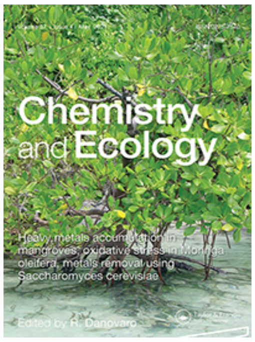

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Influencing factors on transfer and bioaccumulation of soil cadmium and assessment of its environmental risk in a typical karst area, China

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ABSTRACT

Cadmium has become the foremost inorganic pollutant in farmland in China. It is widely accumulated in karst areas in southwest China, especially in Guangxi Province. In this study, vertical soil profiles, rice grains and corresponding root-soil samples were collected in Guangxi. The Cd contents of soil and rice grain samples and several soil properties, including organic matter, Fe_2O_3, Al_2O_3, CaO, MgO, SiO_2 contents and pH were tested. Meanwhile, sequential extraction procedure were used to analysis the chemical fractions of Cd in root-soil samples. Based on the measured data, influencing factors on bioaccumulation of Cd and its contamination risk in study area were discussed. The results indicate that the bioaccumulation and mobility of Cd from soil to rice grain are restrained by high soil pH, high contents of organic matter, clay mineral and carbonate. Guangxi has strong development of karst landform, with the continuous process of soil formation, the activity of Cd in the soils in such areas will gradually become stable. In general, the environmental risk of soil cadmium contamination in karst areas of Guangxi is relatively low. But preventing soil acidification is the significant measure to prevent the environmental risk of it.

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KEYWORDS
Karst; soil contamination; cadmium; bioavailability; risk assessment

1. Introduction

Cadmium (Cd) is one of the most harmful elements in the environment [1,2]. Excessive intake can cause damage to human bones, lungs, kidneys, etc. [3]. In 2014, the 'National Soil Pollution Status Survey Bulletin' issued by China reported that Cd was the foremost inorganic pollutant in farmland. According to the «Geochemical Investigation Report of Cultivated Land in China» published in 2015, there is a large area of soil with Cd content exceeding limit in southwest karst area of China. Karst region is a typical geological high background area of soil heavy metals. A majority proportion (over 97%) of the parent carbonate is leached during the weathering and the subsequent soil formation

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processes, resulting in significant enrichments of trace elements in the residual materials [4,5]. A review of soil contamination in China showed that soils developed from sedimentary parent materials (especially limestone) tend to accumulate much higher Cd concentrations than others [6].

The karst area of Guangxi is about 76,600 km^2, accounting for 32.24% of the total area of the province. It is the region with the strongest karst development in China and one of the provinces with the most serious soil Cd contamination in China. Since 2013, a large number of land quality geochemical surveys have been carried out in Guangxi Province, and the preliminary results show that Cd content in the soil of Guangxi is significantly higher than other regions in China [7–11]. The karst geological background is the main natural controlling factor for the high Cd content in the soil. In addition, the intensive interference of mining activities and other human activities also causes local soil Cd pollution [12–19].

Excessive cadmium can migrate from soil to crops, subsequently threatening the health of local people. At present, there are few studies revealing the risk of crops exceeding the standard in karst areas of Guangxi Province. Moreover, the understanding of the effect of soil properties on the bioavailability of cadmium is still not comprehensive. In this study, we present analytical results of vertical soil profiles, rice grains and corresponding root-soils in karst areas of Guangxi Province, and discuss the effects of six different soil properties on transfer and bioaccumulation of Cd from vertical and horizontal perspectives. This is of great significance for understanding the geochemical behaviour of Cd, and so as to guide the prevention and control of soil Cd contamination in karst areas.

2. Materials and methods

2.1. Sample collection

Considering that the degree of soil development will affect the soil properties, using the existing work data, three research areas with different degrees of soil development in Guangxi Province were selected, from strong to weak in order: Guigang, Nanning, Laibin (Figure 1). The study area is characterised by a subtropical climate with strong chemical weathering, and has an average annual precipitation of 1450 mm and average annual temperature of 22°C.

