# Parent material modulates land use effects on soil selenium bioavailability in a selenium-enriched region of southwest China

Kongcao Xiao[a,b], Lufeng Lu[c], Junjie Tang[a,b,d], Hao Chen[a,b], Dejun Li[a,b,*], Yongxian Liu[e,*]

[a] Key Laboratory for Agro-ecological Processes in Subtropical Region, Institute of Subtropical Agriculture, Chinese Academy of Sciences, Changsha 410125, China
[b] Huanjiang Observation and Research Station for Karst Ecosystem, Chinese Academy of Sciences, Huanjiang 547100, China
[c] Huashan State-owned Forest Farm, Huangjiang County, Huangjiang 547100, China
[d] University of Chinese Academy of Sciences, Beijing 100049, China
[e] Institute of Agricultural Resources and Environment, Guangxi Academy of Agricultural Sciences, Nanning 530007, China

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## ABSTRACT

Land use effects on the biogeochemical cycling of the essential trace element selenium (Se) and underlying mechanisms are not well understood yet. Here, total Se ($Se_{total}$) and its potential bioavailability, as represented by phosphate extractability ($Se_{phosphate}$), were evaluated in top soils (0–15 cm) of cropland and woodland over two contrasting geological parent materials (i.e. limestone versus clasolite) in a Se-enriched region of southwest China. After about twenty years of conversion of land use from cropland to woodland, $Se_{total}$ in soils significantly ($P < 0.05$) and marginally significantly ($P < 0.1$) increased over limestone and clasolite, respectively. In terms of $Se_{phosphate}$, it was not significantly changed after land use change from cropland to woodland in soil over limestone, whereas remarkably increased in soil over clasolite ($P < 0.01$). Furthermore, a significant positive relationship between $Se_{phosphate}$ and $Se_{total}$ was found in soil over clasolite, but not in soil over limestone. Land use conversion from cropland to woodland caused a decrease in the proportion of $Se_{phosphate}$ in $Se_{total}$ over limestone, while the opposite was found over clasolite. In comparison, the proportion of $Se_{phosphate}$ in $Se_{total}$ was greater in cropland over limestone than that in cropland over clasolite, indicating that limestone derived soils may have an advantage of producing Se-rich food over clasolite derived soils. SOC content and quality (i.e. C:N ratio) crucially determined Se accumulation and bioavailability in soil over limestone, while pH played a vital role in soil over clasolite. Taken together, our results provide strong evidence that land use effects on Se biogeochemistry can be substantially modulated by the underlying geology, and have important practical implications for effective utilization and management of Se-enriched soil resources in other parts of the world.

## 1. Introduction

Selenium (Se) is a naturally-occurring metalloid element that is essential for humans, animals, microorganisms and some other eukaryotes, but is toxic at excess amounts (Hartikainen, 2005; Fairweather-Tait et al., 2011; Natasha et al., 2018). The range between dietary Se deficiency and toxicity is very narrow (40–400 µg d$^{-1}$) (Rayman, 2000; Lenz and Lens, 2009). Globally, Se deficiency is much more prevalent than Se toxicity, arousing widespread concerns. Since soil is the fundamental source of Se for human and livestock via food chain (Navarro-Alarcon and Cabrera-Vique, 2008; Winkel et al., 2015), understanding the bioavailability of Se in soil and its dependence on environmental parameters is thus of crucial importance for preventing Se deficiency and improving human Se status (Sharma et al., 2015).

Land use has been well recognized to be a major factor causing far-reaching consequences for element biogeochemical cycles (Xiao et al., 2009). To date, however, most studies examining the influence of land use focus on macroelements such as carbon, nitrogen, and phosphorus (Herpin et al., 2002; Potthast et al., 2012; Cherubin et al., 2016), not the trace elements (Anguelov and Anguelova, 2009; Tu et al., 2013; Islam et al., 2015), especially Se (Tuttle et al., 2014; Yanai et al., 2015; Plak and Bartminski, 2017). The direction and magnitude of land use effect on soil Se dynamics is not well known at present. Based on a nationwide investigation in China, Tan et al. (2002) reported that, on average, agricultural soils had a slightly higher Se concentration (269 µg kg$^{-1}$) than uncultivated soils (206 µg kg$^{-1}$). In addition, Zhu et al. (2008) found that agricultural soils were much more Se-enriched (3370 µg kg$^{-1}$) relative to uncultivated soils (1140 µg kg$^{-1}$) in a typical high-Se area Yutangba, Enshi in Hubei Province. However, the opposite pattern, i.e., lower Se concentrations in agricultural soils than

that in uncultivated soils, has also been demonstrated in several researches (Tolu et al., 2014; Shang et al., 2015; Xing et al., 2015). The inconsistency indicates that some other factors which are crucial in determining soil Se dynamics may have been overlooked when comparing among land uses.

