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Land-Use Types Regulate Se:Cd Ratios of Natural Seleniferous Soil Derived from Different Parent Materials in Subtropical Hilly Areas

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Abstract: As natural selenium (Se)-rich soil in China is generally characterized by a high geological background of cadmium (Cd), the safe utilization of such seleniferous soil remains a challenge. The accumulating evidence shows that the threshold value of the Se:Cd ratio is a determinant of regulating Cd accumulation in plants. However, the factors modulating the soil's Se:Cd ratio in selenium-enriched regions are not well understood. Here, a comprehensive study aimed at quantitatively analyzing the effects of land-use types, parent-material types, and soil properties on the distribution and influencing factors of Se, Cd, and the Se:Cd ratios. According to land use and parent-material types, 77 soil samples were collected in Yuanzhou District, a typical naturally seleniferous area in the subtropical hilly area. The results suggested that, compared with quaternary red clays (qrc), the Se content of soils derived from river and lake sediments (rls) and weathered acidic crystalline rocks (wacr) decreased by 5.81%–19.75%, while the weathered quartzite (wq)-derived soils was increased significantly. The soil Cd content in an orchard was significantly reduced compared with that in a paddy field. A redundancy analysis (RDA) revealed that SOM, Total K, and Total P significantly affected the changes in Se and Cd contents. In addition, the land-use type had the most significant effect on the Se:Cd ratio, with a regression coefficient of −0.6999 analyzed by the binary logistic regression model ($p < 0.05$). Furthermore, pH and Total K were the critical soil properties in controlling the Se:Cd ratio. The study indicated that the Se:Cd ratio in natural selenium-rich soil was mainly regulated by land-use types. Therefore, it is a feasible measure to regulate the Se:Cd ratio by using agronomic practices, mainly regulating soil pH, for the safe utilization of selenium-rich soil with a high Cd background.

Keywords: soil Se:Cd ratio; land-use type; parent material; seleniferous soil; soil pH; subtropical hilly area

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Citation: Sun, C.; Rong, Q.; Guo, X.; Guo, J.; Chen, Y.; Chang, Y.; Chen, J.; Zhang, Q.; Zhou, C.; Cai, H.; et al. Land-Use Types Regulate Se:Cd Ratios of Natural Seleniferous Soil Derived from Different Parent Materials in Subtropical Hilly Areas. *Forests* **2023**, *14*, 656. https://doi.org/10.3390/f14030656

Academic Editor: Anna Zavarzina

Received: 28 January 2023
Revised: 8 March 2023
Accepted: 21 March 2023
Published: 22 March 2023

1. Introduction

Selenium (Se) is an essential trace element for microorganisms, animals, and humans, and also a beneficial element for plants [1,2]. As approximately one billion people's health is threatened by Se deficiency in the world [3], obviously it is a worldwide environmental issue. Seeing that about one-half of croplands are composed of Se-deficient soil in China, it is estimated that 39%–61% of the population in China suffers from inadequate Se [4]. Fortunately, there are several natural selenium-rich areas in China, such as Enshi City in

Hubei Province, Yichun City in Jiangxi Province, Shitai County in Anhui Province, and Ziyang County in Shaanxi Province [5,6]. Plants growing in selenium-rich soil can uptake and transform the Se in the soil, thus providing an excellent source of selenium nutrition for the human body through the food chain [7]. However, natural selenium-rich areas in China generally have a high geological background of cadmium (Cd) [8]. Se and its associated heavy metals in seleniferous soil are mainly derived from the same parent materials, such as carbonaceous siliceous black shale and pyrite [8–10]. Although Cd contamination has become a severe environmental issue worldwide and caused extensive concern [11,12], it is worth noting that some natural selenium-rich areas are facing the problem of soil Cd pollution, which seriously limits the utilization of the selenium-rich soil.

As Se can alleviate the toxic effects of Cd through antioxidation, reducing the cadmium bioavailability and uptake, obvious antagonistic effects between Se and Cd have been observed in many studies [13,14]. It can be seen that Se alleviating Cd toxicity in plants is dependent on the Se dosage application [15]. Accordingly, a threshold value of the Se to Cd ratio is a determinant of regulating Cd accumulation in plants [8,15]. In dryland, a Se addition accelerated Cd accumulation in corn plants when the bioavailable Se:Cd molar ratios were less than 0.7 in the soil, but a significant decrease in Cd accumulation was observed, accompanied by increased levels of bioavailable Se in the ground when the Se:Cd ratio was more than 0.7 [16]. In the paddy field, when the ratio of Se to Cd was higher than 1, the Cd level and TF-Cd in all the parts of the rice simultaneously reached the lowest levels, indicating that Se and Cd exhibited a mutual suppression threshold for the uptake and transport in rice [17]. The possible mechanisms underlying the threshold effect between the soil Se:Cd ratio and the accumulation and translocation of Cd are forming insoluble CdSe complexes by Se and Cd in the soil at 1:1 of the Se:Cd ratio [18], and/or bounding to Cd by SeCysCysSe at a lower Se:Cd ratio and by SeMet at a higher Se:Cd ratio in plants [16]. Owing to the key mechanism that Se chelates Cd metal ions in soil and plants [19], antagonistic effects between Se and Cd were observed in fruits, vegetables, and grains [20–22]. However, few studies on Se:Cd ratios in Se-rich soil have been reported. Accordingly, a deep understanding of this topic benefits the safe utilization of natural Se-rich soil.