Three soil profiles were laid out in paddy field derived from carbonate rocks in each study area. Samples were collected during the paddy field drainage period, the actual depth of profile was subject to bedrock or water encounter, and the sampling density was 1 sample/10 cm. A total of 137 soil samples were collected from nine soil profiles. Meanwhile, 6–10 rice grains and corresponding root-soil samples were collected at the top of each profile within a radius of 200 m during the rice harvest period, and a total of 86 sets of samples were collected from the nine profiles. The rice specie we collected is indica.

2.2. Chemical analysis

Soil profile samples and root-soil samples were analysed for Cd, pH, Corg, Fe_2O_3, Al_2O_3, CaO, MgO, SiO_2 in accordance with soil sample analytical schemes [20]. Soil pH value was determined by an ion-selective electrode method. A volumetric technique was

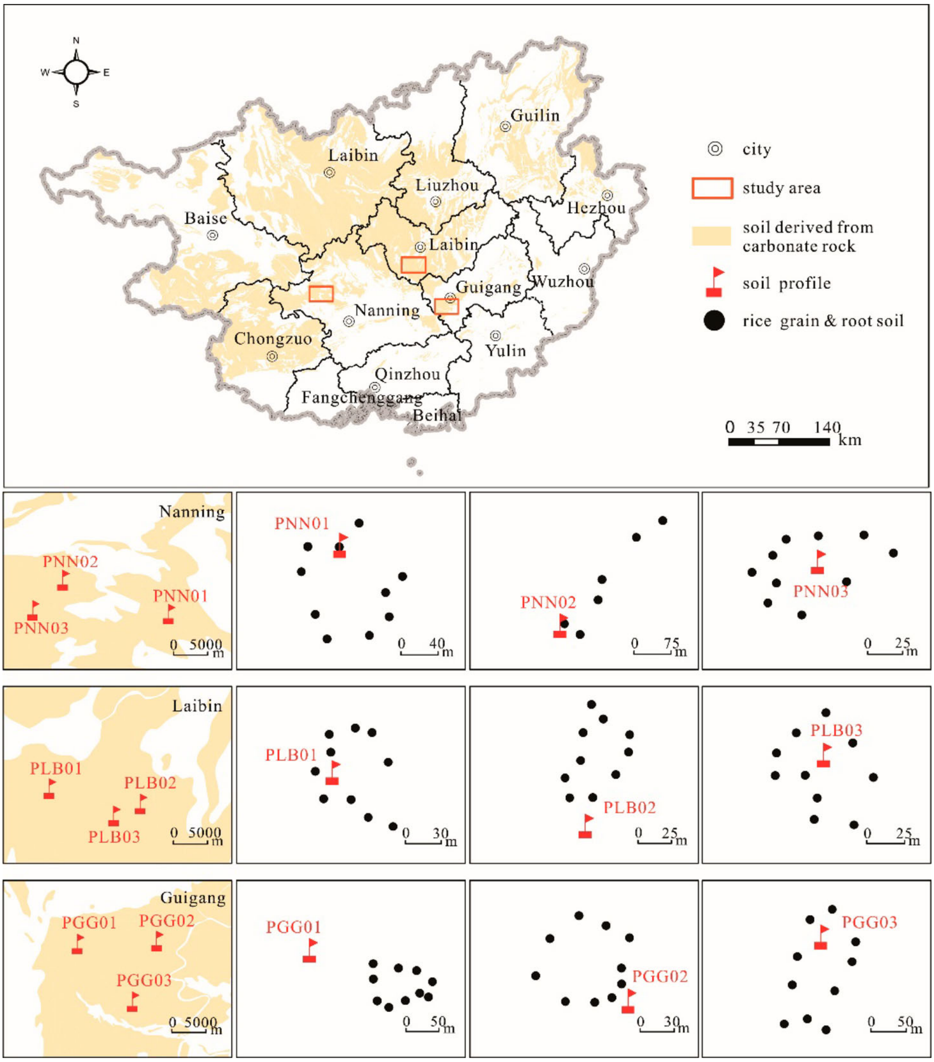

Figure 1. Study area and sampling sites.

