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Prediction of Cadmium Threshold Using Regression Model for Soils Under Cd-Containing Organic Residue Application

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Abstract Organic residue is a good resource of organic matter and nutrients to improve soil physico-chemical properties, but the toxic trace elements, including cadmium (Cd), borne with it may limit its suitability for arable land. This study aimed to develop regression models for Cd critical concentration in the soils amended with Cd-containing organic residue in view of food safety. First, paired data of Cd concentrations in cereal grains and soils from the peer-reviewed literature were collected and screened. Then the plant–soil models between the $\log_{10}$-transformed values of Cd concentrations both in grains and the soils based on soil properties were regressed. The plant–soil model predicted the Cd concentration in the grain (Cd_{grain}) based on Cd concentration in soil (Cd_{soil}) and soil properties well with the values of R^2 ranging from 0.856~0.946. Cd_{soil} and soil pH were the major controlling factors for Cd_{grain} and explained more than 85.6 percent of variation in Cd_{grain}. At last, the critical concentration of Cd based on soil property was then back-calculated according to the food safety standard which was presented as continuous or pH-range criteria for Cd-containing organic residue amended soil. With the models derived in this study, a species-specific Cd threshold can be calculated based on soil pH and a default limit value of Cd in grain for food safety. The species-specific Cd threshold will be conducive to scientific application and management of Cd-containing organic residue in agriculture.

Keywords Cadmium · Critical concentration · Organic residue · Regression model

1 Introduction

Organic residue include animal manure, food processing waste, crop residue, municipal biosolids, waste from some industries and the products processed from them. Animal manure and food processing organic by-product have been applied to land for improving soil fertility, soil physico-chemical properties and crop productivity for a long time. Municipal biosolids have also been utilized for the same purpose (Risse et al., 2001). Land application of organic residue is a beneficial way to recycle organic matter and nutrients, e.g., nitrogen and phosphorus, and reduce the waste volume for disposal at the same

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time (Embrandiri et al., 2012). Agricultural land application of organic residue has been increasing in recent years following the ban on incineration and landfill disposal due to environmental risk associated with these processes. Biosolids have been applied in the agricultural areas which involves nearly all the main food crops and vegetables in China (Cao & Ikeda, 2010). The percentage of the biosolids used for land reached up to 57% and 60% in Belgium and France, respectively (Maisonnave et al., 2002). The increasing use of biosolids has also been reported in the UK (Gove et al., 2002).

Besides all the benefits and values of land application, the concerns related to adverse environmental and food chain effects of trace elements in organic residue have a history as long as the history of applying organic residue on land (Granato et al., 2004; Li et al., 2000). Several studies demonstrated the increase of inorganic contaminants like copper (Cu), zinc (Zn), lead (Pb) or cadmium (Cd) in horizons receiving organic amendments issued from urban wastes, even under conditions that conform to relevant regulations (Baldantoni et al., 2010; Cambier et al., 2014).

The risk of Cd contamination in arable land caused by organic residue application has always been attracted extensive attraction since Cd is a non-essential trace element, and does not play any identified role in the growth and development of human and organism (Khan et al., 2017). Also, Cd is more efficient in translocating from soil to plant than other trace metals frequently concerned in the environment due to its high mobility within the environment and bioavailability in the soil–plant system (Chang et al., 2014; Shahid et al., 2016). The primary route for Cd exposure in non-smoking population is food, through ingestion of contaminated grains, vegetables, fruits and their products (Brus et al., 2009; Norvell et al., 2000). Dietary exposure to Cd constitutes the breast, ovarian, endometrial or prostate, cancer risk factor (Adams et al., 2014; Lin et al., 2016). Aiming for food safety, maximum permitted levels of toxic trace metals (including Cd) in foodstuffs had been strictly defined by many regulatory agencies, e.g., the World Health Organization (WHO) and the Food and Agriculture Organization (FAO).

The critical soil trace metal concentration in view of human health can be back-calculated from maximum permitted levels of trace metal in plants and the ratio of its concentration in plant to that in soil, namely bioconcentration factor (BCF) from soil to plant. However, various studies showed that there is no such a fixed linear relationship for most metals. Moreover, the total soil metal concentration is a poor indicator of metal availability in soil and not the only factor affecting metal accumulation in plants (Simmons et al., 2008). Soil physico-chemical properties including soil pH, organic carbon (OC), cation exchange capacity (CEC) and clay contents affect metal bioavailability in soil significantly and then accumulation in plants (Shahid et al., 2016). The simple regression-based soil–plant models, taking into account the effect of soil properties on Cd bioavailability, describe the relationship between the Cd concentration in soil and uptake by plants (Brus et al., 2009; Koopmans et al., 2008; Simmons et al., 2008). The models are normally represented with a nonlinear quantitative relationship in the form as following,

$\log [Cd]_{plant} = a + b\ pH + c \log [Clay] + d \log [OC] + e \log [Cd]_{soil}$,

where.