It is demonstrable that the chief control of Se and heavy metal concentrations lies in the soil parent material in the pristine environment, as it is believed to be the primary source of Se and Cd in soil [23]. The parent materials also could affect the soil formation processes and soil properties, which may indirectly change the content of soil trace elements [24,25], as the mobility of Se and Cd are both affected by soil physical and chemical processes, including precipitation–dissolution, the reduction–oxidation reaction, and adsorption-desorption [5]. Moreover, in the process of soil formation, the influence of soil's physical and chemical properties on the Se content tends to increase [26]. Other factors that alter the concentrations of Se and Cd in the soil include biological action, precipitation, and human activities [10]. However, the content of Se and Cd in different land-use patterns was quite different. It was reported that cultivated land had higher Cd contents than forestland in the topsoil of selenium-rich areas [27]. In addition, the distribution characteristics of soil heavy metals under different land-use types had a significant correlation with the soil properties [28]. Along with returning farmland to forest or land exploitation, changes in land-use patterns were common in subtropical hilly areas [29,30]. However, both parent materials and land-use types contributed to the variability of soil properties [31]. As a result, the previous studies were primarily conducted through incubation, pot, or plot experiments. How Se and Cd contents and the Se to Cd ratio change in the area with a high geological background of Se and Cd is barely known. Therefore, this study aims to provide a more profound understanding concerning the critical control factors of Se and Cd distribution and Se:Cd ratios in seleniferous subtropical hilly areas, which are the main producing areas of agricultural products in China.

2. Materials and Methods

2.1. Study Area

Yuanzhou District is located in the western part of Jiangxi Province, China, 27°33′–28°05′ N, 113°54′–114°37′ E, which has an area of 2205 km^2 (Figure 1). The region comprises 28 towns. The climate is a humid subtropical monsoon climate, rich in light and heat, with an annual average temperature of 14–17 °C and annual precipitation of 1500–1800 mm. The terrain of the area is characterized by low mountains and hills, surrounded by mountains to the south, west, and north. The central and eastern parts are widely covered with hills, and there are narrow valleys and plains between the hills. The soil-forming parent materials here mainly include quaternary red clays (qrc), river and lake sediments (rls), weathered acidic crystalline rocks (wacr), weathered argillaceous rocks (war), weathered carbonate rock (wcr), weathered quartzite (wq), and weathered red sandstone (wrs) (Figure 1a). The soil types mainly include red soil, paddy soil, and calcareous soil. Red soil and paddy soil can develop from various parent materials, such as wacr, wq, war, wrs, and qrc. However, calcareous soil is mainly derived from wcr. The land-use types mainly include paddy fields, dryland, orchards, and forestland (Figure 1b). Citrus and kiwifruit are mainly planted in the orchards, and peanuts, rape, and vegetables are very common in the dryland. The study area is mainly dominated by the tree species of Masson pine (*Pinus massoniana*), China fir (*Cunninghamia lanceolata*), and camellia (*Camellia oleifera*). The amount of fertilizer applied in the paddy field was N 180 kg/ha, P_2O_5 75 kg/ha, and K_2O 90 kg/ha from urea, superphosphate, and potassium chloride, respectively. The distribution area of selenium-rich soil (Se > 0.4 mg/kg) was 1819.49 km^2, accounting for 82.50 % of the total area.

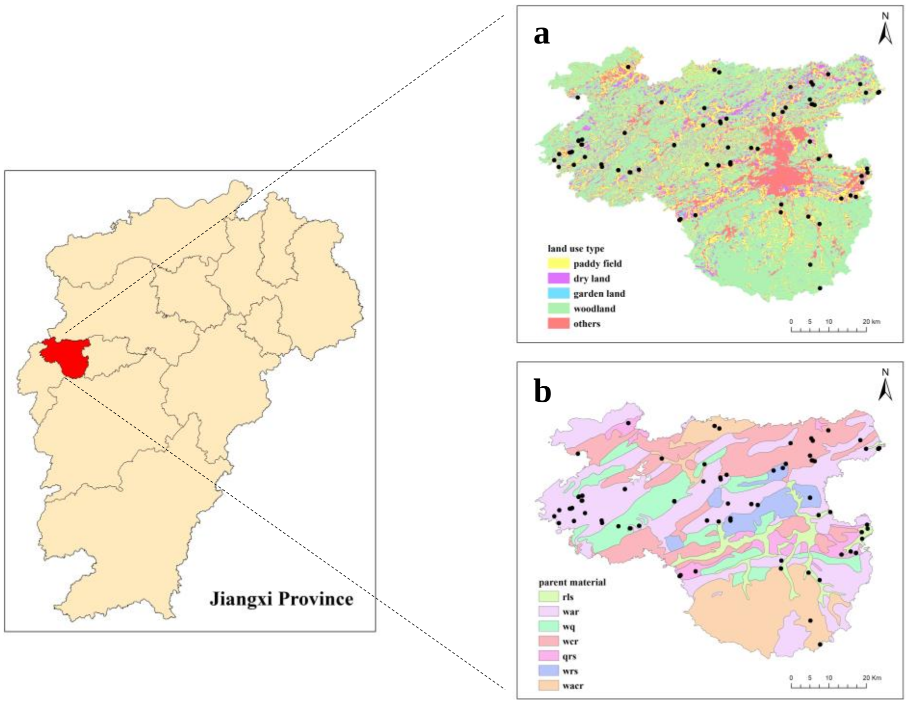

Figure 1. Overview of the study area and distribution of sampling points under different land-use types (**a**) and parent materials (**b**). Parent materials: qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, and wrs—weathered red sandstones.

