also indicated that the mean concentration of Cd in cereal grains of different countries is almost less than 100 g/kg DW, ranging from 20 to 200 g/kg DW in regular soils or 20 g/kg to 3.50 g/kg DW in sludge-amended soils. These experiments were collected from the cereal
crops including wheat, barley, and corn growing in the Netherlands, the United States, UK, Germany and Sweden.

In Taiwan, Li et al. (1994) also made a detailed survey in food crops collected from Taiwan. The conclusion of the study proposed that the Cd concentration in food is lowest in the brown rice (only $70 \text{ g/kg DW}$) highest in vegetables ($>200 \text{ g/kg DW}$), but almost lower than $500 \text{ g/kg DW}$.

In Japan, large scale survey of Cd concentration in rice has been carried out in area where the environmental contamination was suspected on metal mines (Japanese Environmental Agency, 1982). In some area of health effect reports, the Cd concentration in the rice always ranged from 500-2000 $\text{ g/kg FW}$ and daily Cd intake ranged from 200-600 $\text{ g/day/person}$. But in areas of health non-effect reports, the Cd concentration in rice always ranged from 200-700 $\text{ g/kg FW}$ and daily Cd intake ranged from 180-390 $\text{ g/day/person}$ (WHO, 1992).

Page et al. (1987) and Li et al. (1994) also indicated that marked differences also occurred in the accumulation of Cd between various plant organs including seed, root/bulb, fruit, and leaf of fruit, vegetables, and cereal grain. The general conclusion is that the concentrations of Cd in plant tissues were reported to decline in the following order: Roots $>$ stems and leaves $>$ tubers (Page et al. 1987; and Chen 1991).

**Assessment methods of dietary exposure.**

It is known that a number of different methods were used to evaluate the assessment of Cd exposure, but they always gave the same result for a given case study. Four main methods of exposure assessment were proposed for these studies including (1) the standard or total diet study, (2) the duplicate meal, (3) fecal analysis, and (4) diary study (Jackson and Alloway 1992; WHO 1992).

The standard diet method was used in the national survey of developed countries. This technique involves an assessment including the survey of mean consumption of food from a variety of food classes and a mean concentration of Cd in each food class. The most common problem is the potential source of analysis error calculated from the Cd concentration in the food, which was less than the detection limit of Cd in the analysis.

The other methods, duplicate diet and diary study are used to assess the exposure of Cd for specific persons. This method needs a preparation of an extra meal for each meal. Then the meal or its components were analyzed. The results indicated that the predicted Cd exposure made by the dietary record studies, were underestimated.

Another method is use to estimate the daily intake of Cd from the determination of daily fecal output, which is less frequently used than other two methods mentioned above. The main problem is the high variability in the absorption capacity of the diet. In the USA, the estimated dietary exposure based on the fecal analysis is considered lower than direct estimates of dietary intake (WHO 1992). The more cost-effective method for people was the diary studies proposed by Coomes et al. (1982).

**Cd concentration in the total diet studies.**

In European community, the diet generally exposes people to between 18 to 48 $\text{ g Cd/day}$, of which only about 5% is coming from water consumption. The Cd concentration in food crops largely depends upon the concentration of Cd in growing soils. The national mean dietary exposure to Cd varies quite differently in Finland, Germany, Japan, and Britain, ranging from 15 to 57 $\text{ g Cd/day}$. Based on these studies, the variation of dietary exposure to Cd has three components including; 1) total mass of food consumed, 2) composition of the diet, that means how much of the daily intake is derived from what food classes, and (3) the Cd concentration of the food classes.

The Cd exposure profile can be quantitatively calculated from the diet composition which is based on the dietary exposure model. Some countries including in Finland, Germany, Japan, Taiwan, and Britain have supported valuable Cd exposure profile based on the basic and detail survey (Chen et al. 2000a), WHO (1992) also reported that daily intakes of Cd in European countries, New Zealand and the USA are usually about 10 to 25 $\text{ g Cd/day/person}$. These data indicated that the overall Cd exposure from Germany and Japan diets is significantly higher ($>50 \text{ g Cd/day}$) than that of Finland and Britain diets ($<20 \text{ g Cd/day}$). The daily Cd intake in Japan estimated from WHO (1992) is consistently higher than the other parts of the world,
generally ranging from 30 to 50 g Cd/day. In some areas of elevated exposure, average daily intakes ranging from 150 to 250 g Cd/day is considered (WHO 1992). For the areas of normal Cd exposure such as Germany, Sweden, and USA, the mean estimate of Cd daily intake is about 10-30 g Cd/day in but the daily Cd intake in Japan was still higher than other countries which ranges from 30 to 80 g Cd/day. For the contaminated sites of Cd exposure, the mean estimate Cd daily intake is greater than 150 g Cd/day in Japan and 50 to 500 g Cd/day in New Zealand.