As a primary source of Se in soil, parent materials are usually considered to play a fundamental role in determining soil Se levels (Wadgaonkar et al., 2018; Jia et al., 2019). Strong positive correlations between Se levels in underlying parent rocks with those of overlying soils have been recorded (Li et al., 2008; Pan et al., 2017). Beyond serving as the main source, parent materials also exert strong controls on the physicochemical properties of the overlying soil (Neff et al., 2006; Vestin et al., 2006; Andersson et al., 2015), further affecting Se behavior in soil phase (Tolu et al., 2014; Carvalho et al., 2019). With the progress of pedogenesis, the influence from parent materials on the Se status of overlying soils gradually weakens while roles of soil physicochemical attributes strengthen (Wang et al., 2013). Recently, accumulating evidence shows that land use often causes different effects on soil properties and processes in various geological areas (Ozdemir and Askin, 2003; Tu et al., 2013; Deng et al., 2015; Li et al., 2017a; Wang et al., 2018b), suggesting that the underlying geology may greatly modulate land use effects on the biogeochemical cycles of elements, including Se.

Guangxi, a province located in southwest China, is well-known for longevity, partly ascribing to the high background levels of Se in soil (Liu et al., 2013). Since 1990s, a significant proportion of cropland has been converted to woodland in Guangxi, mainly due to the implement of 'Grain-for-Green' program. To the best of our knowledge, however, how this massive land use conversion affects Se biogeochemistry in the environment has rarely been investigated in Guangxi and other Se-enriched areas undergoing land use change. Land use change is often accompanied by significant changes in a variety of soil attributes including pH, SOC and metal oxides contents (Maia et al., 2010; Malik et al., 2018; Lizaga et al., 2019), all of which are in close relation to Se behavior in the environment (Winkel et al., 2015; Li et al., 2017b). Therefore, it can logically be assumed that soil Se cycling would be substantially altered by land use conversion. To enhance our ability to manage soil Se resources and improve Se status in human populations under future land use change scenarios, a better understanding of factors and underlying mechanisms controlling the direction and magnitude of land use effects on soil Se status is undoubtedly needed.

Numerous studies have indicated that conversion of cropland to woodland causes increase in the soil organic carbon (SOC) stock (Song et al., 2014). SOC is widely regarded as the most important attribute that closely links with Se concentrations in soils (Roca-Perez et al., 2010; Shand et al., 2010; Tolu et al., 2014; Xu et al., 2015; Li et al., 2017b). Therefore, we hypothesized that land use conversion from cropland to woodland would lead to an elevation in soil Se level, as a consequence of SOC accumulation (**Hypothesis I**). Many researches, including our previous studies, have demonstrated that some soil physicochemical properties responded differentially to land use change over different geological areas (Tu et al., 2013; Li et al., 2017a; Wang et al., 2018b). Given that soil Se bioavailability is synthetically influenced by soil physicochemical properties (Winkel et al., 2015; Li et al., 2017b), we thus hypothesized that land use effect on soil Se bioavailability would also vary over different geological parent materials (**Hypothesis II**). In the present study, concentrations of total Se and phosphate extractable Se were investigated in top soils of cropland and woodland over areas of contrasting parent materials (i.e., limestone vs. clasolite) in huangjiang, northwest Guangxi. The major objective was to test whether land use effects on Se levels and bioavailability would be modulated by the underlying geology.

## 2. Materials and methods

### 2.1. Study area

The study area (23°40′N–25°25′N, 107°35′E–108°30′E) was located in the northwest of Guangxi Zhuang Autonomous Region, southwest China, which belongs to subtropical monsoon climate and has a mean annual air temperature ranging from 17.8 °C to 22.2 °C and a mean annual precipitation ranging from 1346 mm to 1640 mm. This region is geologically interwoven with limestone, dolomite, clasolite and their mixtures. Limestone is the most common parent material for soils in this region, accounting for approximately 85% of the total area (Wang et al., 2018b). The soil developed from limestone and clasolite can be categorized into calcareous lithosols (limestone soil) and ferralsols, respectively, according to the FAO/UNESCO classification system.

### 2.2. Field sampling

Field sampling was carried out from the end of March to early June 2015. The sampling sites were selected based on the type of parent material, i.e., limestone and clasolite, and land use, i.e., cropland and woodland. The information about land use history was obtained by interviewing local residents. Croplands were mainly managed under a corn-soybean rotation with annual fertilizer inputs of nitrogen (N), phosphorous (P), and potassium (K) at about 150, 60, and 120 kg ha$^{-1}$ in the form of compound fertilizer, respectively. Woodlands (stand age ~20 years) were all converted from cropland and received no fertilization after conversion. The major species of woodlands is Masson pine (*Pinus massoniana* Lamb.), which is a very important timber species widely distributed in south China.

In total, 60 sampling sites were selected using a paired-site approach, including 15 croplands and 15 woodlands over limestone and 15 croplands and 15 woodlands over clasolite. During the sampling time, all the croplands were planted with corn. At each site, a 20 × 20 m sampling plot was established and surface mineral soil samples (0–15 cm) were collected with a stainless steel auger (5 cm in diameter). 10–15 soil cores were randomly sampled in each plot and then mixed to form a composite sample. Soils were air-dried at room temperature and then sieved through a 2 mm-mesh sieve, with roots and stone fragments removed by hand-sorting. The 2 mm sieved soil samples were used to measure phosphate extractable Se (Se$_{phosphate}$) which can to a great extent represent the overall amount of potentially bioavailable Se fractions (Bajcan et al., 2001; Zhao, 2004; Zhao et al., 2005; Favorito et al., 2017), and to analyze some edaphic properties such as pH, size fractions, exchangeable cations, and Fe-Al oxides. An aliquot of 2 mm sieved sample was further ground to pass through a 0.15 mm-mesh sieve to measure total soil Se (Se$_{total}$), SOC, total N (TN), and total P (TP).