2.2. Sampling and Analysis

Soil samples were collected in Yuanzhou District in mid-May 2019. According to the land use, parent-material types, land area, and topography, atypical areas such as ditches, ridges, and roadsides were avoided. The location of samples was determined flexibly according to the diagonal distribution method and plum blossom distribution method.

Five topsoil samples (0–20 cm) around each sampling point were taken. After mixing, a quartering method was used. About 1 kg of each soil sample was reserved to be brought back to the laboratory. In total, 77 soil samples were collected, including 40 paddy field samples, 20 dryland samples, 10 orchard samples, and 7 forestland samples (Figure 1a). The samples distributed by qrc, rls, wacr, war, wcr, wq, and wrs were 6, 6, 6, 10, 36, 5, and 8, respectively (Figure 1b).

After the plant residues were removed, the soil samples were air-dried indoors, passed through 60-mesh and 100-mesh nylon sieves, respectively, and packed in ziplock bags for analysis. The soil samples' physicochemical properties were investigated according to Bao [32]. Potentiometry was used to measure the soil pH with a 1:2.5 soil-to-water paste mixture. The soil organic matter (SOM) and total nitrogen (Total N) contents were measured by the dichromate oxidation and the Kjeldahl methods, respectively. Total phosphorus (Total P) was determined by the molybdate colorimetric method after perchloric acid ($HClO_4$) digestion, and total potassium (Total K) was measured by flame spectrometry after melting with sodium hydroxide. To determine the Se concentration, the digestion solution was reduced with 6 mol/L HCl and then measured by hydride generation atomic fluorescence spectrometry (XGY-1011A). HNO_3-$HClO_4$ (3:2, v/v) was used to digest the soil samples, and the Cd concentration in the digestion solution was measured by an inductively coupled plasma optical emission spectrometer (Agilent Technologies, Santa Clara, CA, USA).

2.3. Statistical Analysis

For the initial calculations, the data processing and descriptive statistical analyses were performed with Excel 2016 (Microsoft Corp., Redmond, WA, USA). Boxplots were used to present the features of soil Cd, Se, and Se:Cd ratios among the different parent materials or land-use types by SigmaPlot 12.0. A redundancy analysis (RDA) with the Monte Carlo permutation test (499 permutations) was performed to determine if the soil Cd and Se could be correlated with the soil properties, as implemented in Canoco 5.0. Taking the qrc and paddy field as controls of different parent materials and land-use patterns, respectively, the data were analyzed by calculating the natural logarithm of the response ratio (lnRR) for the soil Cd or Se among the different parent materials and land-use patterns [33].

The variance (v) of the lnRR was calculated by using Equation (1).

$$\mathrm{v} = \frac{S_t^2}{\left(\mathrm{n}_t \times \mathrm{X}_t^2\right)} + \frac{S_C^2}{\left(\mathrm{n}_c \times \mathrm{X}_c^2\right)} \quad (1)$$

where S_c and S_t represent the standard deviations of the control and the other groups, respectively; and where n_c and n_t represent the number of replicates for the control and the other groups, respectively, and where X_c and X_t represent the means of the control and the other groups, respectively. The meta-analysis was performed by using the restricted maximum likelihood estimator (RMLE) estimation in the rma.unl model of the 'metafor' package programmed using the R (v4.1.1) [34]. The mean effect size (lnRR) and its 95% confidence interval (95% CI) were calculated with bias correction and generated by using bootstrapping. When the 95% CI of the response variables did not include zero, the parent materials or land-use pattern were considered to be significantly different from the control group. The samples were divided into high Se:Cd ratio and low Se:Cd ratio categories according to whether the soil bioavailable molar Se:Cd ratio was greater than 0.7 [16]. The determined class values of the dependent (the Se:Cd ratio) and independent variables (parent materials, land-use patterns, pH, SOM, Total N, Total P, Total K) were used to create the input table for logistic regression modeling. By using the function glm(), the coefficients were calculated, which were then used to estimate the influences of the independent variables in the binary logistic regression model [35].

3. Results

3.1. Soil Properties in the Area

The soil properties in the area are shown in Table 1. The concentration distribution of soil pH in this area was in the range of 5.54–6.82, with an average value of 6.19. The soil derived from wcr had the highest pH. The soil pH in the paddy fields and dryland was lower than that in the orchards and forestland, while the SOM was the opposite. The distribution of Total N in the soil was consistent with the distribution of SOM. Although the average soil Total P was 0.61 g/kg, it was found to be 0.33 g/kg and 0.29 g/kg in the wrs-derived soil and forestland soil, respectively. The Total K of the soil derived from the wcr, wq, and wrs was relatively lower than that from the others, but the highest content was found in the paddy field, which reached 1.66%.

