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Relative Uptake of Cadmium by Garden Vegetables and Fruits Grown on Long-Term Biosolid-Amended Soils

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Cadmium contamination of soils has been viewed as the most dangerous form of trace element contamination of soil. The primary risk posed by Cd contamination is through the ingestion of vegetables grown on Cd-contaminated soil. A variety of garden vegetables selected as representatives of the major food groups were grown on long-term sludge and control plots at high and low pH levels to determine their patterns of Cd uptake in reference to an indicator crop. This was done to determine the potential for a relative uptake index. This relative uptake index (RUI) can be used to assess the risk potential for transfer of soil Cd to the food chain for contaminated sites. Lettuce (*Lactuca sativa*) was used as the indicator crop. For all vegetables excluding dry bean (*Phaseolus vulgaris*), which shows very low Cd uptake, it was possible to define a statistically significant log linear relationship with Cd accumulation in lettuce. When only the more contaminated treatments were included, a more quantitative relation between Cd in the indicator crop and Cd in the other vegetables (including bean) was defined. This indicates that a RUI may be appropriate for risk evaluation on more heavily Cd-contaminated soils.

Introduction

Since the identification of Cd health effects in subsistence farm families in Japan in 1969, which resulted from Zn mine- and smelter-contaminated rice paddies, the presence of Cd in soil has been viewed as one of the most dangerous forms of trace element contamination of soil (*1*). The primary risk pathway associated with Cd-contaminated soils has been identified as soil−plant−human pathway and the consumption of staple grains or garden crops grown on these soils (*2*, *3*). Cadmium can be added to soils through many processes, including phosphorous fertilization (as a contaminant), mining and smelting of Zn and Pb ores (aerial deposition and erosion of slag piles), and application of biosolids to cropland. The growing acceptance of biosolids as a fertilizer and soil conditioner is still met with concern over increased Cd concentration in sludge-amended soil (*4*).

The precise nature of Cd uptake and transfer to edible parts of plants is not completely understood. Several soil factors, including total soil Cd, pH, organic matter, chloride concentration, Zn status, hydrous iron, manganese, and aluminum oxides, and soil particle size have been identified as effecting Cd uptake by plants (*4*−*7*). In addition, plant species and cultivar are factors that influence the Cd concentration in plant tissues grown on a particular soil (*8*−*10*).

Chaney et al. (*9*) suggested that an efficient method for testing the potential human dietary risk associated with ingesting food crops grown on a particular site would be to grow an indicator crop, such as lettuce, on that site. An indicator crop would be chosen based on its relatively high Cd uptake. Measurement of Cd in the indicator crop is much less difficult than with other crops with relatively lower Cd in the edible plant tissues. This would reduce the potential for measurement error. The Cd concentration in the indicator crop relative to other garden foods could be established through a series of field trials, which included a variety of soil series, levels of contamination, and a range in soil pH levels. Chaney et al. (*9*) suggested that this index would be particularly pertinent for soils containing sufficiently high Cd levels to be perceived as posing a potential risk for food chain Cd. Although the factors influencing Cd uptake are not completely understood, the behavior of plant species remains relatively consistent across sites (*8*, *9*, *11*, *12*). A research-based relative uptake index (RUI) could be used to quantify the risks associated with growing a variety of plants on a particular site by growing a single species on that site. The RUI is defined as the ratio of plant Cd:lettuce Cd. By using lettuce, a crop with a high plant Cd:soil Cd transfer factor, a more precise estimate of Cd risk from all garden crops could be made. Further, because a high Cd uptake crop is used to estimate the soil−plant transfer coefficient (leading to a lower coeficient of variation), experimental characterization should also minimize errors in slope measurement.

Several studies have grown a variety of plants on sludge-amended soils in pot studies (*11*−*13*). Kim et al. (*11*) used a variety of sludge-amended soils and 12 species of food plants. Swiss chard was used as the reference crop. All soils used in the study were at relatively high pH (>6.5). Although there were significant differences between soils, the relative concentrations of Cd for all plants across the soils used in the study provided a qualitative basis for an uptake index.

