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Evaluation of cadmium transfer from soil to leafy vegetables: Influencing factors, transfer models, and indication of soil threshold contents

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ABSTRACT

Food chain contamination by soil cadmium (Cd) through leafy vegetable consumption poses a threat to human health. It is imperative to understand the relationship between Cd phytoavailability in soils and its uptake in common leafy vegetables. A large-scale field survey in Zhejiang Province, southeast China, was conducted to develop models to evaluate the Cd phytoavailability to leafy vegetables based on soil properties and to establish soil Cd thresholds based on food safety. The empirical models developed in this study explained the combined effects of soil properties and diethylenetriaminepentaacetic acid (DTPA)-extractable Cd content on Cd phytoavailability to leafy vegetables. The Cd accumulation in celery, pak choi, and amaranth was quantitatively predicted by measurement of DTPA-extractable soil Cd and soil pH, organic matter, cation exchange capacity and clay content. For predicting Cd accumulation, the DTPA-extractable Cd, pH and clay content had a major influence in lettuce; and for water spinach, the DTPA-extractable Cd, pH, and cation exchange capacity had a major influence. Soil DTPA-extractable Cd was suitable to be used as Cd thresholds in soils cultivating celery, amaranth, pak choi, lettuce, and water spinach, with values of 0.24, 0.13, 0.23, 0.32, and 0.37 mg kg^{-1}, respectively. However, the threshold values of soil total Cd were 0.26, 0.34, and 0.83 mg kg^{-1} for amaranth, celery, and pak choi fields, indicating that the current soil quality standard (GB 15618-1995) for soils cultivating different types of vegetables could be overestimated or underestimated for Cd contamination and the associated risk. This study will provide a useful reference for controlling Cd contamination in common leafy vegetables and developing sustainable production of leafy vegetables.

1. Introduction

Cadmium (Cd) is an important environmental pollutant that is toxic to humans. Among all the toxic heavy metals, Cd is the most mobile and bioavailable (Rafiq et al., 2014). Its bioaccumulation in the food chain surpasses all other heavy metals due to its high mobility in soil (Mahler, 1978). In humans, Cd exposure can cause multiple adverse effects, including renal and hepatic dysfunction and testicular damage (Stohs et al., 2001).

With improved economic development and standards of living, leafy vegetables have become a significant part of the human diet. However, leafy vegetables can accumulate higher concentrations of Cd in edible parts even when grown in soils with low levels of Cd (Baldantoni et al., 2016; Chen et al., 2010). Leafy vegetable consumption is one of the most important pathways of human exposure to Cd in several areas of the world including China (Datta and Young, 2005; Huang et al., 2014; Yang et al., 2016). Therefore, security of leafy vegetables should be a high priority to ensure human health (Ye et al., 2015).

To limit the accumulation of Cd in leafy vegetables, a good understanding of their accumulation properties is crucial. Accumulation of Cd in leafy vegetables is dependent on surrounding environmental factors (Rafiq et al., 2014; Ye et al., 2015). Significant factors contributing to vegetable uptake of Cd are vegetable species and soil properties: e.g. pH, organic matter (OM), cation exchange capacity (CEC), and texture (Datta and Young, 2005; Nabulo et al., 2012; Yang et al., 2016). Therefore, risk assessments in soil–vegetable systems should include physicochemical characteristics of soils.

Including soil variables in predictive models for Cd accumulation in leafy vegetables would provide a more risk-based approach to the improvement of soil quality standards (Ding et al., 2013a). Some models

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have been developed to describe Cd transfer from soil to leafy vegetables (Rafiq et al., 2014; Yang et al., 2009). However, these models were developed on greenhouse pot experiments using Cd-spiked soils, which differ from real agricultural soils. Therefore, it is important to develop models factoring in soil variables for predicting Cd accumulation in leafy vegetables based on field data.

To assess the risk of Cd contamination, soil Cd thresholds for leafy vegetables should be established using predictive models (Rafiq et al., 2014; Xiao et al., 2013). The current soil quality standard in China (GB 15618-1995) does not consider such factors as specific crop, metal phytoavailability, and soil properties (National Environmental Protection Agency of China, 1995). However, vegetable uptake and transfer of Cd is especially affected by vegetable type and species (Hu et al., 2017; Pan et al., 2016; Ye et al., 2015). Therefore, setting up a single standard of Cd for soils cultivating different types of vegetables might overestimate or underestimate Cd contamination and the associated risk.

Zhejiang Province in the southeast of China is a rapidly developing region with a high population density. Intensive management such as fertilization and high cropping index has caused significant soil problems including heavy metal accumulation in field soils. Hence, Zhejiang Province was chosen as the case study area: (i) to evaluate the combined effect of soil properties on the phytoavailability of Cd to leafy vegetables, using regression-based predictive models developed to correlate Cd phytoavailability with common soil properties and with Cd concentrations in soil extractable fractions; and (ii) to establish Cd thresholds for soils used for cultivating common leafy vegetables. The investigation relied on a large-scale agricultural survey accompanied by paired soil–vegetable sampling. The study would provide a theoretical reference for safe production and sustainable development of leafy vegetables in Zhejiang.