### 2.3. Chemical analyses

Se$_{total}$ was measured according to the method as described in Xing et al. (2015) with a slight modification. Briefly, ~0.5 g of sample was added with a mixed acid solution of HNO$_3$ and HClO$_4$ (10 ml, vol:vol 4:1) and allowed to stand overnight at room temperature. Then, the mixture was electrothermally heated first at 60 °C for 30 min, followed by at 120 °C for 30 min, then at 180 °C for 30 min, and finally at 220 °C until the resulting solution volume was < 1 ml. After cooling, 5 ml of 6 M HCl was added and boiled for 1 min to completely convert Se$^{6+}$ to Se$^{4+}$. Afterwards, the solution was transferred to a 25 ml test tube and diluted with Milli-Q water for Se analysis. Blank samples were included throughout the analysis as a contamination control. A standard reference material of calcareous soil (GBW07404) purchased from the Institute of Geophysical and Geochemical Exploration, Geological Survey of China was used for quality control.

Se$_{phosphate}$ was extracted by 0.1 mol L$^{-1}$ (pH = 7.0) KH$_2$PO$_4$-

$K_2HPO_4$ buffer solution. Briefly, ~2 g soil was weighed into a 50 ml polycarbonate centrifuge tube and then 25 ml of phosphate buffer solution was added. The tube was subsequently shaken on a reciprocal shaker for 4 h at a speed of 200 rpm. Afterwards, the tubes were centrifuged at a speed of 4000 rpm for 10 min and the supernatants were decanted to a 50 ml conical flask. Then, a mixed acid solution of $HNO_3$ and $HClO_4$(10 ml, vol:vol 4:1) was added to conical flask and the resulting solution was further digested at 220 °C for 30 min to transform all dissolved Se to $Se^{6+}$. After cooling, 5 ml of 6 M HCl was added and boiled for 1 min to completely reduce $Se^{6+}$ to $Se^{4+}$. After cooling, the solution was transferred to a 50 ml test tube and treated with 1 ml concentrated HCl to maintain a reducing environment, and diluted with Milli-Q water for subsequent analysis.

The prepared samples for determining $Se_{total}$ and $Se_{phosphate}$ were analyzed by hydride generation-atomic fluorescence spectrometry (HG-AFS830, Titan Instrument Co., Ltd., Beijing, China). Data for $Se_{total}$ and $Se_{phosphate}$ were finally expressed as µg Se $kg^{-1}$ dry soil. $Se_{total}$ of the standard reference material was 651 $\pm$ 83 µg Se $kg^{-1}$ (the certified values of 640 $\pm$ 140 µg Se $kg^{-1}$).

Soil gravimetric water content (GWC) was measured by drying soil at 105 °C to constant weight. Soil pH was measured in a suspension of 1:2.5 soil:water ratio using a pH meter (FE20K, Mettler-Toledo, Switzerland). SOC was analyzed by wet oxidation with potassium dichromate redox colorimetric method. TN was determined using an elemental analyzer (EA 3000, EuroVector, Italy). TP was pretreated by acid digestion with a $H_2SO_4$ + $HClO_4$ solution. Soil available P (AP) was extracted with 0.5 M $NaHCO_3$. Then, TP and AP were determined using the molybdenum colorimetric method. Particle size fractions, i.e. clay (< 2 µm), silt (2–50 µm), and sand (50–2000 µm) were determined on a laser diffraction particle size analyzer after removal of organic matter (Mastersizer, 2000, Malvern, UK). Exchangeable cations, including calcium (Ca), magnesium (Mg), K, and sodium (Na) were analyzed after extracting with 1 mol $L^{-1}$ ammonium acetate at pH 7.0 with inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Iron (Fe), aluminum (Al) and manganese (Mn) were extracted by ammonium oxalate solution (0.2 mol $L^{-1}$, pH = 3), which can extract free amorphous oxides and hydrous oxides and Fe and Al-humus complexes, and were measured on ICP-AES.

### 2.4. Statistical analyses

Statistical analyses were performed with SPSS software package version 16.0 ((SPSS Inc., Chicago, IL, USA). All data were first checked for normality and homogeneity before variance analysis. Two-way ANOVA was adopted to examine the effects of parent material and land use and their possible interactions on $Se_{total}$, $Se_{phosphate}$, and other edaphic variables. Since significant interactions between parent material and land use were observed for some variables, one-way ANOVA was further performed to examine the effect of land use on $Se_{total}$, $Se_{phosphate}$, and other edaphic variables in the soil over limestone and clasolite, respectively. Stepwise multiple linear regression approach was used to identify the key controlling factors for soil Se content and bioavailability. All reported significant differences are at $P < 0.05$ level unless otherwise stated.