Table 1. Soil properties of different parent materials and land-use types in the study area.

Type		pH	SOM (%)	Total N (g/kg)	Total P (g/kg)	Total K (%)
Parent material *	qrc	5.75 + 0.22	3.17 + 1.62	1.63 + 0.71	0.65 + 0.21	1.55 + 0.22
	rls	5.68 + 0.48	3.27 + 1.15	1.41 + 0.50	0.65 + 0.19	1.91 + 0.54
	wacr	5.82 + 0.33	3.63 + 1.15	1.68 + 0.52	0.60 + 0.30	2.48 + 0.29
	war	6.48 + 0.95	4.05 + 2.01	1.96 + 0.80	0.61 + 0.20	1.54 + 0.54
	wcr	6.58 + 0.98	3.14 + 1.57	1.55 + 0.92	0.59 + 0.34	0.97 + 0.46
	wq	5.80 + 0.69	5.59 + 2.49	2.12 + 0.72	0.78 + 0.42	1.00 + 0.21
	wrs	5.61 + 1.22	2.48 + 1.37	1.20 + 0.73	0.33 + 0.15	0.89 + 0.18
Land use	dryland	6.31 + 0.74	3.45 + 1.92	1.55 + 0.68	0.78 + 0.31	1.40 + 0.68
	orchard	5.31 + 0.91	1.86 + 0.88	1.06 + 0.36	0.47 + 0.25	1.42 + 0.58
	paddy field	6.44 + 0.86	4.69 + 1.60	2.20 + 0.65	0.61 + 0.15	1.66 + 0.52
	forestland	5.68 + 0.83	2.01 + 0.48	0.85 + 0.25	0.29 + 0.13	0.72 + 0.12

* Parent materials and land-use types in the study area. Parent materials: qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, wrs—weathered red sandstone.

3.2. Distribution of Se and Cd among Different Parent Materials and Land-Use Types

The average content of Se and Cd in the soil was 0.51 mg/kg and 0.32 mg/kg, respectively, but obvious differences were found in different parent materials or land-use types (Figure 2 and Table S1). The wq-derived soil and rls-derived soil both had higher Cd contents, with an average content of 0.37 mg/kg and 0.38 mg/kg, respectively; the lowest content of Cd was found to be 0.27 mg/kg in the wrs-derived soil. The average content of Se in the wacr-derived soil was 0.31 mg/kg, which was lower than that from the other parent material-derived soils. The black rock series and their metamorphic rock parent materials (wq, war, and wcr) all had higher Cd and Se contents (Figure 2a,c). There was little difference in the soil's Se content among different land-use types, whereas a considerable variation was found in the soil's Cd content (Figure 2b,d).

The meta-analysis was applied to analyze the response ratio (RR) of Se and Cd for different categories (Figure 3). Compared with that from the qrc, the Se content of the soil derived from the rls and the wacr decreased by 5.81%–19.75%, while that from the wq-derived soils significantly increased. There was no significant difference in the soil Cd content among different parent materials (Figure 3a). For different land-use types, there was no significant difference in the soil's Se content compared with that in paddy fields. However, compared with the paddy fields, the soil's Cd content in the dryland, orchards, and forestland decreased by 34.30%–72.75%, among which the soil's Cd in the orchards decreased significantly (Figure 3b).

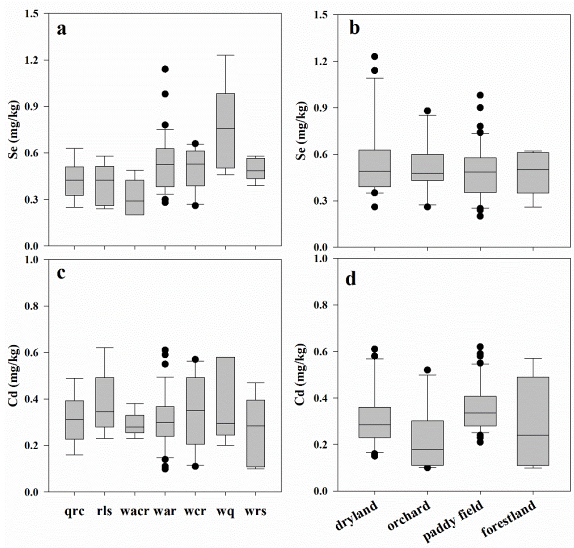

Figure 2. Boxplots of soil Se and Cd contents among different parent materials and land-use types. (**a**) soil Se contents among different parent materials; (**b**) soil Se contents among different land-use types; (**c**) soil Cd contents among different parent materials; (**d**) soil Cd contents among different land-use types. Parent materials: qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, and wrs—weathered red sandstones.