adopted in order to test Corg concentration. X-ray fluorescence spectrometry (XRF) was implemented to measure the contents of Fe_2O_3, Al_2O_3, CaO, SiO_2. MgO content was detected by inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). In addition, we used inductively coupled plasma mass spectrometry (ICP-MS) to analyse the concentrations of Cd after the soil samples were decomposed by $HCl–HNO_3–HClO_4–HF$.

We used sequential extraction procedure for analysis the metal speciation of Cd in root-soil samples. This procedure was modified from the methods developed by Tessier [21] and Quezada-Hinojosa [22]. This method separated Cd into the following seven fractions. F1:water soluble fraction (using ultrapure water for 2h at 25°C; F2:ion-exchangeable fraction (using 1 M $MgCl_2$ for 1 h at 25°C); F3:fraction bound to carbonates (using 1 M NaOAc and buffer at pH = 5.0 for 5 h, 25°C); F4:fraction bound to humic acid (using 0.1 M $Na_4P_2O_7$ and buffer at pH = 10.0 for 1 h, 25°C); F5:fraction bound to

Fe-Mn oxides (using 0.25 M $NH_2OH{\cdot}HCl$/20% HOAc); F6:fraction bound to OM (using 1 M $NH_2OH{\cdot}HCl$/25% CH_3COOH); F7:residual fraction (using HF–HNO_3–$HClO_4$ for 72 h, 65°C).

Moreover, based on the national standard for food safety [23], ICP-MS was applied to measure the concentrations of Cd after the rice grain samples were digested by HNO_3 and H_2O_2.

The accuracy and precision of the analyses of all the samples were in accordance with the required specifications [24].

2.3. Biological enrichment coefficient (BCF)

BCF can reflect the ability of crops to absorb and enrich various elements from the soil, which is generally expressed by the ratio of the content of an element in a certain part of the crop to the content of this element in the root-soil [25].

$$\mathrm{BCF} = C_{\mathrm{grain}}/C_{\mathrm{soil}}$$

where C_{grain} and C_{soil} represent the contents of heavy metals in the crop grain and its corresponding root-soil, respectively.

2.4. Migration coefficient (MF)

The effect of soil properties on the solubility of Cd essentially affects the chemical fraction of Cd in the soil, that is, the association mode of Cd in soil. Some researchers have proposed that the migration capacity of heavy metals in the soil can be described by the migration coefficient MF [26,27].

$$\mathrm{MF} = \frac{F_1 + F_2}{F_1 + F_2 + F_3 + F_4 + F_5 + F_6 + F_7}$$

Therein, $F1$–$F7$ corresponds to the above seven fractions.

2.5. Si–Al ratio (Sa)

Si exists in the form of quartz (SiO_2) and silicates. Quartz is the most common mineral in crustal rocks and is abundant in soil as a residual primary mineral. Al mainly exists in secondary clay minerals, so the Si–Al ratio is an important parameter to reflect the degree of soil mineral weathering. The smaller the Sa value, the stronger the degree of soil weathering development [28].

$$\mathrm{Sa} = C_{\mathrm{SiO_2}}/C_{\mathrm{Al_2O_3}}$$

where $C_{\mathrm{SiO_2}}$ and $C_{\mathrm{Al_2O_3}}$ represent the contents of SiO_2 and Al_2O_3 in the soil.

2.6. Correlation analysis

Kolmogorov–Smirnove (K–S) and Q–Q charts were used to test the normal distribution of the research data. The test results showed that some indicators such as BCF did not conform to the normal distribution, so Spearman correlation coefficient was selected.