### 3. Results

#### 3.1. Parent material and land use effects on soil Se content and bioavailability

Since no significant interactive effect of parent material and land use was found for $Se_{total}$ (Table S1), only the main effects were presented (Fig. 1). There was no significant difference in $Se_{total}$ between soils over limestone and clasolite, but there was a significant ($P < 0.05$) or marginally significant ($P < 0.1$) difference between the cropland and the woodland over limestone and clasolite, respectively
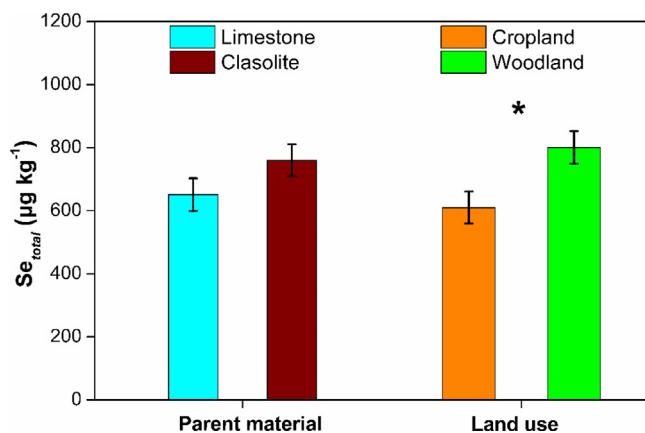


**Fig. 1.** Main effects of parent material and land use on $Se_{total}$. The bars represent mean $\pm$ standard error (n = 30). The symbol * denotes a significant difference in $Se_{total}$ between the cropland and woodland at $P < 0.05$.

**Table 1**
Land use effects on $Se_{total}$, $Se_{phosphate}$, the proportion of $Se_{phosphate}$ in $Se_{total}$, and other edaphic variables over limestone and clasolite, respectively.

| Dependent Variable | Land use effects over limestone | | Land use effects over clasolite | |
|---|---|---|---|---|
| | Cropland | Woodland | Cropland | Woodland |
| $Se_{total}$ (µg $kg^{-1}$) | 570 (44) | 732 (51) | 649 (74) | 870 (102) |
| $Se_{phosphate}$ (µg $kg^{-1}$) | 69 (13) | 56 (9) | 35 (6) | 90 (18) |
| $Se_{phosphate}/Se_{total}$ (%) | 13 (2) | 7 (1) | 5 (0.7) | 9 (1.1) |
| Moisture (%) | 27.0 (1.9) | 27.1 (1.7) | 21.2 (1.3) | 35.2 (2.1) |
| SOC (g $kg^{-1}$) | 26.4 (2.5) | 28.7 (2.9) | 14.4 (1.0) | 24.0 (1.9) |
| C:N ratio | 7.0 (0.3) | 10.8 (0.5) | 7.8 (0.4) | 12.4 (0.5) |
| pH | 7.2 (0.1) | 7.2 (0.2) | 4.9 (0.1) | 4.5 (0.1) |
| TP (g $kg^{-1}$) | 1.1 (0.8) | 0.8 (0.1) | 0.5 (0.07) | 0.3 (0.03) |
| Available P (mg $kg^{-1}$) | 13.3 (2.0) | 6.2 (1.1) | 32.5 (5.9) | 7.0 (0.4) |
| K (cmol $kg^{-1}$) | 0.4 (0.04) | 0.3 (0.03) | 0.2 (0.01) | 0.2 (0.01) |
| Na (cmol $kg^{-1}$) | 0.4 (0.02) | 0.6 (0.04) | 0.4 (0.04) | 0.6 (0.03) |
| Ca (cmol $kg^{-1}$) | 30.9 (2.7) | 27.9 (3.2) | 7.1 (0.6) | 6.5 (0.3) |
| Mg (cmol $kg^{-1}$) | 26.4 (3.4) | 1.82 (0.1) | 9.6 (1.2) | 1.0 (0.08) |
| Al (%) | 0.2 (0.03) | 0.3 (0.03) | 0.2 (0.02) | 0.2 (0.02) |
| Fe (%) | 0.3 (0.03) | 0.4 (0.05) | 0.4 (0.05) | 0.4 (0.05) |
| Mn (%) | 0.1 (0.02) | 0.08 (0.02) | 0.05 (0.01) | 0.04 (0.01) |
| Clay (%) | 20.6 (1.3) | 20.8 (1.3) | 18.2 (0.9) | 19.2 (0.9) |
| Silt (%) | 71.4 (1.2) | 68.1 (1.4) | 73.9 (1.4) | 73.6 (0.8) |
| Sand (%) | 8.0 (0.7) | 11.1 (0.9) | 7.9 (0.9) | 7.1 (0.5) |

Values are presented as means with standard errors in parentheses.

(Fig. 1, Tables 1 and S2). Relative to the cropland, the mean value of $Se_{total}$ in the woodland were 28.4% and 34.1% higher over limestone and clasolite, respectively (Table 1).