$[Cd]_{plant}$ and $[Cd]_{soil}$ are Cd contents in plant and soil, respectively.

pH is soil pH value.

$[OC] =$ organic carbon content in the soil (%).

$[Clay] =$ clay content in the soil (%).

Values for the various coefficients (a, b, c, d, e) were obtained by multiple regression analysis (Adams et al., 2004; Brus et al., 2009). Some human health risk-based Cd concentrations for Cd-spiked soils calculated with the predictive models using one particular species as reference have been reported (Ding et al., 2013; Melo et al., 2012). However, metal salts added into soils with organic amendments are less phytoavailable than metal salts spiked directly into the soil (Benjamin et al., 2016). The Cd in organic residue is bound in solid phase with different forms, and the sum of all chemical forms of Cd is rarely 100% bioavailable (Udovic & McBride, 2012). Moreover, Cd transfers from roots to aerial tissues via transpiration-driven xylem loading, with apoplastic and/or symplastic transport (Hasan et al., 2009; Kranner & Colville, 2011) of free Cd^{2+}, or Cd complexed with various chelates (Lux et al., 2011; Maestri et al., 2010). The differences between Cd contamination sources need to be considered when dealing with Cd accumulation in cereal grains.

Therefore, in this study, paired data of Cd content in grain and Cd-containing organic residue-amended soils were collected (Table S1 in supplementary information). This research aimed to 1) derive species-specific Cd soil–plant models for different crops, 2) develop empirical prediction models for Cd threshold in view of food safety for arable land soils amended with Cd-containing organic residue, 3) apply the obtained empirical prediction models into the management of organic residue agricultural application.

2 Procedure and methods

2.1 Data collection and screening

The concentrations of Cd in the grains of cereal plants grown in organic residue, e.g., biosolids, manure and other organic amendments amended soils, were used in this study. The data were retrieved from all the available peer reviewed literature, in which Cd contamination resulted from organic residue. All the data obtained were screened first with the availability of Cd accumulation in grains and Cd concentrations in soils or the possibility of transforming them from the original data reported in the literature. The methodology of the experiments (either greenhouse or field) and the pattern of results described should be reliable, e.g., it was confirmed that the data of Cd concentrations in the plants were based on dry or fresh weight, the Cd concentrations in soils were based on total content. At last, the soil property parameters (pH, OC, CEC and Clay) should be available from the literature or can be obtained from other referred literatures. The screened data contained Cd data for 5 species includes rice, maize, bread wheat, durum wheat and barley. Considered the small quantity of data for durum wheat and barley and the biological similarity of them with bread wheat, the data for bread wheat, durum wheat and barley was combined into a set of data, named data for wheat in this study. The screened dataset of Cd accumulation in grains used here includes 47 for wheat, 10 for rice and 24 for maize, respectively (Table S1).

2.2 Soil–plant models regression

In view of that each soil property parameter e.g., pH, OC or OM (organic matter) content, CEC and Clay content reported in the literature was determined with different methods. Soil pH used in this study was set as the value measured with the method of soil/water ratio of 1:5. The pH value determined with other methods was corrected to pH value (1:5 H_2O) with quantitative relationship (Wang, 2012). Also, the soil organic matter content was corrected to the soil organic carbon content (%) with quantitative relationship (European Union (EU) 2006), too. Then the Cd concentrations in cereal grains and soils, soil property parameters (except pH) were $\log_{10}$-transformed first. The $\log_{10}$-transformed Cd concentrations in cereal grains were related to $\log_{10}$-transformed Cd concentrations in soils and soil property parameters with multiple regressions (SPSS 19.0). Soil cation exchange capacity (CEC) is well correlated with pH, OC and clay (van Erp et al., 2001). CEC was not included as an independent variable in the regression analysis to avoid issues of collinearity (Pérez & Anderson, 2009). To decide whether the plant–soil model can be used in this approach, the predicted Cd concentrations in wheat and maize grains calculated with the regression models were compared with the measured Cd concentrations to evaluate the prediction accuracy for combination of plant–soil model.