3. Result and discussion

According to the risk control standard for soil contamination of agricultural land [29], a total of 70 samples of 86 root-soil samples exceeded the risk control standard of Cd, with the exceeding rate of 81.40% (Table 1). According to the contaminant limit in food [30], Cd content in 8 rice grains exceeded the limit, which is 0.2 mg/kg, and the exceeded rate was only 9.30%, indicating that excessive Cd in soil is not equal to that in rice grain.

The comparison of BCF corresponding to different profiles shows that there are significant differences among the profiles. Generally speaking, the BCF in Guigang area is lower, while that in Laibin area is higher (Figure 2).

Meanwhile, the over standard condition of rice grain is basically consistent with the BCF. Among the eight exceed standard rice samples, four are at the top of PNN02 profile, three are at the top of PLB01 profile and one is at the top of PLB03 profile.

According to the excessive situation of rice grain and the difference of BCF, four profiles, PLB01 (high BCF), PNN02 (high BCF), PGG03 (low BCF) and PNN03 (low BCF), are taken as representatives, and the effects of different soil properties on the bioavailability of Cd are further investigated by combining the data of 86 sets of rice grain and root-soil.

3.1. Total Cd

From the profile data, PLB01 with the highest BCF has the lowest Cd content (Figure 3(A)). The bubble plots of Cd contents in root-soils, Cd contents in rice grains and BCF do not show a good rule, on the contrary, Cd contents in rice grains and BCF are higher at a relatively low level of Cd contents in root-soils (Figure 4(A)). There is a negative correlation between BCF and root-soil Cd according to correlation analysis, but it was not significant (Table 2). In the box figure of MF and Cd in root-soils, MF do not increase or decrease obviously with the increase of Cd (Figure 5(A)), and there is a slightly positive correlation between them (Table 2). No clear relationship between the total Cd contents of soils and its bioavailability most likely indicate that total Cd contents do not significantly affect the bioavailability of Cd.

3.2. PH

The profiles PLB01 and PNN02 with high BCF have the lowest near-surface pH, indicating that the roots are in a relatively acidic environment (Figure 3(B)). The bubble diagram shows that Cd contents in rice grains and BCF increase with the decrease of pH (Figure 4(B)), and there is a significant negative correlation between BCF and pH (Table 2).

Table 1. Risk control standard for soil Cd contamination of agricultural land in China.

Pollutants	Land use types	pH range	Screening value (mg/kg)
Cd	paddy field	$pH \leq 5.5$	0.3
		$5.5 < pH \leq 6.5$	0.4
		$6.5 < pH \leq 7.5$	0.6
		$pH > 7.5$	0.8