A significant interactive effect of parent material and land use was observed for $Se_{phosphate}$ ($P < 0.01$, Table S1). $Se_{phosphate}$ was not significantly different between the cropland and woodland over limestone, but was significantly different between the two land uses over clasolite ($P < 0.01$, Fig. 2, Table S2), with the mean $Se_{phosphate}$ in the woodland about 2.6 times greater than that in the cropland (35 µg $kg^{-1}$). In terms of the proportion of $Se_{phosphate}$ in $Se_{total}$, it was also interactively influenced by parent material and land use (Fig. 3, Table S1). On average, the proportion of $Se_{phosphate}$ in $Se_{total}$ in the cropland was significantly higher than that in the woodland over limestone, while the opposite was detected over clasolite (Fig. 3, Tables 1 and S2).

#### 3.2. Factors controlling soil Se content and bioavailability

Stepwise multiple linear regression analysis revealed that controlling factors for soil Se content and bioavailability were greatly
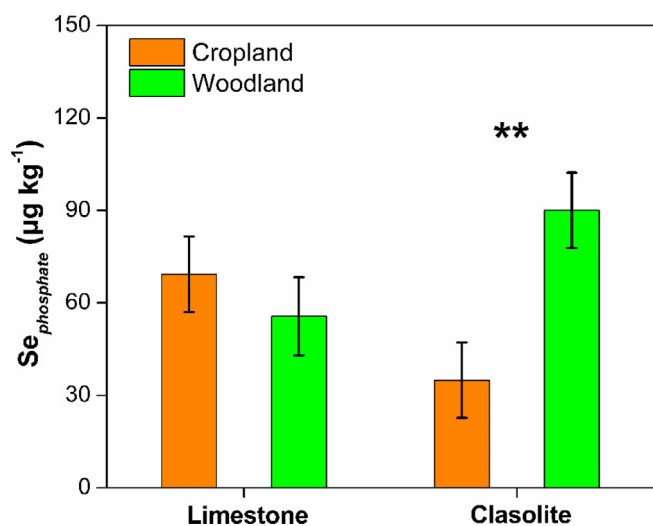
**Fig. 2.** The effect of land use on $Se_{phosphate}$ over limestone and clasolite, respectively. The bars represent mean $\pm$ standard error (n = 15). The symbol ** denotes a significant difference in $Se_{phosphate}$ between the cropland and woodland over clasolite at $P < 0.01$.
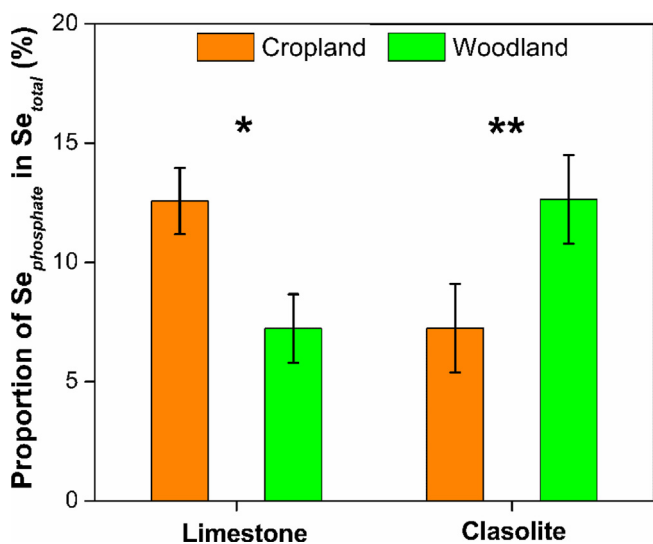


**Fig. 3.** The effect of land use on the proportion of $Se_{phosphate}$ in $Se_{total}$ over limestone and clasolite, respectively. The bars represent mean $\pm$ standard error (n = 15). The symbol * and ** denote a significant difference in the proportion of $Se_{phosphate}$ in $Se_{total}$ between the cropland and woodland over limestone and clasolite at $P < 0.05$ and $P < 0.01$, respectively.

dependent on parent material. For soil over limestone, C:N ratio, Fe and Mn were the best predictors for variation in $Se_{total}$ (Table 2), while SOC concentration and C:N ratio were the major explanatory variables for $Se_{phosphate}$ (Table 2). In addition, C:N ratio and SOC concentration were the primary explanatory variables for the proportion of $Se_{phosphate}$ in $Se_{total}$ (Table 2). For soil over clasolite, pH was the strongest explanatory variable for variation of $Se_{total}$, explaining 45% of $Se_{total}$ variance (Table 2). SOC concentration and pH were the major two explanatory variables for $Se_{phosphate}$ variance (Table 2), and they together explained about 61% of $Se_{phosphate}$ variance. Furthermore, pH and AP were the best predictors for and together explained approximately 61% of variation in the proportion of $Se_{phosphate}$ in $Se_{total}$ (Table 2).