**Maximum permissible concentration (MPC) of Cd in food.** After studies on the Cd exposure assessment, several attempts have been made to restrict the dietary exposure to Cd. The most convenient method is to control the Cd levels in the aquatic ecosystem, soils or amended material (such as sewage sludge, biosolid, or other manure composts applied into the soils) and propose the limited Cd concentration in a specific food group. Maximum permissible concentration (MPC) of Cd in the specific group of food has been issued in the developed countries (WHO 1992). Most MPC level in foods is around 100 g/kg, ranging from 50 to 200 g/kg.

**Total intake and uptake of Cd from all pathways.** In uncontaminated regions, assuming an air Cd concentration of 10ng/m³ and a daily inhalation rate of 15 m³ for adult, then the average intake of Cd from the atmosphere per day is about 0.15 g Cd/day, of which about 25% or 0.04 g Cd/day will be absorbed. For persons smoking a pack of 20 cigarettes daily they can accumulate up to about 3 g Cd/day. Cd intake from drinking water based on the daily consumption of 2 liters per day is usually <1 g Cd/day. The average Cd intake from the food including meat, fish, cereal, vegetables and fruit is about 10-25 g Cd/day.

In contaminated regions, assuming an air Cd concentration may reach to the level of 0.5 g/m³ and a daily inhalation rate of 15 m³ for adult, then average intake of Cd from the atmosphere per day is about 7.5 g Cd/day, of which about 25% or 2 g Cd/day will be absorbed. For smokers, a pack of 20 cigarettes daily can accumulate about 3 g Cd/day. Cd intake from the drinking water based on the daily consumption of 2 liters is usually <1 g Cd/day. The average Cd intake from the food including meat, fish, cereal, vegetables and fruit will be variable and related to the contamination and the food and water supplies, ranged from 150 to 200 g Cd/day. The absorption in the body was estimated to be about 5%, the daily uptake from diet would be 8-10 g Cd/day. Therefore, the total daily Cd uptake will depend on the environmental conditions, whether food, water, aquatic organisms and air levels, not exceeding 20 g Cd/day/person.

The major pathways of Cd exposure in the aquatic and agro-ecosystem are via food chain. The total daily Cd uptake will not exceed 20 g Cd/day/person. The concentration from other pathways to the total uptake of Cd is relatively small (less than 3 g Cd/day). In contaminated regions, the total Cd exposure via food chain will reach to greater than 150 or several hundred g Cd/day. It is known that the usual maximum permissible concentration (MPC) level in foods are limited to around 100 g/kg or ranged from 50 to 200 g/kg in order to keep good health through these complicated Cd exposure pathways.

**CONCLUSION**

The dependence of various metals for all aspects of modern life, the increasing world population, and the increasing metal usage makes the soil pollution an inevitable problem and a big challenge to scientists and environmental policy makers. For healthy and sustainable future generations, soil resource should be protected against a slow and insidious poisoning by heavy metals released from mining, industrial, and agricultural activities. Even though there were lots of available literatures on soil pollution, it did not result in greater similarity in the guidelines established by countries worldwide to control the pollution of agricultural soils. This identified the complication of heavy metal behavior in agro-environmental system, the various climatic, geologic, and hydrological conditions in different countries, and the political and nonscientific community factors also affecting the establishment of regulations. Therefore, the site-specific and health-based risk assessment will be more reliable and practical approaches to tackle soil pollution. More scientific evidence from case-specific research, especially the long-term field trial, involving all kinds of key conditions and
factors are necessary to understand the bioavailability of heavy metals in various soil types after long period of time and provide reliable parameters for health-based risk assessments.

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