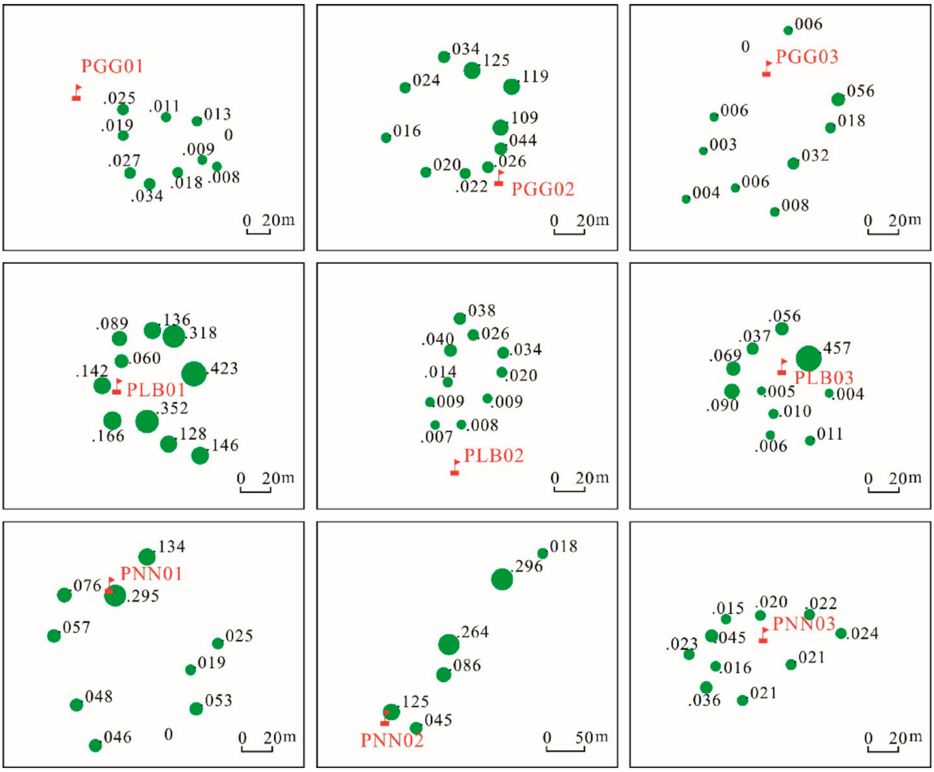

Figure 2. Comparison of BCF to different profiles.

Meanwhile, as pH decreases, MF shows an increasing trend (Figure 5(B)), and it is negatively correlated with pH significantly (Table 2). All of the above indicates that a decrease in pH will lead to an increase in the ability of Cd to migrate and be easily absorbed by crops.

Various studies have confirmed that pH is the most important factor affecting Cd adsorption in soil [31–37]. The adsorption capacity of soil to Cd is mainly determined by the negative charge on the adsorption surface. With the increase of pH, the concentration of OH^- in the soil will decrease. That is to say, the negative charge on the soil surface will increase, and the retention capacity of soil to Cd will become stronger [38,39].

In addition, the increase of pH can effectively drive the hydrolysis reaction of metal ions, which is mainly manifested in Fe, Al, Mn oxides and hydroxides [40–42]. Many researchers have reported that Cd adsorption on metal oxides increases dramatically a pH increases from 6 to 9 [43–45]. XAFS detect that at pH 7, Cd is adsorbed as an outer complex, at pH 8 as a combination of external and internal complexes, and at pH 9 as primarily an inner complex [46].

The study area is characterised by a hot and humid subtropical climate. The soil in this area is widely distributed with iron and manganese nodules with large differences in particle size. Although it is conducive to the fixation of Cd, but if the soil is continuously acidified, it will have an activation effect on the Cd adsorbed and the risk should not be ignored.

