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Assessing cadmium exposure risks of vegetables with plant uptake factor and soil property☆

Yang Yang [a, b], Andrew C. Chang [c], Meie Wang [a], Weiping Chen [a, b, *], Chi Peng [a]

[a] State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, PR China
[b] University of Chinese Academy of Sciences, Beijing, 100049, PR China
[c] Department of Environmental Sciences, University of California, Riverside, CA 92521, United States

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A B S T R A C T

Plant uptake factors (PUFs) are of great importance in human cadmium (Cd) exposure risk assessment while it has been often treated in a generic way. We collected 1077 pairs of vegetable-soil samples from production fields to characterize Cd PUFs and demonstrated their utility in assessing Cd exposure risks to consumers of locally grown vegetables. The Cd PUFs varied with plant species and pH and organic matter content of soils. Once normalized PUFs against soil parameters, the PUFs distributions were log-normal in nature. In this manner, the PUFs were represented by definable probability distributions instead of a deterministic figure. The Cd exposure risks were then assessed using the normalized PUF based on the Monte Carlo simulation algorithm. Factors affecting the extent of Cd exposures were isolated through sensitivity analyses. Normalized PUF would illustrate the outcomes for uncontaminated and slightly contaminated soils. Among the vegetables, lettuce was potentially hazardous for residents due to its high Cd accumulation but low Zn concentration. To protect 95% of the lettuce production from causing excessive Cd exposure risks, pH of soils needed to be 5.9 and above.

1. Introduction

Cadmium (Cd) polluted agricultural soils were found around the world (Chen et al., 2008; Anwar et al., 2016; Rizwan et al., 2017) and exposures to food grown on Cd-tainted soils might constitute a public health hazard (Reeves and Chaney, 2001; Rizwan et al., 2016). For non-cigarette smokers, dietary intakes of cereal grain and vegetable were the primary sources of one's Cd exposure (Shi et al., 2009; Chaney, 2012), accounting up to 80% of the daily body load (Clemens et al., 2013; Swartjes et al., 2013; Zhao et al., 2017). To properly assess the human health risks, it would be imperative that Cd concentrations of harvested crops be put in perspective.

Plant uptake factor (PUF), denoting the ratio of pollutant contents in edible plant tissue to that of the soil where crops were grown, characterizes how crops accumulate soil-borne pollutants (McBride et al., 2014; Antoniadis et al., 2017). Normally, the Cd pool of polluted soils would be significantly larger than the amounts absorbed by harvested plants (Augustsson et al., 2015). Assuming a steady-state equilibrium and linear mass transfer of Cd from soil to plant, the PUF would be crop species and be constant across soils (Chen et al., 2009; Boim et al., 2016). In this manner, the Cd concentrations of vegetables grown on a contaminated soil might be estimated via their respective PUFs (Swartjes et al., 2013). The relationships however were empirical and were derived from results of controlled experiments in which the plants were cultivated in pots filled with metal-spiked soils (Anwar et al., 2016; Ding et al., 2016). The linear Cd mass transfer model broke down when the PUF was extrapolated to depict realistic situations of the production fields (McBride et al., 2014; Novotná et al., 2015).

The Cd PUF of vegetables produced even in the same vicinities would be subject to the influences of spatially varied micro-growing environment (Zhang et al., 2011; Rehman et al., 2017). Cultivated side by side, lettuce, peanut, and potato resulted in higher Cd PUF than those of bean, cabbage and onion (Chen et al., 2009; Rehman et al., 2017; Yang et al., 2017). In addition to the Cd level of soils, the Cd uptake of field-harvested plants was

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* Corresponding author. State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, PR China.

E-mail address: wpchen@rcees.ac.cn (W. Chen).

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susceptible to variability the field's pedological, microclimatological, and agronomical attributes that changed across the landscape (Wang et al., 2008; Chaney, 2012). Soil factors contributing to the PUF variations were soil pH, organic matter, cation-exchange capacity, texture, and iron/manganese oxide content (McBride et al., 2014; Ding et al., 2016; Yang et al., 2017). For vegetables harvested *in situ*, the PUFs were classified according to plant species and within a species the PUFs instead of being constant followed a lognormal distribution (Chen et al., 2009; Zhang et al., 2011). The Cd PUF of a vegetable therefore should be a probability distribution when they were used in assessing human exposure risks (Swartjes, 2009, 2013; Boim et al., 2016). Accordingly, the resulting dietary exposures to Cd would be probabilistic instead of deterministic depending on probability distributions of the PUFs (Augustsson et al., 2015) and the outcomes of health risk assessments would also be probabilistic in nature (Hosseini Koupaie and Eskicioglu, 2015; Hu et al., 2017).

Zhuzhou (27.83°N lat., 113.16°E long.) was the center of a metal mining/smelting/processing region in southern China (Fig. SI1). It encompassed 4000 km^2 and had 2.4 million residents. Agricultural soils in this region are primarily acidic Ultisoils and have been used for growing vegetables for many years (Chen et al., 2008). In this area, the Cd concentrations of agricultural soil were frequently found to exceed the threshold and the levels were rising (Chen et al., 2008; Wang et al., 2008). The Cd concentrations of harvested vegetables and soils from production fields in this densely populated metropolis were sampled to illustrate the changes of Cd PUFs in nine vegetables residents consumed year around. The data were used to demonstrate the probabilistic nature of Cd PUF distributions and the PUF data might be normalized based on soil factors. A probability-based model was developed to assess the residents' dietary exposures to Cd through consumption of locally harvested vegetables.