## 4. Discussion

### 4.1. Variations in total soil Se

In the present study, we found a significant difference in $Se_{total}$ content between land uses but not between parent materials (Fig. 1, Table S1), suggesting that land use may exert a stronger influence on soil Se content than parent material in the study region. Woodland soils had significantly ($P < 0.05$) or marginally significantly ($P < 0.1$) higher $Se_{total}$ than cropland soils over limestone and clasolite, respectively (Table 1), in line with our first hypothesis that conversion of cropland to woodland would generally lead to accumulation of Se in soil. However, the reason for Se accumulation after land use conversion from cropland to woodland in different geological regions may be different (Table 2).

In the current study, we find that soil $Se_{total}$ was strongly associated with C:N ratio instead of SOC content for soil over limestone (Table 2). Since soil C:N ratio is a general index that largely reflects soil organic matter quality (Springob and Kirchmann, 2003; Khalil et al., 2005), the strong correlation of $Se_{total}$ with soil C:N ratio likely implies that SOC quality exerts greater controls than its quantity on Se accumulation in limestone soils. This might be related to the fact that land use conversion from cropland to woodland over limestone did not cause significant change in SOC content but led to a remarkable rise in C:N ratio (Tables 1 and S2). As the decomposability of SOC usually decreases with increasing C:N ratio, woodland soils may favor Se accumulation due to the high stability of organically-bound Se.

For soil over clasolite, however, soil pH was the most crucial attribute that controls the variation in $Se_{total}$ (Table 2). In our study, soil pH was found to negatively correlate with $Se_{total}$ (Table 2), consistent with findings of previous studies (Xing et al., 2015; Cao, 2017; Luo et al., 2018; Jiang et al., 2019). One explanation for the negative relationship between pH and $Se_{total}$ is that rise/drop in pH usually causes decrease/increase in the positive charges on soil particles, thus reducing/enhancing adsorption of the anionic Se species (Goh and Lim, 2004). Another possible reason is that decline in soil pH suppresses microbial activity and then slows down the decomposition of SOC (Anderson and Domsch, 1993; Andersson and Nilsson, 2001; Aciego Pietri and Brookes, 2009), indirectly promoting Se retention in soils. In support of this view, we found that conversion from cropland to woodland over clasolite led to a significant reduction in soil pH by 0.4 and concurrently a substantial increase in SOC content by ca. 70% on average (Table 1). For areas over limestone, no clear relationship was observed between pH and $Se_{total}$, likely due to the insensitivity of pH change to land use conversion owing to high buffering capacity (Zhang et al., 2016; Jalali and Moradi, 2019). In consistent with our results, Pan et al. (2017) reported that total Se content was not significantly correlated with pH value in a karst area near Guiyang.

### 4.2. Variations in bioavailable Se

It is increasingly recognized that the amounts of biologically available fractions of Se, rather than total Se content, crucially determines the level of Se entering into food chains, reflecting the potential Se bioavailability in the environment (Zhang et al., 2014). Since $Se_{phosphate}$ frequently correlates well with Se accumulation in plants (Zhao, 2004; Zhao et al., 2005; Favorito et al., 2017), it is thus widely considered as a proxy of Se bioavailability (Keskinen et al., 2009). Although it has traditionally been thought that bioavailable Se is highly associated with total Se content (Stroud et al., 2010; Wang et al., 2011; Jia et al., 2019), there are also results showing no significant (Zhang et al., 2002; Ma et al., 2017) or weak positive correlation (Chilimba et al., 2011) between bioavailable Se and total Se content. In the current study, we only find a clear positive relationship between $Se_{phosphate}$ and $Se_{total}$ over clasolite, but not over limestone (Fig. 4), suggesting that Se bioavailability in limestone soils will not proportionally increase

**Table 2**
Results of stepwise multiple linear regression analyses showing the dependence of $Se_{total}$ (µg kg$^{-1}$), $Se_{phosphate}$ (µg kg$^{-1}$), and the proportion of $Se_{phosphate}$ in $Se_{total}$ on edaphic variables.

**Over limestone**

| $Se_{total}$ | | | | $Se_{phosphate}$ | | | | Proportion of $Se_{phosphate}$ to $Se_{total}$ | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Explanatory variable | Coefficient | Model R$^2$ | P value | Explanatory variable | Coefficient | Model R$^2$ | P value | Explanatory variable | Coefficient | Model R$^2$ | P value |
| C:N ratio | 0.351 | 0.296 | 0.029 | SOC | 0.785 | 0.227 | 0.000 | C:N ratio | −0.747 | 0.213 | 0.000 |
| Fe | 0.351 | 0.406 | 0.021 | C:N ratio | −0.507 | 0.389 | 0.014 | SOC | 0.743 | 0.393 | 0.000 |
| Mn | −0.348 | 0.513 | 0.027 | | | | | | | | |

**Over clasolite**

| $Se_{total}$ | | | | $Se_{phosphate}$ | | | | Proportion of $Se_{phosphate}$ to $Se_{total}$ | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Explanatory variable | Coefficient | Model R$^2$ | P value | Explanatory variable | Coefficient | Model R$^2$ | P value | Explanatory variable | Coefficient | Model R$^2$ | P value |
| pH | −0.673 | 0.453 | 0.000 | SOC | 0.459 | 0.520 | 0.010 | pH | −0.645 | 0.466 | 0.000 |
| | | | | pH | −0.395 | 0.607 | 0.024 | AP | −0.375 | 0.606 | 0.005 |

Positive and negative values of coefficients denote positive and negative relationship, respectively, between the explanatory variables and Se indicators.

with increasing total Se. Similarly, Tan et al. (2002) found that the correlation between available Se and total Se varies depending on soil type, e.g., the correlations are often significant in desert soil, solonchak, chestnut soil and meadow soil, but are always insignificant in red soil, brown earth and drab soil.

