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Impact of land use/land cover change on the topsoil selenium concentration and its potential bioavailability in a karst area of southwest China

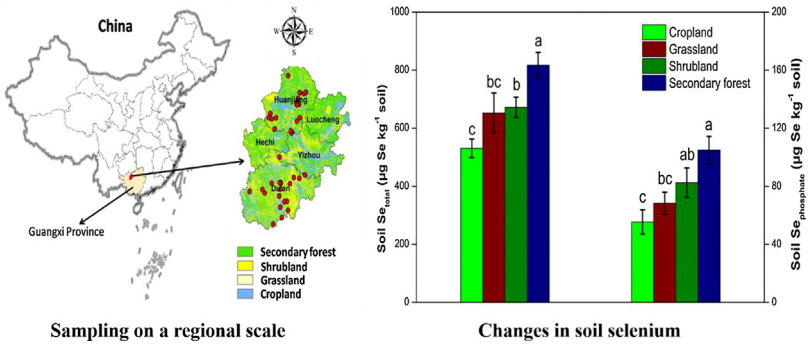
Kongcao Xiao [a,b], Junjie Tang [a,b,c], Hao Chen [a,b], Dejun Li [a,b,*], Yongxian Liu [d,*]

[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] University of Chinese Academy of Sciences, Beijing 100049, China
[d] Institute of Agricultural Resources and Environment, Guangxi Academy of Agricultural Sciences, Nanning 530007, China

HIGHLIGHTS

- Land use/land cover change affects total soil Se content and its bioavailability.
- Soil organic carbon was the overriding edaphic factor controlling soil Se status.
- Climate variables strongly influenced soil Se status at the regional scale.
- Soil Se retention increased after cropland abandonment mainly due to SOC accumulation.

GRAPHICAL ABSTRACT



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ABSTRACT

Selenium (Se) is an essential micronutrient for human health, and its abundance and potential bioavailability in the soil are of increasing concern worldwide. To date, how total soil Se and its bioavailability would respond to human disturbance or future environmental change is not yet clear, and associated controlling factors remain incompletely understood. Here, we collected soil samples (0–15 cm) from different land use/land cover types, including active cropland, grassland, shrubland, and secondary forest, in a Se-enriched area of Guangxi, southwest China. Total Se concentration and its potential bioavailability, as estimated by phosphate extractability, were investigated. Total soil Se concentration (Se_{total}) for all samples ranged from 220 to 1820 μg kg^{-1}, with an arithmetic average value of 676 ± 24 μg kg^{-1} (Mean ± SE, the same below). The concentration of phosphate extractable Se ($Se_{phosphate}$) varied between 1 and 257 μg kg^{-1}, with an arithmetic mean value of 79 ± 5 μg kg^{-1}, accounting for on average 13 ± 1% of the Se_{total}. Among the four land use/land cover types, Se_{total} and $Se_{phosphate}$ were generally more enriched in the secondary forest than those in the grassland and cropland. The content of soil organic carbon (SOC) was the overriding edaphic factor controlling the abundance and potential bioavailability of Se in topsoils. In addition, climatic variables such as mean annual precipitation and mean annual temperature

* Corresponding authors at: Institute of Subtropical Agriculture, Chinese Academy of Sciences, Changsha 410125, China (D. Li). Institute of Agricultural Resources and Environment, Guangxi Academy of Agricultural Sciences, Naning 530007, China (Y. Liu).
E-mail addresses: dejunli@isa.ac.cn (D. Li), liuyx27@163.com (Y. Liu).

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were also key factors affecting the abundance and potential bioavailability of soil Se. Our results suggest that changes in land use/land cover types may deeply influence Se biogeochemistry likely via alterations in soil properties, particularly SOC content.

