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Evaluating the potential health risk of toxic trace elements in vegetables: Accounting for variations in soil factors

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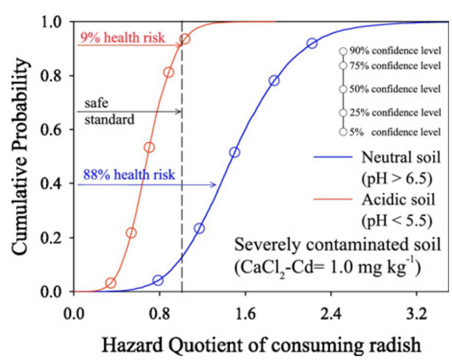
HIGHLIGHTS

- Accumulation of toxic trace elements in vegetables is governed by multi-factors.
- The Cd-Zn relationships depend on the source of Cd contamination.
- Radish is toxic for its high consumption rate and high Cd content but low Zn accumulation.
- Soil pH below 5.3 is a poor practice for vegetables growth.
- Site-specific risk assessment is needed for better and safer vegetable production.

GRAPHICAL ABSTRACT



Hazard Quotient of consuming radish in contaminated soil in different pH conditions

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ABSTRACT

Vegetable crop consumption is one of the main sources of dietary exposure to toxic trace elements (TEs). A paired survey of soil and vegetable samples was conducted in 589 agricultural sites in the Youxian prefecture, southern China, to investigate the effect of soil factors on the accumulation of arsenic, cadmium, mercury, and lead in different vegetables. A site-specific model was developed to estimate the health risk from vegetable consumption. The TE concentration varied in different plant species, and rape can be cultivated in contaminated areas for its potential use in restricting the transfer of TE from soil to edible plant parts. The accumulation of TEs in vegetables was governed by multiple factors, mainly element interaction, metal availability (extractable $CaCl_2$ fraction), and soil pH. Soil Zn may promote Cd accumulation in vegetables when soil Cd/Zn ratio > 0.02. Cadmium is a major hazardous component. About 80.8% of the adult populations consuming locally produced vegetables had a daily Cd intake risk above the safe standard. Among investigated vegetables, radish is potentially hazardous for populations because of its high consumption rate and high Cd content but low Zn accumulation. The consumption of radish cultivated in highly acidic soil ($4 < pH \le 5$) and high Cd contamination ($CaCl_2$-Cd = 1.0 mg kg^{-1}) had a significant probability (89.4%) to be above the safe standard; while this risk was significantly decreased to 8.9% in soil of near-neutral pH ($6 < pH \le 7$). The wide range of TE concentrations and soil factors suggests that a site-specific risk assessment is needed for better and safer vegetable production.

1. Introduction

Toxic trace elements (TEs) contamination in agricultural soils is of great concern as a result of anthropogenic activities (Samsøe-Petersen

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et al., 2002; Reeves and Chaney, 2008). Arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb), which are highly toxic, are the most closely monitored metals and result in different chronic and acute conditions (McLaughlin et al., 1999; Chaney et al., 2004; Zhang and Wong, 2007; Spanu et al., 2012). Metal concentration in vegetables is a major health concern (McLaughlin et al., 2011). In some areas, vegetable consumption contributes up to 49%, 70%, 45%, and 40% of the dietary intake of As, Cd, Hg, and Pb, respectively (Hough et al., 2004; Wang et al., 2005; Liu et al., 2010; Nabulo et al., 2011, 2012).

China has the highest global vegetable production (Liu et al., 2004). However, vegetable fields in China are often close to industrial areas that discharge part of their chemical wastes into agricultural soils (Liu et al., 2004; Zeng et al., 2008). In a prefecture of Hunan, As and Cd concentration in agricultural soil affected by the Pb and Zn mine discharges exceeded the limits specified by Chinese standards 24 and 13 times, respectively, and the concentration of these metals in vegetables exceeded the standard 6.6 and 8.5 times, respectively (Liu et al., 2006). Zeng et al. (2008) reviewed reports of TE accumulation in vegetable fields in China since 1989, showing that Cd in 24.1% of soils exceeded the national soil quality standard, followed by Hg (10.3%) and As (9.2%). Arsenic, Cd, Hg, and Pb can be co-absorbed by vegetables (Samsøe-Petersen et al., 2002; Wang et al., 2005; McLaughlin et al., 2011). The daily intake of vegetables in China is as high as 402 g FW d^{-1}(SEPAC, 2013), thus posing a greater health risk than in other regions.

The availability of TEs to vegetables depends on both cultivars and soil factors (Mckenna et al., 1992; Liu et al., 2006; Ding et al., 2016). An analysis of 16 potato cultivars grown in a clean agricultural area in Turkey showed that the Cd and Pb levels were in the range of 0.08–0.32 mg kg^{-1} and 0.5–0.8 mg kg^{-1}, with significant differences between cultivars (Öztürk et al., 2011). Ding et al. (2013, 2014, 2016) cultivated carrots in soils from different regions of China covering a wide range of soil properties, showing that TEs in carrots varied greatly, 180-fold for Cd, 21-fold for Hg, and 360-fold for Pb, respectively. Increased cation-exchange capacity helps decrease Cd accumulation in crops (McBride et al., 1981). Near-neutral pH with high organic matter content is reported to limit the availability of TEs in soils (Mitchell et al., 2014). It is worth noting that these soil factors interact for the accumulation of TEs in vegetables (McLaughlin et al., 2011; Ding et al., 2013). An analysis of the relationship between vegetables accumulating TEs and soil factors is needed to comprehensively investigate the influences of TEs on agricultural ecosystems and evaluate the risks posed by contaminated vegetables.