2.3 Derivation of critical soil Cd concentration from food quality criteria for cereal

The link between critical metal concentration for soil and those for plant mainly involves food quality in view of human health, fodder quality in view of animal health and phytotoxic level in view of negative effect on the plant itself. As for Cd, phytotoxic levels in food crops are much higher than food and fodder quality limits. The critical Cd concentrations in soil back-calculated from food quality criteria are much more stringent than those from phytotoxic levels for plants. With the regression soil–plant models obtained as showed above, the critical Cd concentration in soil with specific soil property condition referred to a particular cereal species was calculated. In view of some data of Cd concentration from the literature was given as fresh weight, the moisture percentage of 85% was applied to grain of the crops to calculate dry weight-basis criteria (Vries et al., 2007).

3 Results and Discussion

3.1 Regression plant–soil models

Linear regressions between the $\log_{10}$-transformed values of Cd concentrations in grains and soils and the corresponding soil parameters were carried out for the screened data. The coefficients of the plant–soil models are shown in Table 1. The plant–soil model predicted the Cd concentration in the grain (Cd_{grain}) with Cd concentration in soil (Cd_{soil}) and soil pH well with the values of R^2 ranging from 0.856 ~ 0.946. Inclusion of organic carbon content (OC) into the soil–plant models improves the correlation performance slightly with R^2 from 0.856 and 0.891 to 0.878 and 0.923, respectively, for wheat and maize. The partial correlation coefficients (r) of the three factors indicated that Cd_{soil} was the major factor controlling Cd_{grain}. Soil pH was negatively related with Cd_{grain} while Cd_{soil} and OC was positively related with it. Among soil characteristics, soil pH is usually considered the most important in controlling the uptake of Cd by plants (Mcbride, 2002; Peijnenburg et al., 2000) and normally the Cd concentrations in plants decreased with pH increased. As for rice, pH has a greater r value than the $\text{LogCd}_{\text{soil}}$ or LogOC, which means pH plays a key role ($r = -0.991$) in the Cd uptake for rice grain. Bingham et al. (1980) stated earlier that Cd_{grain} of rice was highly dependent upon the soil pH. Several findings also have shown that the Cd concentration in the rice grown in Cd contaminated soil decreased when added amendment increased soil pH (Tariq et al., 2014; Yong et al., 2011). Moreover, the strong effect of pH on the Cd uptake in the rice grain may be due to strong correlation of soil pH with redox potential in paddy soil, another important soil property affecting Cd accumulation in rice grain (Liu et al., 2013).

For multiple linear regressions, values of R^2 can be used to explain variation of the dependents (Wang et al., 2004). Table 1 shows that more than 85.6 percent of variation in Cd_{grain} can be attributed to both Cd_{soil} and soil pH. Soil pH has been reported to play the most important role in determining Cd phytoavailability in soil in numerous studies (Muhammad et al., 2012; Yu et al., 2016; Zeng et al., 2011). Zhuang et al. (2021) developed the multiple linear regression models for the relationships between soil properties, Cd content in soil and grain Cd level. Those models showed that soil pH exhibits strong effect on Cd transfer from soil to grain with the coefficients of pH from -0.49 to 0.17, -0.58 to -0.08 for wheat and maize, respectively. While the models developed in this study showed weaker effect of soil pH on Cd transfer from soil to grain. The discrepancy of soil pH coefficients for the models maybe caused by difference of data source. Only the data originated from organic residue amended soil were used in the present study and the Cd are less phytoavailable than that in Cd-spiked soils (Benjamin et al., 2016).

Table 1 The regression plant–soil models

Species	Regression models	Coefficient of determination (R^2)	Partial correlation coefficient (r)		
			$\text{LogCd}_{\text{soil}}$	pH	LogOC
Wheat	$\text{Log}_{10}\text{Cd}_{\text{grain}} = 0.002\text{pH} + 0.059\text{Log}_{10}\text{OC} + 0.952\text{Log}_{10}\text{Cd}_{\text{soil}} - 0.946$	0.878 (n = 47)	0.638	-0.497	0.423
	$\text{Log}_{10}\text{Cd}_{\text{grain}} = -0.008\text{pH} + 0.968\text{Log}_{10}\text{Cd}_{\text{soil}} - 0.854$	0.870 (n = 47)			
Rice	$\text{Log}_{10}\text{Cd}_{\text{grain}} = -0.630\text{pH} - 5.536\text{Log}_{10}\text{OC} + 1.511\text{Log}_{10}\text{Cd}_{\text{soil}} + 2.540$	0.946 (n = 10)	0.942	-0.991	0.500
	$\text{Log}_{10}\text{Cd}_{\text{grain}} = -0.478\text{pH} + 1.511\text{Log}_{10}\text{Cd}_{\text{soil}} + 2.223$	0.946 (n = 10)			
Maize	$\text{Log}_{10}\text{Cd}_{\text{grain}} = -0.047\text{pH} + 0.654\text{Log}_{10}\text{OC} + 0.480\text{Log}_{10}\text{Cd}_{\text{soil}} - 1.181$	0.923 (n = 24)	0.770	-0.230	0.481
	$\text{Log}_{10}\text{Cd}_{\text{grain}} = -0.075\text{pH} + 0.518\text{Log}_{10}\text{Cd}_{\text{soil}} - 0.995$	0.891 (n = 24)			