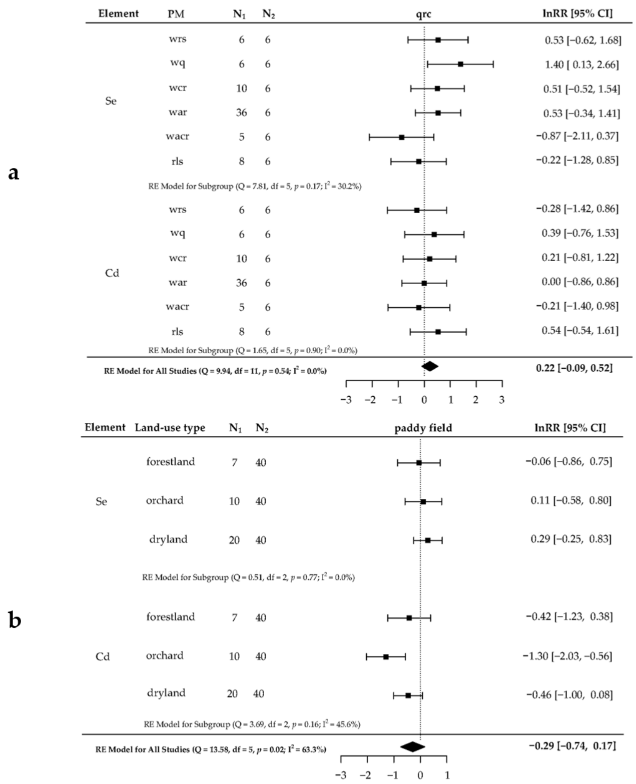

Figure 3. Comparison of mean effect sizes of parent materials (**a**) and land-use types (**b**) on soil Se and Cd contents. N_1 and N_2 represent the sample size for each variable and control; error bars represent 95% confidence interval (CI). Categories whose 95% CI does not cross 0 (represented by the vertical dotted lines) have significant differences between the variables and the control. PM—parent materials, qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, wrs—weathered red sandstone.

3.3. Effects of Soil Characteristics on Soil Se and Cd

The correlations between the Se and Cd and the soil characteristics were analyzed (Figure 4). The correlation analysis suggested that both Se and Cd were significantly positively correlated with the SOM, Total N, and Total P, but the Se was significantly negatively correlated with the Total K. There was a significant positive correlation between the Cd and the soil pH, with no significant correlation between the Se and the soil pH. The changes in the soil Se and Cd contents and soil properties among the soil samples of the different parent materials and land-use types were analyzed by redundancy analysis (RDA) to reveal the effects of the soil's physical and chemical properties (Figure 5). The results showed that SOM ($F = 26.6$, $p = 0.002$), Total K ($F = 19.2$, $p = 0.002$), and Total P ($F = 7.9$, $p = 0.002$) had significant effects on the changes in the Se and Cd contents, and the SOM, Total K, and Total P explained 26.2%, 15.2%, and 5.7% of the changes in the soil's Se and Cd contents, respectively. The results of the RDA analysis showed that the order of soil characteristics affecting the changes of Se and Cd was as follows: SOM > Total K > Total P > Total N > pH.

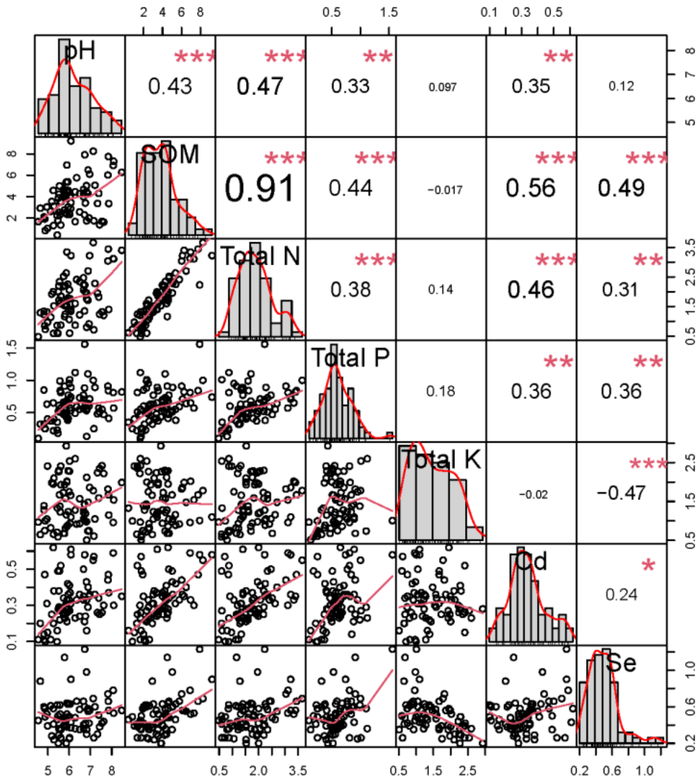

Figure 4. The correlations between Se and Cd and soil characteristics. *, **, *** means significantly different at $p < 0.05$, $p < 0.01$ and $p < 0.0001$, respectively.