2. Materials and methods

2.1. Selection of sites and sampling

Five main vegetable production regions in Zhejiang Province were selected for sampling, including Hangzhou, Shaoxing, Jiaxing, Ningbo, and Wenzhou City. A total of 123 above-ground vegetable samples of 5 usual types from Zhejiang were collected from vegetable fields, and 123 agricultural soil samples (0–20 cm in depth) were obtained from the corresponding sites using a stainless-steel auger. Each vegetable/soil sample was made from a mixture of five sub-samples. Samples were placed in plastic bags and brought to the laboratory for analysis. A global positioning system (GPS) was used to locate the sampling sites and the soil type was marked along it (Fig. 1). All sampling sites were chosen according to field survey characteristics such as vegetable types, their planting patterns, planting years, and soil types and properties. The vegetable and sampling number were *Apium graveolens* L. (celery, 20), *Lactuca sativa* L. (lettuce, 23), *Brassica chinensis* L. (pak choi, 31), *Amaranthus tricolor* L. (amaranth, 22), and *Ipomoea aquatica* Forsk. (water spinach, 27). For each cultivar, the sampling sites, corresponding soil types, and sample numbers in each site are listed in Table 1. Sampling was conducted during March–June 2016.

Leafy vegetable samples were washed carefully with tap and deionized water. All samples were stored at 4 °C and analyzed within 24 h. Soil samples were firstly air-dried at room temperature and then ground to pass a 2-mm sieve. A small portion (50 g) of each soil sample was further ground with an agate mortar and pestle to pass a 0.149-mm sieve to determine the concentration of total Cd.

2.2. Chemical analysis

Soil pH was measured using soil/de-ionized water at 1:2.5 (w:v). Soil CEC was determined according to Hendershot and Duquette (1986). Soil OM was determined by the modified Walkley–Black titrimetric procedure (Storer, 1984). Particle size distribution was determined according to Day (1965).

Soil samples were digested by HNO_3–$HClO_4$–HF (5:1:1) and vegetable samples by HNO_3–$HClO_4$ (5:1) in a microwave digestion system (Xiao et al., 2013) in preparation for measuring total Cd contents. Soil samples of 5 g were shaken with 25 mL of 5 mmol L^{-1} diethylenetriaminepentaacetic acid (DTPA) for 2 h, and then the suspension was centrifuged at 4000 rpm for 30 min, and filtered through 0.45-μm filter paper. Then the Cd concentration was determined by inductively coupled plasma-mass spectrometry (X-Series II, Thermo Electron Corporation, Madison, USA).

For Cd analysis, two certified reference materials (soil GBW07457 and plant GBW10048) approved by General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China were used to validate the digestion and analysis procedures as part of the QA/QC protocol. The Cd values determined in reference materials were within their certified concentration ranges, with recovery rates of 93.5% and 95.2% for GBW07457 and GBW10048, respectively. Three replications were conducted for each sample.

2.3. Statistical analysis

Means of data were compared by least significant difference tests at $p < 0.05$. Linear regression and multiple regression analyses were performed using the statistical package SPSS 18.0 for Windows (CoHort Software, Berkeley, CA, USA).

3. Results and discussion

3.1. Characteristics of soils

A summary of selected soil physicochemical properties and Cd concentrations are presented in Table 2. Soil pH ranged from strongly acidic (4.14) to mildly neutral (7.48) with mean pH levels ranging between 5.30 and 6.61. Soil organic matter (OM) ranged from 8.01 to 44.68 g kg^{-1} with average value between 20.12 and 28.60 g kg^{-1}, CEC values ranged from 9.36 to 48.15 cmol kg^{-1} with average value between 15.29 and 22.43 cmol kg^{-1}, and soil clay fractions ranged from 9.20% to 43.60% with average value between 28.74% and 35.60%, indicating that soils collected from different areas varied widely in their physicochemical characteristics, and could possibly influence soil Cd phytoavailability.

Total Cd in soils ranged from 0.08 to 1.20 mg kg^{-1} covering a range that has been classified as background levels to moderate polluted soil, and had distinct geographical distribution. The average contents of total Cd in soils decreased in order: Wenzhou > Hangzhou > Shaoxing > Jiaxing > Ningbo, respectively with corresponding values of 0.44, 0.29, 0.25, 0.21, and 0.17 mg kg^{-1}. Compared with the Chinese Soil Quality Criterion (GB 15618-1995), the number of soil sample exceeding the proposed limit (0.3 mg kg^{-1}) also followed the order of Wenzhou (16) > Hangzhou (8) > Shaoxing (7) > Jiaxing (6) > Ningbo (0). Song et al. (2012) and Zhang et al. (2005) reported that Cd pollution in Wenzhou is a serious concern due to rapid industrialization and urbanization. Industrial pollution, sewage irrigation, and fertilizer application could have contributed to Cd contamination in these areas (Ye et al., 2015).

The DTPA-extractable Cd accounted for 10.0–43.8% (with a mean value of 19.6%) of total Cd in soil. Compared to other investigations, the results showed much higher Cd phytoavailability. This may be due to the anthropogenic input of Cd into the soil, in addition to the soil properties, especially the acidity of soil in the study areas (Yang et al., 2015; Ye et al., 2015).

3.2. Cd concentrations in different types of leafy vegetables

The Cd contents varied among the celery, lettuce, pak choi,