The current study was conducted as part of a U.S. EPA-sponsored cooperative three location field trial (Ohio State University, University of Maryland, and University of California−Riverside) to further assess the potential for the development of a relative uptake index. Potential differ-

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TABLE 1

Total and DTPA-Extractable Soil Metals, pH, and Carbon Content of Long-Term Biosolid Field Plots Used To Measure the Relative Uptake of Zn, Cd, and Cu by Representative Garden Vegetables[a]

				total metal (mg/kg)			DTPA metal (mg/kg)		
sludge	rate	pH	carbon (%)	Zn	Cd	Cu	Zn	Cd	Cu
control		6.85 5.17	1.32 cd	43.8 d	0.33 c	8.3 e	2.15 c	0.09 d	1.0 d
heat-treated	224	6.51 5.07	2.07 a	144. b	1.01 c	35.0 c	20.8 a	0.2 d	6.57 c
Nu-Earth	50	6.40 4.77	1.46 c	119. c	3.0 b	27.0 d	13.0 b	1.38 c	7.1 c
	100	6.35 5.18	1.71 b	181. a	5.5 a	45.1 b	22.5 a	2.49 b	12.6 b
	5 × 20	6.44 5.20	1.87 b	189. a	6.6 a	56.8 a	22.5 a	2.9 ab	15.84a
Cd salt		6.90 5.12	1.25 d	43.8 d	5.7 a	7.2 e	2.9 c	3.28 a	0.62 d

[a] Means followed by the same letter are not significantly different ($p < 0.05$) using the Duncan Waller means separation procedure.

ences between relative uptake estimates from field and pot studies, the importance of pH affects on Cd uptake, the differences in uptake due to soil series, and the possible changes in Cd uptake from sludge-amended soil over time were all factors that could be examined in the field trials (*2, 9*). Each could potentially influence the relative uptake by different crops.

Materials and Methods

The Maryland portion of the cooperative study was conducted at the Hayden Farm research facility of the University of Maryland located at Beltsville, MD. The field plots used in this study had been established in 1976−1978 using five sludges, each applied at several rates, and a Cd salt treatment. Where practicable, treatments were set up at high (6.5) and low (5.5) pH levels using a randomized complete block design with three blocks on a Christiana fine sandy loam soil (Typic Paleudults; clayey, kaolinitic, mesic). Original plots measured 6.4 × 7.9 m (21 × 26 ft.) (*14*). Since they had been established, plots have been rototilled to avoid soil dispersal due to tillage and maintained in no-till crops when not used in metal uptake research.

For this study, a subset of the original plots was used. These included control plots, "Nu-Earth" sludge (210 mg kg^{-1} Cd, 4140 mg kg^{-1} Zn), applied at 50, 100, and 5 × 20 t/ha, Cd salt ($CdCl_2$) applied at the same Cd rate as Nu-Earth 100 t/ha, and a low Cd heat-treated sludge from Annapolis, MD (13.4 mg kg^{-1} Cd, 1329 mg kg^{-1} Zn) applied at 224 t/ha. All of the treatments included in the present study were maintained at both high and low soil pH (Table 1). Before planting each season, the plots received a blanket fertilizer application consisting of 100 kg ha^{-1} each of K as K_2O, P as superphosphate, and N as NH_4NO_3.

Plot boundaries were confirmed by analyzing a series of soil samples taken from outside the plot area into the plot at 30-cm intervals. The integrity of the initial plots was further assured by limiting the area used in this study to an interior section measuring 2.75 × 4.25 m. Soil parameters including total metal concentrations, DTPA (diethylenetriaminepentaacetic acid) extractable metals, organic carbon, and pH were measured (Table 1). Total soil metals were extracted in duplicate using the aqua-regia digestion procedure (*15*). DTPA extractions (*16*) were done in duplicate on combined subsamples collected from the A_p layer of each quadrant of the working plot area. Organic carbon was measured using a LECO C-H-N analyzer (Leco Corporation, 1985). Reported soil pH values are the means of samples collected from the root zone of each crop at the time of each harvest. Soil pH was measured as a 1:1 soil: water (by volume) slurry with deionized water using a combination pH electrode 1 h after mixing.