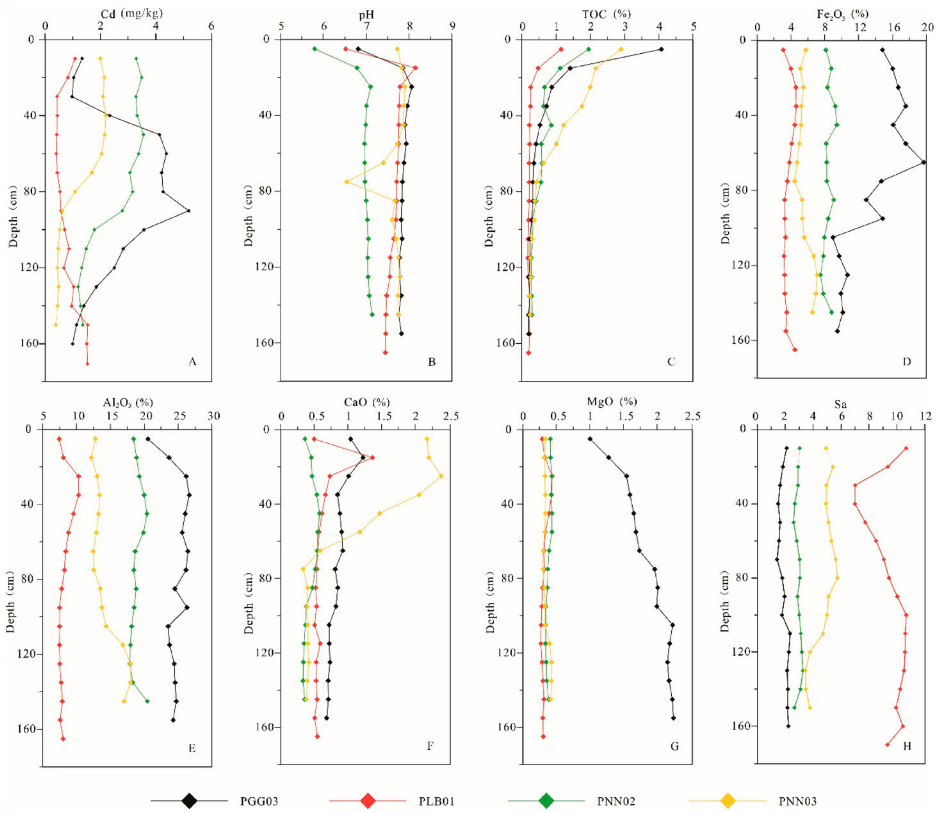

Figure 3. Comparison of soil properties to different profiles.

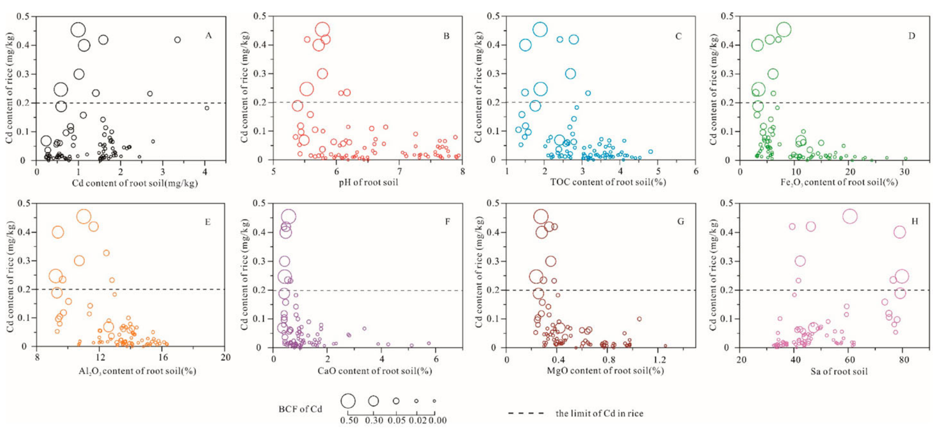

Figure 4. Bubble diagram of soil properties, Cd content in rice grains and BCF.

3.3. Organic matter

PGG03 and PNN03 with low BCF have high near-surface Corg content, indicating that the roots are in an environment with relatively more organic matter (Figure 3(C)). The bubble

Table 2. Correlation coefficients between root-soil indexes, BCF and MF.

	Cd	pH	Corg	Fe_2O_3	Al_2O_3	CaO	MgO	Sa	MF	BCF
Cd(rice)	0.190*	−0.262**	−0.297**	−0.553**	−0.309**	−0.350**	−0.575**	0.529**	0.521**	0.809**
BCF	−0.372	−0.530**	−0.295**	−0.378**	−0.387**	−0.621**	−0.465**	0.500**	0.467**	
MF	0.036	−0.194*	−0.02	−0.534**	−0.331**	−0.237*	−0.454**	0.502**		0.467**

*Significantly correlated at 0.05 level (both sides).
**Significantly correlated at 0.01 level (both sides)

