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Proper management of lead-contaminated agricultural lands against the exceedance of lead in agricultural produce: Derivation of local soil criteria

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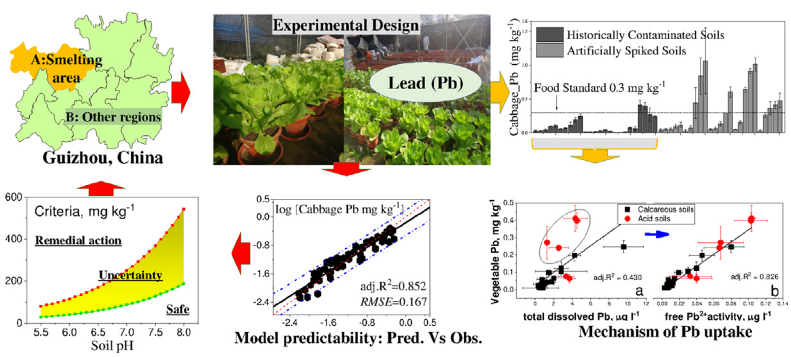
HIGHLIGHTS

- Proper soil management is required by law to lower food contamination risks.
- Derivation of local soil criteria requires reliable Pb soil-crop relationships.
- Pb uptake by cabbage is controlled by free Pb^{2+} ion activity.
- Reliable uptake models are derived using available soil Pb and key soil properties.
- Implementing soil criteria derived can avoid the exceedance of Pb in vegetables.

GRAPHICAL ABSTRACT



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ABSTRACT

The Measures for Management of Soil Environment in Agricultural Land (Trial, Nov. 01, 2017, China) recently came into effect and highlighted the proper management of contaminated croplands to lower risks of exceedances of contaminants, especially toxic trace metals in agricultural produce. We aimed to develop local soil criteria for lead (Pb) in Hezhang county of southwestern China by the inverse use of reliable models linking Pb contamination levels between soils and vegetables. Dilute nitric acid (0.43 M) extraction, a new ISO standard (ISO-17586:2016) for extracting the geochemically reactive Pb fraction (Pb_{NA}), and calcium chloride (0.01 M) extraction (ISO-14255: 1998) for estimating the plant-available Pb (Pb_{CC}) were performed in fifty historically polluted and newly Pb-spiked soils with differing soil types, properties (pH 4.1–8.0), and total soil Pb levels (Pb_T, 20–6153 mg kg^{-1}). Greenhouse experiments for *Brassica pekinensis* L., and in-situ soil porewater measurement for Pb were conducted to investigate the mechanism of Pb uptake, and to establish reliable Pb soil-plant relationships. The results indicated that about 83% of the variation for Pb concentrations in vegetable (Pb_{CL}, 0.009–1.06 mg kg^{-1}) was contributable to free Pb^{2+} activity in soil porewater, which was mainly influenced by pH and dissolved organic matter. Pb_{CL} was satisfactorily predicted using Pb_{NA} and key soil properties (adj. R^2 0.852). Soil Pb criteria for Pb_T and Pb_{NA} are then derived based on food standard. The full implementation of criteria derived for Pb_{NA} (i.e., 27–127 mg kg^{-1}, soil pH 5.5–8.0) can avoid the exceedance of Pb in 95% of cabbage samples in this study, 95% of cabbage cultivars by model extrapolation, and one widely cultivated root vegetable,

Abbreviations: Pb_T, Total soil Pb concentration; Pb_{NA}, Nitric acid (0.43 M) extractable Pb concentration in the soil; Pb_{CC}, Calcium chloride (0.01 M) extractable Pb concentration in the soil; Pb_{CL}, the Pb contamination level in cabbage leaf; Pb_{DTPA}, DTPA (0.005 M) extractable Pb concentration in the soil; SQS, Soil quality standards; FQS, Food quality standards; MMSEAL, Measures for Management of Soil Environment in Agricultural Land.

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radish, in the study region. We provide a successful case study that has effectively tackled the challenge for the complexity of the soil management in contaminated croplands.

1. Introduction

In humans and animals, exposure to lead (Pb) can cause neurological, renal, reproductive, developmental, immune, and cardiovascular health effects (United States Food and Drug Administration, 2015). The primary human exposure to Pb is via the food chain (Agency for Toxic Substances and Disease Registry, 2007) especially since excessive bioaccumulation of Pb is often found in edible portions of crops such as leafy and root vegetables from Pb-contaminated soils (Finster et al., 2004; McBride, 2013; McGrath and Zhao, 2015; McLaughlin et al., 2011). In Hezhang county, Guizhou province of southwestern China, earlier investigations indicated foods of maize and vegetables were commonly grown in Pb-polluted soils (up to 6153 mg Pb kg^{-1}) due to historical Pb-Zn smelting (He et al., 2013; Li et al., 2015; Yang et al., 2011; Zhang et al., 2018a), and Pb exceedance in foods such as maize and vegetables (>0.3 mg Pb kg^{-1}, fresh weight) has been extensively reported (He et al., 2013; Yang et al., 2011). Dietary intake of Pb was the primary contributor to high blood-Pb concentrations (mean 133 μg l^{-1}, n = 451; target blood-Pb level 10–20 μg l^{-1}) found in residents (He et al., 2013; Yang et al., 2011). Therefore, it is crucial to take measures to lower human exposure to Pb via the food chain in this region.

