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Derivation of soil thresholds for lead applying species sensitivity distribution: A case study for root vegetables

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HIGHLIGHTS

- Normalization relationship was developed and validated to be reliable.
- The $HC5_{add}$ values were calculated from the Burr Type III function fitted SSD curves.
- Soil thresholds based on added Pb depend on the combination of soil pH and CEC.

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ABSTRACT

The combination of food quality standard and soil–plant transfer models can be used to derive critical limits of heavy metals for agricultural soils. In this paper, a robust methodology is presented, taking the variations of plant species and cultivars and soil properties into account to derive soil thresholds for lead (Pb) applying species sensitivity distribution (SSD). Three species of root vegetables (four cultivars each for radish, carrot, and potato) were selected to investigate their sensitivity differences for accumulating Pb through greenhouse experiment. Empirical soil–plant transfer model was developed from carrot New Kuroda grown in twenty-one soils covering a wide variation in physicochemical properties and was used to normalize the bioaccumulation data of non-model cultivars. The relationship was then validated to be reliable and would not cause over-protection using data from field experimental sites and published independent studies. The added hazardous concentration for protecting 95% of the cultivars not exceeding the food quality standard ($HC5_{add}$) were then calculated from the Burr Type III function fitted SSD curves. The derived soil Pb thresholds based on the added risk approach (total soil concentration subtracting the natural background part) were presented as continuous or scenario criteria depending on the combination of soil pH and CEC.

1. Introduction

The generally preferred approach to derive soil quality standard (SQS) is to use species sensitivity distribution (SSD) [1]. A significant purpose of SSD modeling is to determine a concentration protective of most species in the ecosystem, usually the 95% protection level, known as the HC5 [2]. Currently, the SSD method is mainly used in the toxicity risk assessment and the development of ecological risk thresholds for aquatic and terrestrial flora and fauna [3–5]. However, it is rarely applied in the derivation of soil thresholds for heavy metals in view of food safety.

Many early proposed SQSs (using total heavy metal concentrations in soil) were found to be less than the natural background concentrations (NBC) in soil because of the large range of NBC and because of the desire to set SQSs based on the SSD method [6]. To overcome this problem, the added risk approach was developed [3]. This approach is based on the idea that the background concentration of a naturally occurring substance should pose no risk to the environment, and therefore could be better applied to soils with high NBC than the total approach. However, most SQSs across the world are still based on the total metal approach [7].

In many countries including China, the current SQSs used for agriculture are estimated from the literature or "imported" from other countries without considering the influence of soil properties [8], and therefore could not completely ensure the production of crops that meet the food quality standard [9,10]. There is an urgent need to revise and improve the current SQS for heavy metals.

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Establishment of empirical soil–plant transfer model is one of the key steps for improving SQS [11]. Models describing the transfer of Cd from soil to plants have been developed, but those for Pb appear to be unreliable [12]. The poor relationship between vegetable and soil Pb concentrations is attributable to particulate contamination of vegetables and soil properties that influence phytoavailability [13]. However, even after accounting for pH and other soil properties, predictions of Pb uptake by leafy vegetables or cereal grains from soils have been generally unsatisfactory [14,15].

Many earlier studies may have been compromised by aerial contamination that obscured impacts of soil Pb on plant concentrations [13]. However, this is not the case for root vegetables. The transfer of airborne Pb into the underground edible parts of root vegetables was negligible, and the accumulated Pb mainly originated from the soil [16,17], highlighting the predominance of root vegetables over leafy vegetables or cereal grains in investigating the Pb thresholds in soils.

Therefore, with a focus on widely consumed root vegetables, this study aims to derive soil thresholds for Pb based on the food quality standard using SSD method, while taking into account the soil properties that would have the greatest influence on the phytoavailability.

2. Procedure and methods

2.1. Experimental design for greenhouse and field trials

A total of twenty-one soils covering a wide variation in soil properties were collected throughout China (Table. S1). A detailed description of the sampling strategy was given by Ding et al. [18]. Firstly, two typical soils, an acidic Ferrolsols (pH 4.84) and a neutral Cambosols (pH 6.93), were used to test the sensitivity variations of different cultivars for accumulating soil Pb in a greenhouse study in 2010. Three species of root vegetables, radish (*Raphanus sativus* L.), carrot (*Daucus carota* L.), and potato (*Solanum tuberosum* L.) were used in the experiment; four representative cultivars of each species were selected. Then, the soil–plant transfer model was developed from carrot cultivar New Kuroda grown in the twenty-one soils in another greenhouse study in 2011. The followed field study was conducted at three sites through China (Yingtan, Jiangxi Province; Nanjing, Jiangsu Province; and Haikou, Hainan Province) in the year 2012 to validate the developed prediction model. The used three soil types and planted cultivar (carrot New Kuroda) were in accordance with the greenhouse study in 2011.