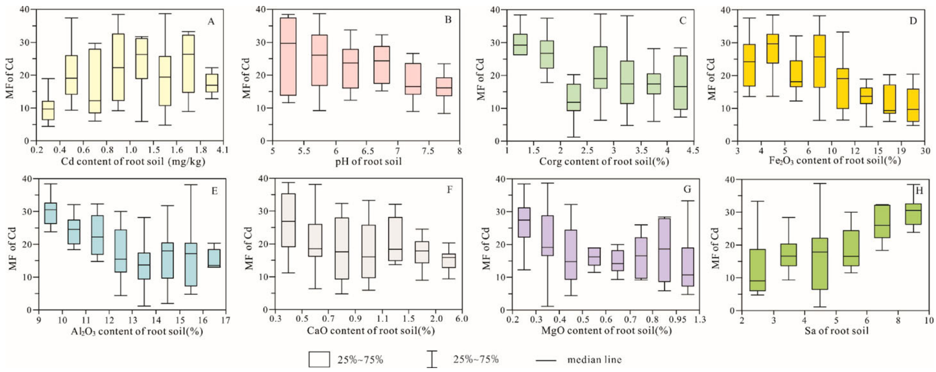

Figure 5. Box diagram of soil properties and MF.

diagram shows that Cd contents in rice grains and BCF have an increasing trend with the decrease of Corg contents in root-soils (Figure 4(C)), and BCF is negatively correlated with Corg (Table 2). The above shows that the reduction of organic matter in soil is conducive to the migration of Cd. MF and Corg contents in root-soils are not well correlated here (Figure 5(C), Table 2). It may be because organic matter is composed of a series of organic compounds with low and high molecular weight, and its influence on the fractions of Cd is complex.

Soil organic matters, such as humus acid or fulvic acid, contain a large number of organic ligands [47,48], among which the most important ones are carboxyl, carbonyl and phenolic groups [49]. Cd can complexize with these organic ligands to form stable complexes and reduce its leaching migration in the soil [50–54].

The accumulative amount of soil organic matter in a karst environment is more abundant than that in other weathered soil under the same climatic conditions [55], and this is beneficial to the fixation of Cd in study area.

3.4. Clay mineral

Clay minerals are important mineral components in soil, mainly including aluminosilicate minerals and oxide minerals. Due to large specific surface area and high surface activity, they have a strong influence on the adsorption of heavy metals. For karst areas with strong soil formation, the content of Fe_2O_3 in the soil can characterise the content of iron oxide minerals to a certain extent, while the content of Al_2O_3 can characterise the content of aluminium oxide minerals and aluminosilicate minerals.

PGG03 with lowest BCF presents the highest Fe_2O_3 and Al_2O_3 content in soil vertically, while PLB01 with greatest BCF presents the lowest Fe_2O_3 and Al_2O_3 contents. However, PNN02 and PNN03 profiles do not exhibit the above rules (Figure 3(D,E)). The bubble diagram shows that with the decrease of Fe_2O_3 and Al_2O_3 contents in root-soils, the Cd contents in rice grains and BCF greatly increase (Figure 4(D,E)), and BCF is negatively correlated with Fe_2O_3 and Al_2O_3 (Table 2). In addition, MF decreases with increasing Fe_2O_3 and Al_2O_3 contents and they have a significant negative correlation (Figure 5(D,E), Table 2). These indicate that the increase of clay mineral contents could effectively retain Cd and reduce its bioavailability.

Forbes et al. proposed that the affinity of clay mineral surface to heavy metal ions could be considered as the affinity of mineral surface to hydroxyl substances of heavy metals [56]. During the hydrolysis process of metal ions, Cd enters the OH^- group by complexing with monodentate or multidentate ligands, forming a hydroxyl complex with stable chemical bond [31].

$$XOH + Cd^{2+} \rightarrow XOCd^{+} + H^{+}$$

$$2XOH + Cd^{2+} \rightarrow (XO)_2Cd + 2H^{+}$$

As pH increases, clay minerals are prone to hydrolyse. Therefore, although the Fe_2O_3 and Al_2O_3 contents of PNN03 are relatively low, due to its highest pH, it will promote the hydrolysis process and effectively inhibit the migration of Cd. Similarly, although the Fe_2O_3 and Al_2O_3 contents of PNN02 are relatively high, its pH is the lowest, which is not conducive to the fixation of Cd and leads to the increase of its bioavailability.