Cd_{grain} = Total Cd concentration in the grain, Cd_{soil} = Total Cd concentration in soil, pH = Soil pH (1:5 H_2O), OC = Organic carbon content (%).

Also for the practicability of the regression models, the regression soil–plant models with two variables Cd_{soil} and soil pH were employed in deriving Cd critical concentration in soil here.

3.2 Validation of the soil–plant model

For the reliability of regression soil–plant models verifying, the data used for regression soil–plant models validation should not be included in the dataset for models development. So, the data for Cd concentrations in both crop grains and soils obtained from a five-year field experiments were used here. In the field experiment, the biosolids were applied into soil with a wheat–maize rotation system from the year of 2007 to 2011 continuously. The concentrations of eight trace elements (including Cd) both in soils and crop grains were analyzed every year (Li, 2012). The predicted Cd concentrations in wheat and maize grain were calculated with the models for wheat and maize shown in Table 1. Then the predicted Cd concentrations in wheat and maize grains were compared with the measured Cd concentrations for the two crops. The results are shown in Fig. 1 with X and Y axes means measured and predicted Cd concentrations in grain, respectively: for wheat, 38 of all the 40 data points fell above the 1:1 line with predicted Cd concentrations higher than the measured ones. For maize, 30 of all the 32 data points fell above the 1:1 line. Most of the predicted Cd concentrations were far higher than the measured ones, that maybe because the soil in the field experiment was calcareous soil with pH 8.9. Normally, the Cd has lower availability in alkaline soil than in acidic soil. The results demonstrated that the predicted Cd concentration in cereal grain is probably higher than the realistic Cd concentration in grain, and then the soil critical concentration of Cd back-calculated from the regression soil–plant models will be conservative, especially in the soil with higher pH.

3.3 Derivation of Cd critical concentration in soil amended with organic residue for cereal crops

Soil critical concentration for Cd can be back-calculated from the regression plant–soil models in Table 1. The derived calculation formula based on Cd concentrations in grains and soil properties is shown in Table 2. Scenario critical value could also be calculated for different combinations of soil pH and OC content as shown in Table 2. The values of Cd critical concentrations ($Cd_{critical}$) ranging from 1.7 to 3.2 mg/kg, 0.3 ~ 4.0 mg/kg and 4.4 ~ 14.1 mg/kg in soils for wheat, rice and maize, respectively, with pH range 4.5 ~ 8.0. The Cd_{soil} for wheat and maize were similar and depended on soil pH slightly, while the $Cd_{critical}$ for rice rose from 0.3 mg/kg for acidic soil to 4.0 mg/kg for alkaline soil. It is generally considered that decrease in soil pH would promote Cd mobility in the soil, then Cd.