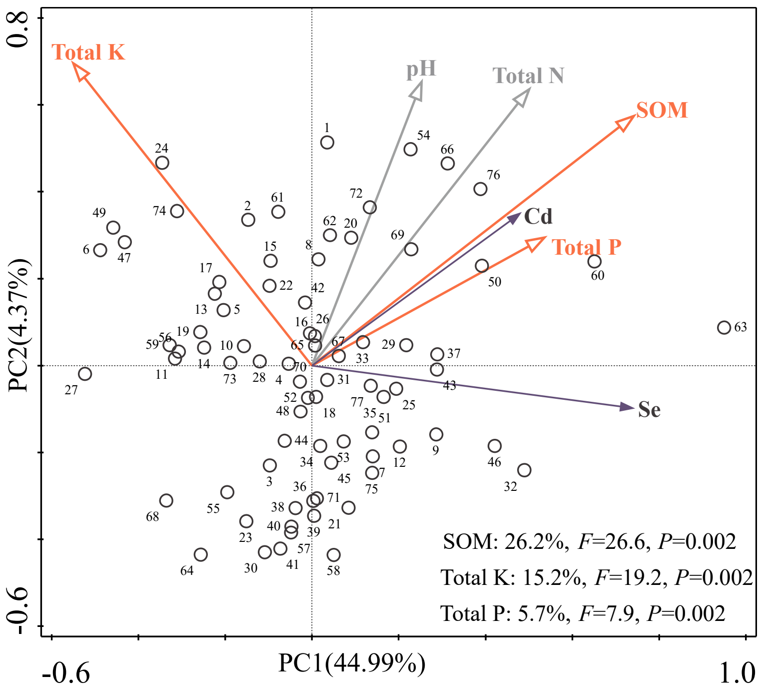

Figure 5. Results of the redundancy analysis of the correlations between soil parameters and soil Se and Cd. Red arrows indicate soil parameters that have a strong and significant impact on soil Se and Cd ($p < 0.05$). The corresponding proportion of the variation is shown in the lower right corner.

3.4. The Se:Cd Ratio Change and Its Influencing Factors in the Area

The Se:Cd ratio in the area is shown in Figure 6. The average values of the Se:Cd ratio in the dryland, orchards, paddy fields, and forestland were 1.90, 3.14, 1.39, and 2.58, respectively. The ratio of Se and Cd in the orchard and forestland changed considerably, while the change in the dryland and paddy fields was small. As to the parent materials, the Se:Cd ratios derived from the war, wcr, wq, and wrs were relatively higher than that from the others (Figure 7).

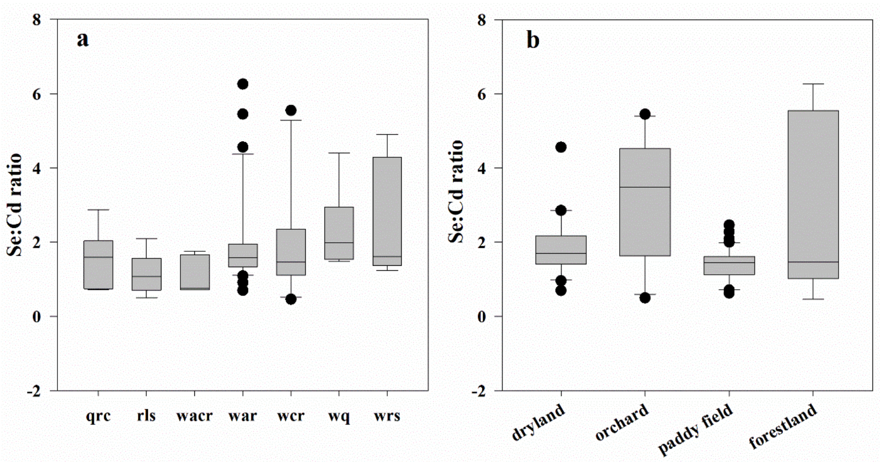

Figure 6. Boxplots of soil Se:Cd ratios among parent materials (**a**) and land-use types (**b**). Parent materials: qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, wrs—weathered red sandstones.

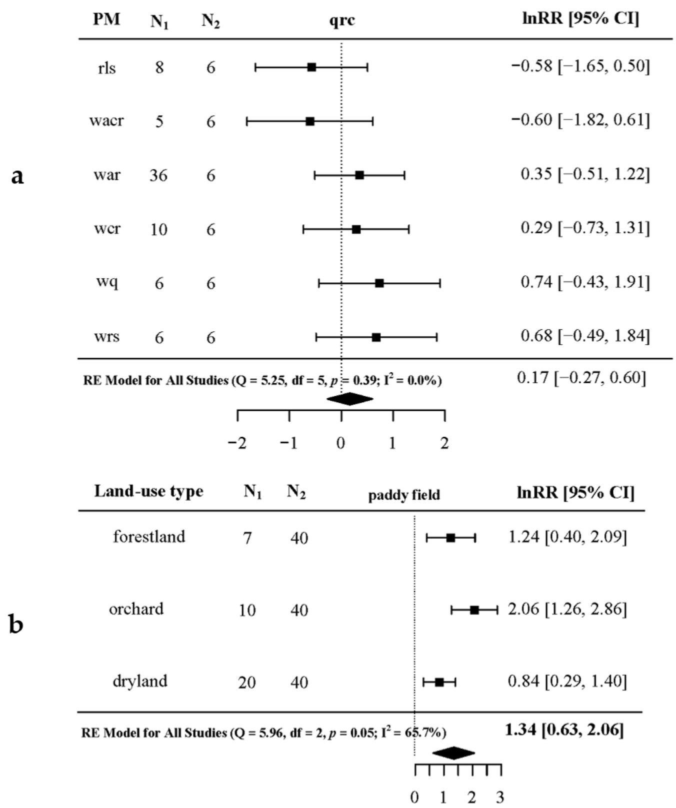

Figure 7. Comparison of mean effect sizes of parent materials (**a**) and land-use types (**b**) on soil Se:Cd ratios. N_1 and N_2 represent the sample size for each variable and control; error bars represent 95% confidence interval (CI). Categories whose 95% CI does not cross 0 (represented by the vertical dotted lines) have significant differences between the variables and control. PM—parent materials, qrc—quaternary red clays, rls—river and lake sediments, wacr—weathered acidic crystalline rocks, war—weathered argillaceous rocks, wcr—weathered carbonate rock, wq—weathered quartzite, wrs—weathered red sandstone.