Consistent with our hypothesis II, land use effects on soil Se bioavailability were affected by parent material, as indicated by the concentration of $Se_{phosphate}$ and its proportion in $Se_{total}$ (Figs. 2 and 3). Furthermore, the controlling factors for $Se_{phosphate}$ also varied with parent material (Table 2). SOC content was found to be the primary factor controlling $Se_{phosphate}$ concentration irrespective of parent material (Table 2). SOC is believed to exert a dual control on Se bioavailability (Li et al., 2017b; Quang Toan et al., 2019). On the one hand, SOC can significantly reduce Se bioavailability via immobilization process. For example, Xing et al. (2015) reported a negative relationship between contents of SOC and bioavailable Se in a Se-enriched area of Anhui province, China. On the other hand, the organically-bound Se can be transformed to water-soluble Se and ligand-exchangeable Se during SOC decomposition, thus increasing the bioavailability of Se (Dhillon et al., 2007; Wang et al., 2018a; Chang et al., 2019). Our result shows that $Se_{phosphate}$ is positively correlated with SOC (Table 2), indicating that organically-bound Se is the predominant form in the phosphate extract.

Our results demonstrate that, besides SOC content, C:N ratio and pH were the most important factors affecting the concentration of $Se_{phosphate}$ over limestone and clasolite, respectively (Table 2). Contrary to $Se_{total}$, $Se_{phosphate}$ was negatively related to C:N ratio over limestone (Table 2). One possible explanation is that the decomposability of SOC decreases with increasing C:N ratio, resulting in a reduction in the solubility of Se-containing organic molecules (Supriatin et al., 2016). For soil over clasolite, $Se_{phosphate}$ exhibited a negative relationship with soil pH (Table 2), in line with the finding of Xu et al. (2018). This is probably because more $Se_{phosphate}$ was lost by leaching with the increase of soil pH under humid climate in our study region.

### 4.3. Variations in the proportion of $Se_{phosphate}$ in $Se_{total}$

The proportion of bioavailable Se in total Se is another important index widely used to reflect the bioavailability of Se in soil (Tan et al. 2002), which greatly varies across studies around the world (Table 3). In the current study, $Se_{phosphate}$ accounts for 8.7% (0.9%~28.3%) of $Se_{total}$ on average for all data set, which falls within the range reported for various soil types across China, i.e. 8%–34% (Wang et al., 2011).

We find that the proportion of $Se_{phosphate}$ in $Se_{total}$ responded differently to land use conversion over limestone and clasolite, respectively (Fig. 3). For soil over limestone, land use conversion from cropland to woodland caused a significant reduction in the proportion of $Se_{phosphate}$ in $Se_{total}$ ($P < 0.05$, Fig. 3). This is because the quality of
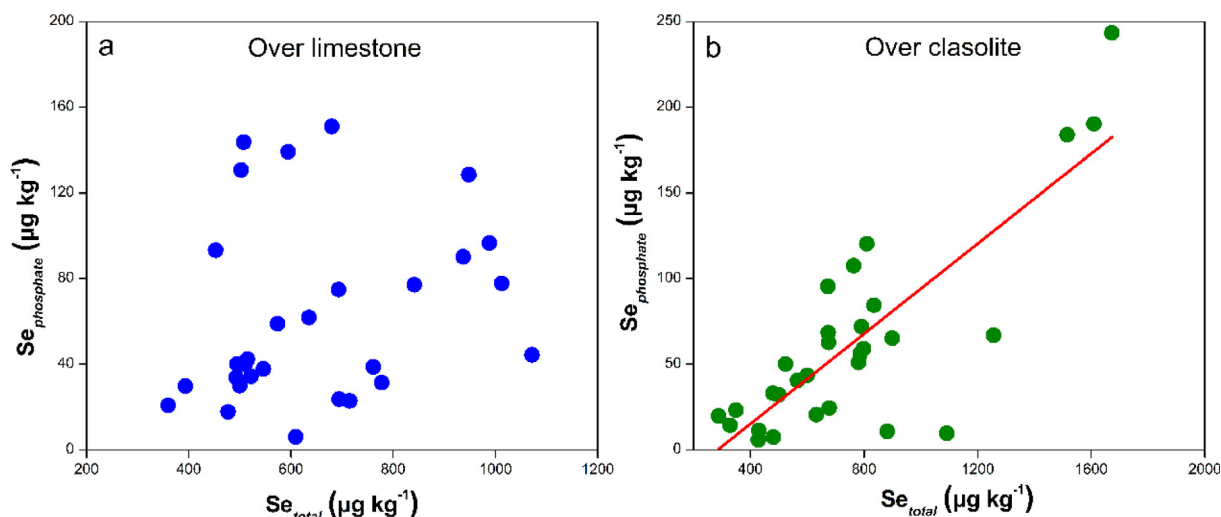