Uncontaminated study soils were spiked with soluble lead salt to minimize the effect of varying sources of Pb on phytoavailability. Specifically, lead nitrate was used in the present study to avoid solubility constraints and related kinetic limitations to dissolution, as has been widely applied in previous studies [19,20]. Based on the current Chinese SQS [21], three Pb addition treatments were used in both greenhouse and field studies: the control (no exogenous Pb added to soil), low-Pb (half of the SQS, i.e. 125, 150, and 175 mg kg^{-1} for soils with pH < 6.5, 6.5–7.5, and >7.5, respectively), and high-Pb (equal to the SQS, i.e. 250, 300, and 350 mg kg^{-1} for soils with pH < 6.5, 6.5–7.5, and >7.5, respectively). After aging for three months, vegetable plants were cultured under regular farming management style. Three replicates were tested per treatment.

For more details about the greenhouse and field studies, it can be referred to Ding et al. [18,22,23].

2.2. Normalization of the bioaccumulation data

Generally, the bioaccumulation data have been generated from different soils. Therefore, the variation in the data will reflect both the inherent species sensitivity and the soil properties. By normalizing the data to a standard soil using empirical soil–plant transfer models, the effect of soil properties on the bioaccumulation data will be minimized and the resulting distribution of the data will reflect more closely the inherent sensitivity of the studied species [24].

The normalization relationship in the present study is the empirical model between bioaccumulation data for Pb to a cultivar (carrot New Kuroda) and the basic soil properties:

$$\log[\mathrm{BCF}] = a \times \mathrm{pH} + b \times \log[\mathrm{CEC\ or\ OC\ or\ clay}] + k \quad (1)$$

where BCF is bioconcentration factor (ratio of Pb concentration in plant to that in soil), a and b are slops of soil properties, the intercept k is the intrinsic sensitivity that characterizes the ability of the cultivar to accumulate soil Pb.

The basis for cross-species extrapolation is the assumption that the slopes of parameters (a and b) which indicate the degree of influence of soil properties on metal accumulation in plants are constant across species, and that only intrinsic sensitivity (k) varies [4,25]. Under the condition of the squared error with the lowest value between the predicted and measured BCF values calculated as $\sum_{i=1}^{n}(\text{measured BCF}_i - \text{predicted BCF}_i)^2$, the intercept ($k$) for different cultivars were obtained through Excel Solver for linear optimization [19]. The accuracy of the predictive models for different cultivars was evaluated by comparing the measured BCF with the predicted BCF from each model.

The intra-species variability before and after the normalization process was calculated as $(\sqrt{\sum(\mathrm{BCFsi} - \overline{\mathrm{BCFs}})^2/n-1})/\mathrm{BCFs}$, where BCFsi is the ith bioconcentration factor normalized to a certain soil condition, $\overline{\mathrm{BCFs}}$ is the average of n BCFs, n is the number of BCFs for the cultivar. The reduction of intra-species variability indicates that the normalization process eliminated the influence of soil properties to some extent.

2.3. SSD construction and HC5 derivation

In this study, Burr Type III distributions which was incorporated into the BurrliOZ program (the software is freely available at https://research.csiro.au/software/burrlioz/#) were used to fit the SSD curves. The Burr III function is a very flexible 3-parameter distribution, which can provide good approximations to many other commonly used distributions such as Log-normal, Log-triangular, and Weibull. The equation for Burr Type III function is as follows [26]:

$$y = \frac{1}{\left[1 + \left(\frac{b}{x}\right)^c\right]^k} \quad (2)$$

where b, c, and k are three parameters. When k and c tend to infinity, Burr Type III distribution may change to ReWeibull and RePareto distribution respectively.

The critical soil concentration for each cultivar under different soil conditions after normalization was back calculated from the corresponding BCF value and the 0.3 mg kg^{-1} Chinese food quality standard for Pb in fresh root vegetables [27]. Then the hazardous concentration in soil for protecting 95% of the cultivars (HC5) at its median estimate (or at its 50% confidence level) was calculated from the BurrliOZ software (Fig. S1).