Six crops were grown as representatives of the major garden vegetable food groups. Lettuce (*Lactuca sativa* var. Paris Island Cos) and cabbage (*Brassica oleracea capitata* var. Copenhagen Market) were selected to represent leafy vegetables, carrot (*Daucus carota* var. Toudo hybrid) was used as a root vegetable, potato was included as a group (*Solanum tuberosum* var. Kennebec), navy bean (*Phaseolus vulgaris* var. Seafarer) as a legume, and tomato (*Lycopersicon lycopersicum* var. Burpee VF) as a garden fruit crop. In addition, a corn (*Zea mays*) inbred that accumulates Cd as compared to other inbreds was grown (*17*). Plants were grown in both the 1991 and 1992 growing seasons. Edible portions of a minimum of five plants per plot were harvested for analysis. Plants were washed in a 0.3% sodium lauryl sulfate solution, rinsed in deionized water, and dried in a 70 °C oven. Dry samples (in duplicate) were ground in a stainless steel Wiley mill. Plant digestions were performed on 2−4-g samples. Samples were ashed for 16 h in a 480 °C oven, dissolved in 2 mL of concentrated HNO_3, and refluxed for 2 h in 10 mL of 3 N HCl. Samples were brought to volume in 0.1 N HCl. Element analysis was conducted using ICP and flame atomic adsorption spectrometry with deuterium background correction.

Statistical analysis was done using SAS version 6.0. Plant Cd was log transformed before statistical analysis to obtain normally distributed data. The year−treatment−pH interaction was not significant for log transformed plant Cd. Data used in calculations include values for all treatments and pH levels for both 1991 and 1992.

Results

Relative Uptake. Lettuce was chosen as the reference crop for this study. The possibility of formulating a relative uptake index was evaluated in two ways: (1) to describe a linear relationship between the change in lettuce Cd and the change in crop Cd across sludge types, soil pH, and total soil Cd levels that might be pertinent to all soils and (2) to determine the reliability of the quotient of plant Cd and lettuce Cd (relative uptake index or RUI) across the more contaminated treatments used in the study for use

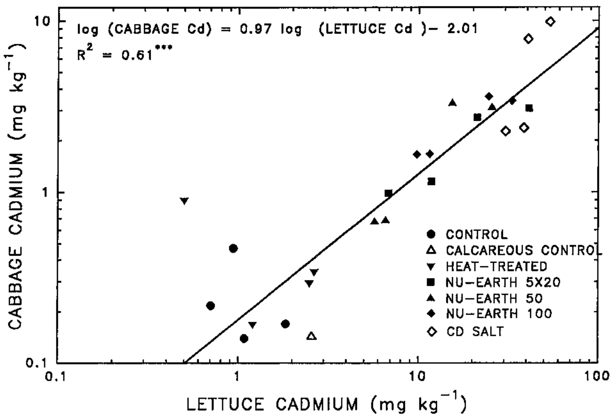

FIGURE 1. Linear regression between log lettuce Cd (mg kg^{-1}) and log cabbage Cd (mg kg^{-1}) for plants grown on long-term biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

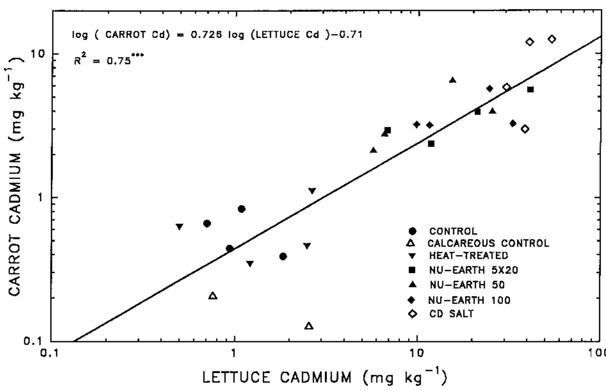

FIGURE 2. Linear regression between log lettuce Cd (mg kg^{-1}) and log carrot Cd (mg kg^{-1}) for plants grown on long-term-biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