To determine the influencing factors of the soil Se:Cd ratios, the samples were divided into high Se:Cd ratio and low Se:Cd ratio categories, and the binary logistic regression model was used to analyze the effects of the independent variables (parent materials, land-use patterns, pH, SOM, Total N, Total P, Total K) on the soil's Se:Cd ratio (Table 2). It was found that the land-use type was the main influencing factor for regulating the ratio of Se and Cd in the soil, with a regression coefficient of −0.6999. The soil pH and Total K also significantly affected the soil's Se:Cd ratio. In addition, under the condition that other factors remained unchanged, the soil's Se:Cd ratio would be about 0.17 times higher for each unit increase in soil pH or Total K.

Table 2. Effects of parent materials, land-use patterns, and soil characteristics on the soil's Se:Cd ratio by binary logistic regression. Goodness of fit (GOF) test: $\chi^2 = 2.8808$, df = 8, $p = 0.9416$; likelihood ratio test: $\chi^2 = 25.4$, $p < 0.001$.

Factor	Regression Coefficient (B)	Standard Error	*p*	Exp(B)
Land-use type	−0.6999	0.3381	0.03847	0.4966
pH	−1.7662	0.5832	0.00246	0.1710
Total K	−1.7683	0.7157	0.01348	0.1706

4. Discussion

4.1. The Effects of Soil Properties on Se, Cd, and Se:Cd Ratios in Natural Seleniferous Soil

Soil Cd contamination in natural seleniferous areas is a critical problem, limiting clean Se-enriched food production [5]. In this study, the soil properties were the essential factors affecting the variation in the soil's Se, Cd, and Se:Cd ratios. A positive correlation between the pH and the soil's Cd content was found, but there was little effect of pH on the Se (Figure 4). The specific mechanism for increased Cd in the soil with increasing pH may be controlled by the decreased Cd solubility [36] and increased density of adsorptive sites on the surface of soil minerals and soil organic matter [37]. In this case, a positive correlation between SOM and soil Cd was also observed (Figures 4 and 5, $p < 0.01$). However, the effect of pH on Se was more complicated than on the soil's Cd, due to the versatile valence states of Se in the soil [2]. The immobilization of Se by the soil's solid phase would be enhanced due to the adsorption processes when the soil pH was low [38]. At the same time, [Se(IV)] was stronger than [Se(VI)] in adsorption by amorphous iron oxyhydroxide [39]. However, anionic Se species would be repulsed and cause adsorption to be decreased with the negative charges on soil particles' surface increasing with the rising pH [40]. As a result, a negative correlation and no significant relationship between soil pH and soil Se contents have been reported [40–42].

A significant negative correlation between Total K and soil Se was also observed (Figures 4 and 5, $p < 0.01$). As the rainy and hot seasons overlapped, the study area, located in a subtropical region, had intense weathering. The soils that experienced intensive weathering alterations were able to adsorb more Se, owing to their higher proportion of clay minerals and Fe/Al oxyhydroxides, promoting Se retention in the soil [43]. However, extensive distribution of the 1:1 clay kaolinite in the area, widely known to contain no K, as well as its poor ability in K retention, and the rainy climate, together led to heavy leaching of K [44]. However, soil potassium was one of the leading factors controlling the Se in alkaline soil, and a positive correlation was reported [45]. Therefore, the correlation between Total K and the soil's Se contents may depend on soil pH and weathering. In the present study, the Se:Cd ratio was negatively correlated with pH and Total K ($p < 0.01$) (Table 2). This was due to the relationship between pH, Total K, and the soil's Se and Cd in our study (Figures 4 and 5, $p < 0.01$), which is related to the soil's physicochemical properties and soil weathering processes.

4.2. The Effects of Parent Materials on Se, Cd, and Se:Cd Ratio in Natural Seleniferous Soil

It was indicated that the parent materials, as the primal sources of Se in soil, make a critical contribution to the soil's Se levels [45,46]. In the present study, a significant difference was found in the soil's Se contents between the wq and qrc, but not in the soil's Cd contents between parent materials (Figures 2 and 3a), suggesting that the parent materials may exert a stronger influence on the soil's Se contents than on the soil's Cd contents in the study region. Although both wq and qrc had high clay contents, the SOM content of the wq was obviously higher than that of the qrc (Table 1). The enrichment in Se was related to the high amount of organic matter in the low-Se geological belts or the excessive Se regions [46,47]. In the study, the Se in the soil from wacr was the lowest (Figure 3a). This could be due to the high sand content and low Se concentrations in the wacr, which was weathered from granite [10]. Generally, the soil with high clay mineral contents, high Fe-Al oxides, and high organic matter in nature had higher Se values [10,47,48].