2. Material and methods

2.1. Data collection

One thousand and seventy seven (1,077) pairs of vegetable versus soil samples were collected from production farms throughout the Zhuzhou industrial center (Fig. SI1). Sampled vegetables included loofah (*Luffa cylindrical* L.), carrot (*Daucus carota* L.), radish (*Raphanus sativus* L.), bok choy (*Brassica rapa var. chinensis*), cabbage (*Brassica oleracea* L.), celery (*Apium graveolens* L.), Chinese cabbage (*Beassica pekinensis* L.), lettuce (*Lactuca sativa* L.), and mustard (*Brassica juncea* L.), and the sample proportion for these nine species were 8.7, 6.1, 11.4, 22.0, 8.6, 12.4, 12.7, 11.2, and 6.7%, respectively.

Soil samples were dried, sieved, and grounded for chemical analysis. The soil pH (at 1:2.5, soil:water ratio), soil organic matter (based on the $K_2Cr_2O_7$–H_2SO_4 digestion of soils), cation-exchange capacity (treated with 1 mol NH_4OAc at pH 7.0), and clay content (based on the hydrometer method) were analyzed according to methods in Lu (2000). Vegetable samples were washed and the edible parts were dried in an oven at 60 °C and then milled for chemical analysis. Soil samples were digested in a mixture of HCl-HNO_3-HF-$HClO_4$ (Shi et al., 2009), whereas vegetables were digested in a concentrated HNO_3-$HClO_4$ solution (Wang et al., 2008). The graphite furnace atomic absorption spectroscopy was used for determination of Cd and Zn concentrations of soil and vegetable samples. For quality control and assurance purposes, certified reference materials GSS-5 for soil and GSB-5 and GSB-25 for vegetables were included in the assays. The basic physicochemical properties of soil and vegetable properties at the 1077 vegetable producing fields was listed in Table SI1 and Table SI2.

2.2. Data normalization

The Cd concentration of soils and harvested vegetables varied spatially across the study area. The resulting PUF reflected the inherent differences of both plant species and properties of soils where they were cultivated (Antoniadis et al., 2017; Rehman et al., 2017). Normalizing the data in terms of their respective soil properties would stabilize the PUFs (Chen et al., 2009; Swartjes et al., 2013). The normalized relationship between PUF and respective soil properties was derived using the extended Freundlich-type function (Boim et al., 2016) that:

$$\mathrm{Log}[\mathrm{PUF_{obs}}] = \alpha_0 + \alpha_1 \times pH + \sum \alpha_\mathrm{i} \log[\mathrm{X_i}] \tag{1}$$

where $\mathrm{PUF_{obs}}$ was the observed PUF, $\mathrm{X_i}$ denoted the ith soil property parameter in which i = pH, clay [%], SOM [%], and CEC [cmol kg^{-1}] corresponding to each $\mathrm{PUF_{obs}}$. The parameters of normalization, α_0, α_1, and α_i were fitted parameters corresponding to the $\mathrm{PUF_{obs}}$.

The resulting function was evaluated according to the root mean square error (RMSE) and explained variance (Swartjes et al., 2013; Novotná et al., 2015). If the obtained parameters (α_0, α_1, and α_i) were significant, the normalized PUF ($\mathrm{PUF_{nor}}$) would be obtained based on eq. (2):

$$\mathrm{PUF_{nor}} = 10^{\alpha_0 + \alpha_1 \times pH + \sum \alpha_\mathrm{i} \log[\mathrm{X_i}]} \tag{2}$$

If the resulting transfer function was not significant, the PUF would then be represented by the geometric mean of population ($\mathrm{PUF_{geo}}$) as the field-acquired PUF followed a lognormal distribution (Chen et al., 2009; Zhang et al., 2011). In this case, the soil property correction factor (CF) (Van den Berg et al, 1993), was employed to normalize the site-specific nature of the non-significant PUF that:

$$\mathrm{PUF_{nor}} = \mathrm{PUF_{geo}}/\mathrm{CF} \tag{3}$$

where CF was related to the organic matter and clay contents of soil that:

$$CF = [(0.4 + (0.007 \times \mathrm{Clay}) + (0.021 \times \mathrm{SOM}))]/0.875 \tag{4}$$

where SOM and clay represented the site-specific soil organic matter and soil clay contents in %, respectively. Detailed description for CF can be found elsewhere (Van den Berg et al, 1993; Swartjes et al., 2013).

The intra-species variation coefficients of observed vs. normalized Cd PUF ($\mathrm{IV_{obs}}$ vs. $\mathrm{IV_{nor}}$) showed the extent of PUF distributions were stabilized by normalization processes (Ding et al., 2016):

$$\mathrm{IV_{obs}} = \sqrt{\sum_{\mathrm{i}=1}^{\mathrm{k}} \left(\mathrm{PUF^i_{obs}} - \overline{\mathrm{PUF_{obs}}}\right)^2 \Big/ (\mathrm{k}-1)} \Big/ \overline{\mathrm{PUF_{obs}}} \tag{5}$$

$$\mathrm{IV_{nor}} = \sqrt{\sum_{\mathrm{i}=1}^{\mathrm{k}} \left(\mathrm{PUF^i_{nor}} - \overline{\mathrm{PUF_{nor}}}\right)^2 \Big/ (\mathrm{k}-1)} \Big/ \overline{\mathrm{PUF_{nor}}} \qquad (6$$

where $\mathrm{PUF^i_{obs}}$and $\mathrm{PUF^i_{nor}}$are the ith observed and normalized PUF where i = 1, 2, 3, …., k. $\overline{\mathrm{PUF_{obs}}}and\overline{\mathrm{PUF_{nor}}}$ were the mean of the observed or normalized PUF population, k is the number of observations.