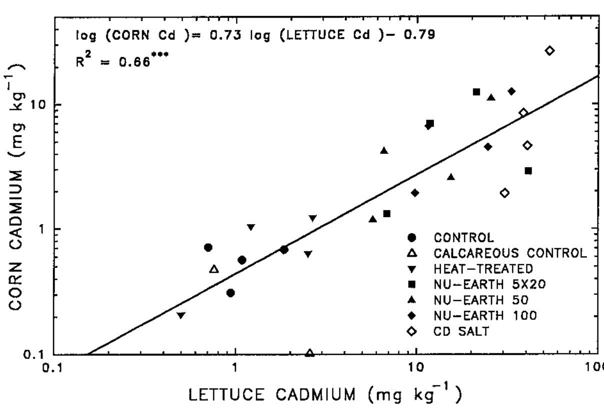

FIGURE 3. Linear regression between log lettuce Cd (mg kg^{-1}) and log corn Cd (mg kg^{-1}) for plants grown on long-term biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

in risk assessment. A linear relationship between log crop Cd and log lettuce Cd was identified for all plants used in this study excluding bean (Figures 1–6). The Cd concentration in dry beans was the lowest of all the crops in the study and was below detection limits for several of the treatments.

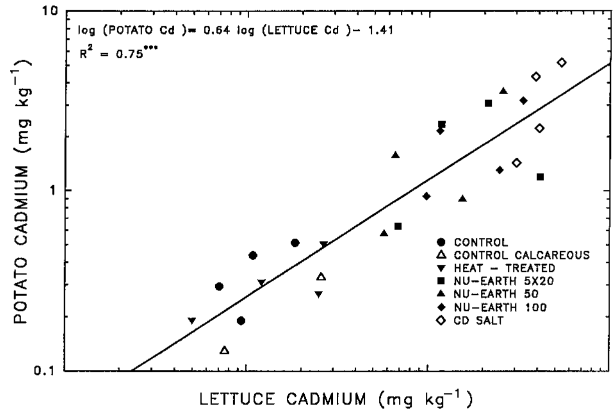

FIGURE 4. Linear regression between log lettuce Cd (mg kg^{-1}) and log potato Cd (mg kg^{-1}) for plants grown on long-term biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

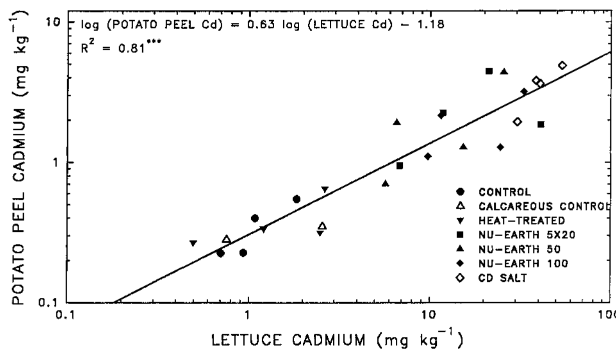

FIGURE 5. Linear regression between log lettuce Cd (mg kg^{-1}) and log potato peel Cd (mg kg^{-1}) for plants grown on long-term biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

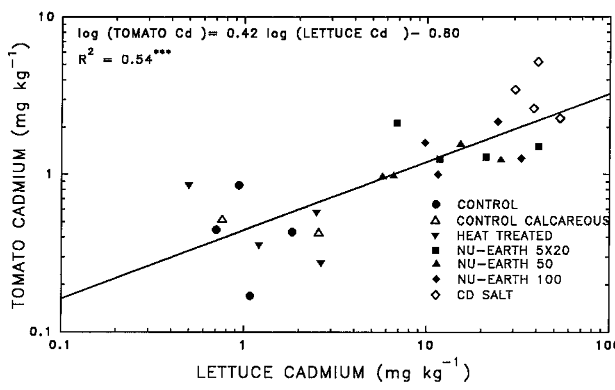

FIGURE 6. Linear regression between log lettuce Cd (mg kg^{-1}) and log tomato Cd (mg kg^{-1}) for plants grown on long-term biosolid-amended plots in Beltsville, MD, during the 1991–1992 growing seasons. Data points shown are sludge treatment × pH means for each year. Different symbols represent different sludge/soil treatments.