Recent policy developments in the USA and Europe aimed at the protection of human health are moving towards regulations that encourage a risk-based approach for the management of TE-contaminated vegetables (Chen et al., 2009; Novotná et al., 2015). Many quantitative and empirical models have been derived to estimate the potential health risks of vegetable consumption (McBride et al., 1981; Nabulo et al., 2012; Wang et al., 2016). Simple generic parameters are widely used in most models (Novotná et al., 2015). In addition, model development in these studies is often based on spike pollutants or container experiments exhibiting a narrow range of soil factors (Zhang et al., 2011; Ding et al., 2013). A site-specific database from field observation, which critically considers metal type, vegetable cultivars, soil properties, and local dietary habits, would help to identify possible risks more accurately. In this study, we use the Youxian prefecture, southern China, as a case study to evaluate the influence of soil factors on the accumulation of TEs in different vegetable plants. A site-specific prediction model is further developed to assess the risk to residents resulting from consumption of locally produced vegetables.

2. Material and methods

2.1. Study area and sampling

The Youxian prefecture (113.32°E long., 27.01°N lat.) is a major crop production region in southern China (Fig. 1) and is known nationwide as the "Cd-laced rice" area (Wang et al., 2016). Contamination of vegetables by TEs is of great concern but still incompletely understood by local government and residents.

A total of 589 paired soil and vegetable samples were collected throughout the Youxian prefecture (Fig. 1). A detailed description of the sampling strategy was given by Yang et al. (2016). Eight major consumed vegetables including bok choy (*Brassica rape* var. chinensis), flowering cabbage (*Brassica rape* subsp. parachinensis), Chinese cabbage (*Brassica pekinensis.* L.), red cabbage (*Brassica campestris* subsp. *chinensis* var. purpuria), mustard (*Brassica juncea.* L.), radish (*Raphanus sativus.* L.), lettuce (*Lactuca sativa.* L.), and rape (*Brassica campestris* L.) were selected and their sensitivity to TEs in different soil conditions was measured. Radish is a root vegetable while the others are leaf vegetables. Detailed information on vegetable sampling is available in Table SI1.

2.2. Chemical analysis

Soil and vegetable samples were prepared according to the procedure described in Yang et al. (2016). Soil organic matter content, clay content, and cation-exchange capacity are listed in Table SI2. In this study, available TEs in soil samples were extracted with 0.01 M $CaCl_2$ (Nabulo et al., 2011). For As and Hg measurement, samples were digested with Aqua Regia (Lu, 2000). In other cases, vegetable matter was digested with concentrated HNO_3-$HClO_4$, and soil was digested with a HCl-HNO_3-HF-$HClO_4$ mixture (Wang et al., 2016). Arsenic and Hg concentrations were determined using an atomic fluorescence spectrometer (Liu et al., 2006), while Cd, Pb, and Zn concentrations were determined using graphite furnace atomic absorption spectroscopy (Wang et al., 2016).

The accuracy of the analyses was assessed by using standard samples for soil (GSS-5) and vegetables (GSB-5). The recovery of each trace element was in the range of 79.4% to 121.5% for vegetables, and 74.2% to 124.7% for soils, respectively. Duplicates were analyzed on 10% of all samples and the standard deviations (SD) were within ± 5% of the mean.

2.3. Modeling TE uptake by vegetables

Accumulation of TEs in vegetables was modeled using a soil-to-plant transfer function described in Hough et al. (2005) and Nabulo et al. (2011, 2012). This empirical model considers competition for uptake from protons and other available TEs. Taking Cd as an example, the accumulation of Cd in vegetables is a function of available Cd and measured H^+ activity (Eq. (1)).

$$Cd_{veg} = (\alpha_1 \times (CaCl_2\text{-}Cd))/\left(1 + \alpha_2 \times (CaCl_2\text{-}Cd) + \alpha_3 \times \left(H^+\right)\right) \quad (1)$$

where Cd_{veg} is the concentration of Cd in vegetables (mg FW kg^{-1}), $CaCl_2$-Cd is the available Cd concentration in soil (mol L^{-1}), and H^+ is the measured H^+ activity (mol L^{-1}) (Hough et al., 2005). The α_1, α_2, and $\alpha_3$are vegetable-specific parameters.

2.4. Health risk assessment

The risk to human health resulting from consumption of individual vegetables was assessed using a non-carcinogenic metal-specific Hazard Quotient (HQ) (Hough et al., 2004). The HQ was calculated as the ratio of individual average daily dose (ADD, μg BW [body weight] kg^{-1} day^{-1}) to a safe reference dose (Hough et al., 2004; Wang et al., 2016). The Total Hazard Quotient (THQ) for specific-metals is calculated as the arithmetic sum of the HQ values of individual vegetables (Wang et al., 2005; Yang et al., 2011).

Site-specific HQ values were derived for adults (58.0 ± 9.1 kg body weight) consuming local produced vegetables including 155 ± 46.1 g FW d^{-1} bok choy, 126 ± 42.6 g d^{-1} flowering cabbage, 177 ±