During the process of soil evolution in the study area, stable new minerals such as ferromanganese nodules and aluminosilicates are continuously formed, and heavy metals are continuously adsorbed [53,57–59]. As the adsorption time prolongs, heavy metals may spread to the mineral crystal lattice structure [60,61], which is of great importance for the fixation of heavy metals such as Cd in study area.

3.5. Carbonate

In the soils derived from carbonate rocks, the carbonate mainly exists in the form of Ca^{2+}, Mg^{2+} binding with CO_3^{2-}. The contents of these two ions reflect the contents of carbonates in the soils.

PLB01 and PNN02 with larger BCF have lower CaO contents in near-surface soils, which indicate that the roots are in a carbonate lack environment (Figure 3(F)). The contents of MgO in the three profiles of PLB01, PNN02 and PNN03 are not different on the whole, while PGGD03 with the smallest BCF corresponds to the highest MgO contents (Figure 3(G)). The bubble diagram present that Cd contents in rice grains and BCF increase significantly with the decrease of CaO and MgO contents in root-soils and BCF is negatively correlated with CaO and MgO (Figure 4(F,G), Table 2). MF decreases with increasing CaO and MgO contents in root-soils and has a significant negative correlation (Figure 5(F,G), Table 2). These prove that the increase of carbonate contents in soils will inhibit the migration of Cd to crops.

Ca^{2+} and Mg^{2+} ions are the most important components in soil base cations, which have an important regulation effect on soil acidification, and Ca^{2+} has stronger inhibition ability on soil pH than Mg^{2+}. The higher contents of carbonates in soil will result in the stronger buffering ability for soil acidification, which is beneficial to the immobilisation of Cd.

Furthermore, since Cd^{2+} and Ca^{2+} have similar ionic radius, some Cd^{2+} will replace Ca^{2+} into carbonate by isomorphism. As the carbonate dissolves, this part of Cd^{2+} enter into the soil solution and increases its bioavailability [37].

Therefore, although the soils in karst areas have a good fixation effect on Cd due to the high contents of carbonates, it is particularly important to prevent the dissolution and loss of carbonates caused by soil acidification.

3.6. Degree of soil development

According to the Sa value, PGGD03 has the strongest soil development degree, corresponding to the smallest BCF, and PLBC01 has the weakest development degree, corresponding to the largest BCF (Figure 3(H)). The bubble diagram shows that with the increase of Sa, the Cd contents in rice grains and BCF have an increasing trend, but some points do not conform to the above rule (Figure 4(H)). These points mainly come from the rice samples around PNN02 (the profile has the lowest pH). Moreover, there is a significant positive correlation between BCF and Sa, MF and Sa (Figure 5(H), Table 2).

The weathering of carbonate rocks mainly includes leaching, hydrolysis, hydration and oxidation. With the progress of soil formation, the soil will gradually approach the aluminium-rich desiliconization stage.

The above indicates that on the general trend, with the continuous process of soil formation, Cd in the soils of karst areas will gradually become stable, but soil acidification will still stimulate the activity of it.

4. Conclusion

The main soil properties that affect the transfer and bioavailability of Cd in soils derived from carbonate rocks include pH, organic matter contents, clay mineral contents, carbonate contents and degree of soil development.

Generally speaking, the abundant carbonates in the soil of karst areas are natural guards against heavy metals. In addition, the high accumulation of organic matter in such soils is an advantageous condition for reducing the bioavailability of Cd. Due to the influence of climate, rainfall and other conditions, karst development is most intense in Guangxi Province area. The soils are basically in the stage of aluminium-rich and desilication, and stable new minerals under surface conditions such as Fe, Mn, Al nodules and aluminosilicate are constantly formed, which are very beneficial to the fixation of Cd. In general, the environmental risk of soil cadmium contamination in such area is relatively low.

Although the above favourable conditions exist in karst area of Guangxi Province, the risk of excessive Cd in this area should not be taken lightly. Preventing soil acidification is the primary measure to prevent and control the risk of soil cadmium contamination. Furthermore, it is necessary to reduce further pollution caused by human activities such as mining and fertilisation.

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Disclosure statement

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