The quotient (RUI) was calculated (percentage of plant Cd relative to lettuce Cd) from the existing data set. The significance of year, treatment, and pH were examined, and means separation procedures were carried out. Conditions during the first season were relatively hot and dry with weather during the 1992 season being cool and wet. Although year was statistically significant, there were no treatment by year interactions so that results have been

TABLE 2

Relative Uptake Values for Plant Cd (mg kg^{-1} Dry Weight) Using Lettuce as Reference Crop for Representatives of Major Food Groups Grown on Long-Term Biosolid Amended Field Plots on a Christiana Fine Sandy Loam Soil in 1991−1992[a]

plant	Nu-Earth (t ha^{-1}) 50	100	5 x 20	Cd salt	all treatments
bean	0.027 a*	0.012 a	0.049 a	0.015 a	0.026
cabbage	0.14 a	0.15 a	0.14 a	0.15 a	0.14
carrot	0.39 a	0.25 b	0.26 b	0.27 b	0.29
corn	0.38 a	0.35 a	0.38 a	0.25 a	0.34
lettuce	1.00	1.00	1.00	1.00	1.00
potato	0.14 a	0.11 b	0.12 ab	0.10 c	0.11
potato peel	0.17 a	0.11 b	0.15 a	0.10 b	0.13
tomato	0.13 ab	0.10 ab	0.13 a	0.10 b	0.11

[a] Means within crop and across treatments followed by the same letter are not significantly different ($p < 0.05$) using Duncan Waller means separation procedure.

averaged over the years. The value of the quotient, however, was much more consistent for the plots that contained higher total Cd. This trend was more pronounced in the 1992 growing season. Overall, plant uptake of Cd was lower in 1992 than in 1991. This made the relative uptake ratios narrower for treatments that were lower in total soil Cd and total plant Cd. Therefore, while it was possible to define a relatively consistent RUI for the more contaminated treatments, it was not possible to determine a consistent value for RUI in the lower Cd treatments (as noted in ref 9). It is possible that, at lower phytoavailable total soil Cd, the relationship between total soil Cd and plant Cd varies more significantly with plant species or that the low Cd:Zn ratio alters the relative Cd uptake among species. Zinc is a necessary plant nutrient and is closely related to Cd. Zinc and Cd uptake mechanisms in plants may be related. It is also possible that, at very low levels of Cd uptake, soil factors other than total Cd, whose relative influence is not clearly understood, may effect each species differently. As the goal of defining the RUI was to facilitate the risk assessment process for food chain transfer of Cd from potentially dangerously contaminated soils, the low reliability of the index in cases that do not pose a risk does not diminish the potential usefulness of the RUI. Calculated values for RUI in the more contaminated treatments are reported (Table 2).

The results of this study indicate that it is possible to develop a useful, quantitative relationship between the Cd concentration of the vegetables used in this study and lettuce. Although there were some significant differences between treatments in the RUI in the more contaminated soils, the differences are relatively small. For the RUI to be applied for actual risk assessment, the values determined in this study need to be compared with the values calculated under different soil conditions in additional studies to determine their applicability on a broader scale. Although this is the first field study to develop an RUI, a comparison to previous pot studies confirms the potential usefulness of the RUI. A RUI developed in a pot study using swiss chard as a reference crop by Kim et al. (*11*) also showed less variation for more contaminated soils relative to control soils. Index numbers for potato and tomato were both 0.10 in the present study. Kim et al. defined RUI values of 0.21 and 0.22 for tomato and potato. Different cultivars were used in each study. This indicates that both vegetables behaved similarly under different environments. In another pot study using sludge-amended soils, Davis and Carlton-Smith (*12*) used a cos lettuce as a reference crop. Their RUI value for cabbage (0.16) is very similar to the value obtained in this study (0.14) despite the use of a different cultivar. Values obtained for tomato are also comparable, 0.07 and 0.11 for Davis and Carlton-Smith and the present study, respectively.

If further field studies find consistent results, relative uptake ratios between garden crops and lettuce can be reliably used to estimate soil Cd transfer to edible tissues of garden foods. It is also important to incorporate the probable range of Cd uptake within a particular species (due to cultivar variation) to fully develop the relative uptake index. This would provide a simple and direct method to assess possible risk associated with Cd contamination.

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