Except for wrs and rls, the distribution patterns of Se and Cd in the different parent materials were generally consistent, although the Cd contents in the different parents just varied slightly without significant differences (Figure 3a). However, the wrs-derived soils had the lowest Cd content and a higher Se content, while the rls-derived soils showed the opposite trend (Figure 3a). Many previous studies had identified that the soil's total Cd concentration had a strong correlation to the soil's Total P concentration [49], as phosphorus could precipitate with Cd as insoluble phosphate [50]. Therefore, the lowest Cd content in the wrs may be caused by the lowest Total P content (Table 1). Furthermore, a significant relationship between the soil's total Cd and Total P was also detected in this study ($p < 0.01$) (Figure 4). The lower Se content in rls-derived soil was in line with the results reported in the literature [43]. As a result, the Se:Cd ratios in different parent materials mainly depended on the abilities of the parent materials to preserve Se but not Cd (Figures 3 and 7). This may be because Se-rich rock weathering was a primary source causing Se enrichment in the soil [46]. In comparison, the higher Cd content soil may be determined by geological parent-material compositions and human activities [51,52].

4.3. The Effects of Land-Use types on Se, Cd, and Se:Cd Ratios in Natural Seleniferous Soil

Land-use types had strong influences on soil properties and thus could cause far-reaching consequences for element biogeochemical cycles [53]. In the current study, it was shown that land-use types had a stronger influence on Cd than on Se, which was different from that of the parent material patterns (Figures 2 and 3). It was reported that uncultivated agricultural soil had a lower Se concentration than that of agricultural soil [54]. Consistent with the findings, the forestland had the lowest soil Se content among the different land-use types in our study, although only a slight variation was found. However, lower Se concentrations in the agricultural soil than in the uncultivated soil were also reported [55]. This might be related to the fact that there were other factors, such as parent materials, and the mean annual precipitation [43], which were crucial in determining the soil's Se content, as the soil's Se was mainly derived from natural sources [56].

The soil's Cd concentrations over different land uses are illustrated in Figure 2d. The data analysis in this study showed apparent differences in the Cd contents in the topsoil under different soil-use types. Cd concentrations were the highest in the paddy fields, followed by the dryland and forestland, and they were the lowest in the orchard. As a result, a significant difference in the soil's Cd content between the paddy fields and orchards was found by the meta-analysis (Figure 3b). For the land use, which was a comprehensive reflection of both natural conditions and human activities [57], it has been reported that the highest Cd content in paddy soils may be related to the application of phosphorus and organic fertilizers [58]. Notably, the highest SOM and pH were observed in the paddy soil; on the contrary, those in the orchard were the lowest (Table 1). The obvious differences in soil properties caused by the land-use types may be another important reason for the distribution of Cd in the topsoil [37,59]. Accordingly, the binary logistic regression suggested that the ratio of Se to Cd was mainly determined by the land use (Table 2). Soil pH was also a dominant factor influencing the soil's Se:Cd ratios, which could directly or indirectly affect the interaction between the Se and Cd ions and the soil components [40,60].

5. Conclusions

In this study, various analytical methods were used to determine the distribution and influencing factors in Se, Cd, and Se:Cd ratios in natural seleniferous soil. The results of our study demonstrate that land-use types have a stronger influence on Cd than on Se, but the effects of parent materials are the opposite. Moreover, the Se:Cd ratio in natural selenium-rich soil is mainly regulated by land-use types, and pH and total K are the critical soil properties in controlling the Se:Cd ratios. Specifically, using agronomic practices is a practical way to regulate soil pH, so as to safely utilize selenium-rich soil with a high Cd background. It is also necessary to consider the variation of the Se:Cd ratios in the process of land-use change to reduce human health risks. However, more research is needed to clarify the threshold effects for different plants and the effects of soil Se and Cd distribution on the Se:Cd ratio in the future.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/f14030656/s1, Table S1: Soil Se and Cd contents of different parent materials and land use types in the study area.

Author Contributions: Conceptualization, C.S., Q.R. and X.G.; methodology, H.C. and X.Z.; software, J.G. and Y.C. (Yi Chen); validation, Y.C. (Yi Chen), Y.C. (Yihua Chang) and Q.Z.; formal analysis, Q.R. and J.C.; investigation, Q.Z. and C.Z.; data curation, Q.Z. and X.G.; writing—original draft preparation, C.S.; writing—review and editing, Q.R. and X.Z.; supervision, Q.R. and X.Z. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Special Key Grant Project of Technology Research and Development of Jiangxi Province ("Take-and-lead" Program) (No. 20213AAF02026); the National Natural Science Foundation of China (No. 32060728); the Youth Foundation of Jiangxi Educational Committee (No. GJJ180232); the Project of Jiangxi Selenium-Rich Agricultural Research Institute (No. JXFX21-ZD06); and the Jiangxi Province Postgraduate Innovation Special Fund Project (No. YC2021-S346).

Data Availability Statement: The data are available on request from the corresponding author.

Acknowledgments: The authors would like to thank anonymous reviewers for their valuable comments and remarks.

Conflicts of Interest: The authors declare no conflict of interest.

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