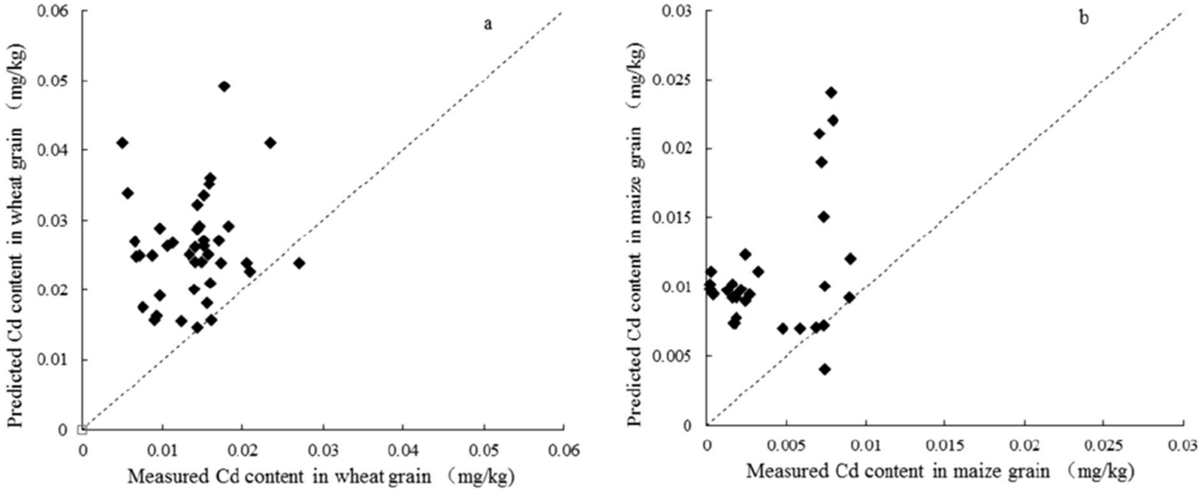

Fig. 1 Measured versus predicted concentrations of Cd in grains for wheat and maize. (The dotted line is 1:1 line)

accumulation in plant significantly at even lower Cd concentration in soil with extreme low pH, but it also has been reported that the Cd accumulation in plants showed no further increase with soil pH decrease when soil pH $<$ 5.5 due to the coexistence of other metals in the soil. With soil pH decrease, the activities of those competitive ions such as Zn^{2+} would increase and compete with Cd^{2+} for the binding site on plant roots (Ueno et al., 2004; Wu et al., 2018). Muhammad et al. (2014) studied the relationship between Cd accumulation in rice grains with soil properties based on pot experiments with Cd-spiked soil and obtained the critical Cd_{soil} value of 0.77 mg/kg and 0.32 mg/kg for Mollisols and Udic Ferrisols, respectively. The risk screening and intervention values for Cd were provided in the soil environmental quality risk control standard for soil contamination of agricultural land in China (GB15618—2018, 2018) (Table 3). The soil with contaminants below the risk screening levels provides sufficient protection for food safety and ecological quality, while the soil with contaminants reach to risk intervention levels will lead to below-standard food and needed to be stringently regulated. The $Cd_{critical}$ derived with regression models in this study (Table 4) for rice were higher than the risk screening levels for paddy land from pH 5.0 to 8.0 but lower than the risk screening levels for paddy land at pH $\leq$ 7.5. The $Cd_{critical}$ derived here for wheat and rice were even higher than the risk intervention levels except in the soil with pH $>$ 7.5. Most of the soil critical Cd concentrations reported in the current literature (Ding et al., 2013; Melo et.al, 2012; Tariq et al., 2014) were derived with data originated from pot experiments based on soil with spiked Cd salt. It is well known that the phytoavailability of Cd added into soils in the form of soluble salts is much

Table 2 Continuous models for soil critical concentration of Cd in view of food quality

Crop species	Models
Wheat	$Log_{10}Cd_{critical} = (0.08pH + Log_{10}Cd_{grain}{}^{*} + 0.854)\ /0.968$
Rice	$Log_{10}Cd_{critical} = (0.478pH + Log_{10}Cd_{grain}–2.223)\ /1.511$
Maize	$Log_{10}Cd_{critical} = (0.075pH + Log_{10}Cd_{grain} + 0.995)\ /0.518$

* The Cd_{grain} was set equal to the limit value of Cd for food quality, the values were referred from GB2762—2017 as 0.1 mg/kg for wheat and maize and 0.2 mg/kg for rice.

Table 3 Soil environmental quality—risk control standard for soil contamination of agricultural land (GB15618-GB15618 2018) (mg/kg)

Limit values	$pH \leq 5.5$		$5.5 < pH \leq 6.5$		$6.5 < pH \leq 7.5$		$pH > 7.5$	
Risk screening	Paddy land	Other	Paddy land	Other	Paddy land	Other	Paddy land	Other
	0.3	0.3	0.4	0.3	0.6	0.3	0.8	0.6
Risk intervention	1.5		2.0		3.0		4.0	

Table 4 Calculated critical Cd concentrations in soil amended with organic residue in view of the food quality criterion as a function of soil properties (all values are given on the basis of dry weight, mg/kg)

Crop	Critical Cd concentration in soil								Food quality criteria*
	pH 4.5	pH 5.0	pH 5.5	pH 6.0	pH 6.5	pH 7.0	pH 7.5	pH 8.0	
Wheat	1.7	1.8	2.0	2.2	2.4	2.7	2.9	3.2	0.1
Rice	0.3	0.4	0.6	0.9	1.3	1.9	2.7	4.0	0.2
Maize	4.4	5.2	6.1	7.2	8.5	10.1	11.9	14.1	0.1