**Fig. 4.** Correlations between $Se_{total}$ and $Se_{phosphate}$ over limestone (a) and clasolite (b), respectively.

**Table 3**
Values of Se$_{phosphate}$ (µg kg$^{-1}$), Se$_{total}$ (µg kg$^{-1}$), and the proportion of Se$_{phosphate}$ in Se$_{total}$ reported in other studies.

| Study region | Se$_{phosphate}$ | Se$_{total}$ | Proportion of Se$_{phosphate}$ in Se$_{total}$ | Reference |
|---|---|---|---|---|
| Finland | 20–240 | 140–1560 | 17% (12–25%) | Keskinen et al. (2011) |
| Malawi | 6 (1–16) | 194 (52–620) | ~3% | Chilimba et al. (2011) |
| UK | 6–11 | 245–590 | 1.1–3.4% | Stroud et al. (2010) |
| China | 22–154 | 138–9540 | 8%–34% | Wang et al. (2011) |
| Enshi, Hubei, China | 960–22260 | 3180–435700 | < 5% | Yuan et al. (2012) |
| Wuhan, Hubei, China | 18 (0–57) | 200–1000 | 5.3% (0%–13.7%) | Zhou et al. (2016) |
| Ziyang, Shaanxi, China | 109 (7–388) | 5474 (370–23530) | 4.0% (1.6%–17.0%) | Zhao (2004) |
| Jiangxi, China | 9 (5–16) | 235 (98–609) | 3.8% | Zhang et al. (2017) |
| Fengchen, Jiangxi, China | 58 | 643 | ~9% | Ma et al. (2017) |
| Yongjia, Zhejiang, China | 34 (8–76) | 382 (157–633) | 9.4% (3.4%–18.9%) | Xu et al. (2018) |
| Qinghai, China | 21 (12–44) | 280 | 6.9% | Ji et al. (2012) |
| Heilongjiang, China | 1–29 | 98–246 | 0.8%–9.4% | Xu et al. (2016) |
| Shanghai, China | 17–28 | 99–234 | 11%–18% | Qu et al. (1998) |
| Hechi, Guangxi, China | 63 (6–244) | 705 (288–1673) | 8.7% (0.9%–28.3%) | This study |

Values are means with the range in the parenthesis.

SOC declined while the quantity of SOC slightly increased after land use conversion from cropland to woodland over limestone (Table 1), resulting in a decrease in soluble Se-containing organic molecules, relative to total Se. However, for soil over clasolite, land use conversion from cropland to woodland significantly decreased soil pH. As soil pH declines, the surfaces of clay minerals, organic matter and metal oxyhydroxides are more positively charged, which is conducive to sorption and retention of Se-oxyanions in soil (Liao et al., 2005; Strahm and Harrison, 2008; Khawmee et al., 2013), and consequently results in an elevation in the proportion of Se$_{phosphate}$ in Se$_{total}$.

The cropland over clasolite had a much smaller proportion of Se$_{phosphate}$ in Se$_{total}$ as well as a much lower concentration of Se$_{phosphate}$, relative to the cropland over limestone (Figs. 2 and 3), indicating that Se bioavailability in the cropland over clasolite may be much lower than that in the cropland over limestone. The results support the idea that alkaline soils have a higher Se bioavailability than acidic soils (Chilimba et al., 2011; Ji et al., 2012; Supriatin et al., 2016; Wang et al., 2018c). In the present study, lower Se bioavailability in the cropland over clasolite may have been resulted from significant loss of bioavailable Se by leaching, because the cropland over clasolite contained a much greater concentration of available P than its counterpart over limestone (Table 1). Phosphate anion in soil solution can compete strongly with Se for adsorption sites (Eich-Greatorex et al., 2010), thereby increasing the potential risk of Se loss by leaching (Lessa et al., 2016) as well as improving the bioavailability of Se in soils (Jia et al., 2019).

In addition, despite well-documented close associations of other soil properties such as Fe/Al oxides and mineralogical characteristics with Se bioavailability in soils (Nakamaru and Altansuvd, 2014; Xu et al., 2018; Jia et al., 2019), the current study failed to find significant relationships between the phosphate extractability of Se and Fe/Al oxides and particle size fractions (including sand, silt and clay) (Table 2), suggesting these parameters may have only a marginal effect on soil Se variation in the study region. Overall, our results have important implications for developing accurate and effective management strategies for utilization of Se-enriched soil resources over different geological areas.

## 5. Conclusions

In summary, our results demonstrated that about twenty years of land use conversion from cropland to woodland significantly and marginally significantly increased total soil Se in soils over limestone and clasolite, respectively. Furthermore, land use effects on soil Se bioavailability were dependent on parent material. SOC content and quality (i.e. C:N ratio) critically controlled soil Se bioavailability in soil over limestone, while pH played a vital role in soil over clasolite. Our findings will help better understand land use effects on Se biogeochemical cycling, and have important practical implications for effective utilization and management of Se-enriched soil resources in the world.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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