Recently, the Measures for Management of Soil Environment in Agricultural Lands (Trial) (MMSEAL) came into effect on Nov. 1, 2017 (MMSEAL, 2017). The MMSEAL highlighted the proper management of contaminated agricultural lands to lower risks of exceedances of contaminants, especially trace metals such as cadmium (Cd), arsenic (As), and Pb in agricultural produce. A prerequisite to effectively carry out MMSEAL is the accurate identification of those soils that are safe for growing crops, and those that are not. However, because of the high variability of geographical landscapes, soil types, soil physiochemical properties (e.g., pH 4.0–9.0), and soil background values for contaminants between regions in China, a nation-wide soil quality standard (SQS) currently is not able to meet the requirement by the MMSEAL. Therefore, a regional management strategy should be explored, and the development of local soil criteria has been suggested by scholars and policy-makers (Song et al., 2016; Xia, 2013; Xia and Luo, 2007).

In order to develop local soil criteria for identifying soils that would result in the exceedance of Pb in agricultural produce, it is crucial to establish reliable site-specific links between Pb contamination levels in soils and foods (Zhang et al., 2018a; Zhang et al., 2018b). Numerous studies have investigated Pb as well as other trace metals soil-plant relationships. For Pb, higher levels in foods such as vegetables are often found where soil levels are relatively higher (Samsøe-Petersen et al., 2002; Zhang et al., 2018b). Bioconcentration factors (BCFs), with a linear uptake assumption, has been used to predict crop contamination levels. Despite some successful attempts (Samsøe-Petersen et al., 2002), results overall were not consistent especially for Pb (Ding et al., 2016; Samsøe-Petersen et al., 2002; Zhang et al., 2018b).

Currently, knowledge of factors controlling Pb uptake into food crops from soils, whether from soils or plants, is inadequate to reliably predict Pb contamination levels in crops (Clemens and Ma, 2016; McGrath and Zhao, 2015; Sharma and Dubey, 2005; Zhao et al., 2004). Specifically, the total Pb levels in food crops and soils are poorly correlated under field conditions (Legind and Trapp, 2010; McBride, 2013; Rooney et al., 2007). This weak correlation is partially due to the insufficient consideration of the (bio)availability of Pb in soils, which is strongly influenced by some key soil properties such as pH and soil organic matter content (McBride et al., 2014; Rieuwerts et al., 1998; Sauvé et al., 1997). For instance, a lower soil pH (e.g., <6) can increase the total dissolved Pb as well as free Pb^{2+} activity, further denoted as aPb^{2+}, in soil porewater thereby increasing the Pb uptake in plants (Rooney et al., 2007; Zhang et al., 2018b). Some studies satisfactorily predicted Pb uptake by root vegetables after accounting for soil pH and other key properties under greenhouse experiments using newly Pb-spiked soils (Ding et al., 2016). However, no such successful site-specific predictions were found. Pb concentrations in soil porewater or the exchangeable fraction in soils from fields can be much lower, and other factors such as aerial Pb-to-plant uptake and the surface-related contamination can significantly mask the real Pb soil-plant relationships (Douay et al., 2008; Hu and Ding, 2009; Legind and Trapp, 2010; McBride et al., 2014; McBride et al., 2013).

Besides the absence of reliable predictions of crop Pb-contamination levels, other factors would hinder the implementation of local criteria worth considering accordingly. Ideally, local soil criteria should be crop type-dependent because of the significant variability for bioaccumulation of Pb between crop types. However, policy-makers and farmers typically prefer to choose a conservative criterion to protect most local crops. For instance, soil criterion for Pb derived from leafy vegetables would be also to protect other crop types such as fruit and root vegetables. In those contaminated soils which would result in Pb exceedance in leafy vegetables, fruit or root vegetables can be alternately cultivated. This is in accordance with MMSEAL preferential use measures of agronomic regulation to reduce the percentage of exceedance of contaminants in foods.

Based on a comprehensive field investigation on soil/crop types and Pb contamination status in historical Pb-Zn smelting areas of Hezhang county, as well as other unpolluted areas in Guizhou province of southwestern China, this study is designed to derive local soil criteria for Pb, total content or bioavailability-based, by carefully examining the process by which a leafy vegetable, Chinese cabbage, became contaminated from both historically contaminated and Pb-spiked soils with a wide range of soil types and properties. The mechanism understanding for Pb uptake and the methodology for deriving Pb criteria, together with uncertainty analysis, were provided. The specific objectives are to (1) assess the Pb availability using the ISO standard procedures (2) determine the factors that control Pb availability in 25 historically contaminated (HC) and 25 artificially Pb-spiked (AS) agricultural soils, and (3) derive reliable soil-plant transfer models for further derivation of soil criteria for Pb regarding vegetable safety for human consumption.

2. Materials and methods

2.1. Field investigation and soil sampling

Both literature and on-site field investigation were performed to determine the soil sampling method, and to ensure the representativeness of collected soils covering main soil types in Guizhou province of Southwestern China. The main soil types in the historically Pb-contaminated area, Hezhang county (104°10′–105°01′ E, 26°46′–27°28′ N), as well as other regions across Guizhou were investigated. A grid sampling method (e.g., Supporting information Fig. A.1) was adopted in typical agricultural fields. All of the soils collected from one particular field, if total soil Pb varied insignificantly, were thoroughly mixed into one typical soil in the sampling site. A portable X-ray fluorescence spectrometer (XRF) was used to ensure the high variability of Pb-contamination levels in collected soils. Overall, a total of 25 historically contaminated and 5 unpolluted topsoils to a depth of 20 cm were collected.