* Adopted from GB2762 2017

greater than that from other common sources of Cd contamination, e.g., biosolids, manure, compost (Rashid et al., 2018; Shuman et al., 2002). Moreover, the increase of organic matter in the soil added with organic residue will reduce the Cd availability in most cases (Ciadamidaro et al., 2017; Filipović et al., 2018). Based on the data originated from Cd-containing organic amended soil, the crop-specific critical Cd concentration ($Cd_{critical}$) derived here provides relevant improvements for the general Cd limit values for soil amended with Cd-containing organic residue. However, it should be cautious to apply the regressed soil–plant models and $Cd_{critical}$ predicted models into Cd-contaminated soil with other source except organic residue or multi-source, since the data originated from soils with other Cd contamination sources were not included here to derive those models.

3.4 Application of critical concentration in soil for organic residue agricultural application

Many countries and jurisdictions have regulated limit concentrations of contaminants (including Cd) or their equivalent for organic residue (e.g., biosolids) land application to protect human health and environment. USEPA (U.S. Environmental Protection Agency) proposed ceiling concentration of 39 mg/kg and annual pollutant loading rate for Cd in the Part 503 of 40 Code of Federal Regulations (https://www.epa.gov/sites/production/files/2018-11/documents/land-application-sewage-sludge.pdf). EU (European Union) proposed the limit value of Cd for safe biosolids agricultural application: 0.5 mg/kg for soil pH of 5 to 6, 1.0 mg/kg for soil pH of 6 to 7, and 1.5 mg/kg for soil pH of 7 (CEC, 2000). The control standard of contaminant for biosolid agricultural application (GB4284-2018, 2018) in China sets 3 mg/kg as the limit value of Cd for arable land application. With the $Cd_{critical}$ derived as above, the application rate of Cd-containing organic residue can be calculated with the $Cd_{critical}$, Cd concentration in the targeted soil before Cd-containing organic residue application, the soil bulk density, crop intersection and mixing depth of soil as follows:

$$\text{Appl} \times \text{Year} = \frac{\left(Cd_{critical} - Cd_b\right) \times D \times BD}{Cd_{residue}(1-k)} \tag{1}$$

where

Appl is the application rate of Cd-containing organic residue on agricultural land for one application per year (kg DW/m^2·a).

$Cd_{critical}$ is the critical concentration of Cd related with soil pH and crop species (mg/kg).

Cd_b is the concentration of Cd in the targeted soil before Cd-containing organic residue applied (mg/kg).

$Cd_{residue}$ is the concentration of Cd in Cd-containing organic residue applied (mg/kg).

k is the fraction intercepted by the crop (%).

D is the mixing depth (m).

BD is the soil bulk density (kg/m^3).

The loss of Cd caused by leaching was not taken into account in Eq. 1, even the crop interception of Cd can be ignored in practical application to get sufficient protection. For large-scale cereal production area, the cropping system is constant normally, the lower crop-specific $Cd_{critical}$ can be chosen to regulate the Cd-containing organic residue land application. The simple predictive models derived in this study will be useful not only for predicting the risk that the Cd concentrations in grains from the plant grown on the Cd-containing organic residue amended soils exceed the regulatory limit, but also for more accurate risk assessment of Cd-containing organic residue amended soils. Using organic residue as a source of organic matter and nutrients is a cost-efficient way to improve soil physico-chemical properties and reduce the need for chemical fertilizers. The $Cd_{critical}$ will be helpful for some authorities to issue a more detailed regulation for Cd-containing organic residue land application.

4 Conclusions

The plant–soil models between the Cd concentrations in grains (Cd_{grain}) and the soils (Cd_{soil}) amended with Cd-containing organic residue based on soil properties were regressed. Cd_{soil} and soil pH were the major controlling factors for Cd_{grain} and explained more than 85.6 percent of variation in Cd_{grain}. The critical Cd concentrations for three cereal crops, in view of food safety, were derived based on the regressed model with two factors Cd_{soil} and pH. The continuous or scenario crop-specific Cd critical model will lead to a more accurate risk assessment of soil amended with Cd-containing organic residue and more precise regulation for organic residue land application.

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Data Availability All data generated or analyzed during this study are included in this article.

Declarations

Conflict of Interest The authors declare no conflict of interest.

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