1. Introduction

Selenium (Se) is an essential trace element for human and animal health (Rayman, 2000), and its deficiency has been identified to be causative for several endemic diseases, such as Kashin–Beck disease and Keshan disease (Tan et al., 2002). It is estimated that up to one in seven people worldwide (Haug et al., 2007; Jones et al., 2017), including approximately 39–61% of Chinese residents (Dinh et al., 2018), have insufficient dietary Se intake. Furthermore, as a critical constituent of selenoproteins, Se has important functions in anti-aging, enhancing immunity, and prolonging life-span (Foster and Zhang, 1995; Hao et al., 2016; Zhang et al., 2018). On the other hand, Se is an acute toxin and environmental pollutant as well, and the quantitative range between Se toxicity and deficiency for human body is rather narrow (Lenz and Lens, 2009; Kieliszek and Blazejak, 2016). Se in the soil is generally the primary source of human food Se (Navarro-Alarcon and Cabrera-Vique, 2008; Sun et al., 2010). For these reasons, soil Se abundance and Se bioavailability and its association with human and ecological health are of growing interest for both the scientific community and policy makers worldwide (Ullah et al., 2019).

Beyond total soil Se content, Se uptake by plants, i.e. bioavailability, depends more on its speciation and binding to soil fractions (Wang and Gao, 2001; Ellis and Salt, 2003; Sun et al., 2009). As such, the available fractions of Se, rather than its total content, serve as a more critical indicator of Se deficiency or excess in ecosystems (Sun et al., 2009). Water-soluble and exchangeable Se are major bioavailable forms of Se (Ellis and Salt, 2003). A wide variety of extractants have been attempted to displace Se that absorbed on soil solid surface (Keskinen et al., 2009), including phosphate buffer, $NaHCO_3$, $CaCl_2$, AB-DTPA, and NH_4F, etc. Among these extractants, phosphate buffer (K_2HPO_4-KH_2PO_4, pH = 7), which can extract soluble and exchangeable forms of Se^{6+}, Se^{4+} and Se^{-2}, is viewed as one of the most efficient extractants to assess Se bioavailability in soils (Bujdos et al., 2005; Zhao et al., 2005; Keskinen et al., 2009). Therefore, a better understanding of variation in phosphate extractable Se, besides total Se, and its governing factors can help more accurately evaluate ecosystem Se toxicity/deficiency.

The abundance and bioavailability of Se in soil can be modified by a wide range of factors, including plant, soil properties, and climate conditions (Fernández-Martínez and Charlet, 2009; Jones et al., 2017). Although Se has not been proven to be essential for plantae, it is considered to be beneficial for many plant species (Pilon-Smits et al., 2009). Plant can not only take up Se from soil, but also secrete kinds of organic acids (including oxalate, malate and citrate etc.) into soil, likely leading to changes in Se mobility and bioavailability through adsorption, complexation and reduction processes (Dinh et al., 2017). However, the effect of plant growth on soil Se bioavailability varies considerably among different plant species (Munier-Lamy et al., 2007). Edaphic variables, including soil organic matter (SOM), pH, clay content, amorphous Fe-and Al-(hydr) oxide, play important roles in regulating Se bioavailability (Tolu et al., 2014; Supriatin et al., 2015; Xing et al., 2015). For example, total soil Se content often correlates positively with contents of SOM, hydrous oxides of Fe and Al, and clay (De Temmerman et al., 2014; Tolu et al., 2014; Supriatin et al., 2015), but correlates negatively with soil pH (Buheaosier et al., 1995; Xing et al., 2015; Shao et al., 2018). In addition, the climate is also considered one of the most important factors affecting the biogeochemistry of Se in soil. It can affect soil Se abundance directly via atmospheric Se deposition (Blazina et al., 2014; Sun et al., 2016) or indirectly through mediating Se uptake by plants (Galeas et al., 2007). All these factors may act alone, or in combination with each other, in mediating soil Se abundance and bioavailability, and various factors have been shown to contribute differently to soil Se bioavailability, depending on soil type and climatic conditions (Nakamaru et al., 2005; Wang et al., 2011; Supriatin et al., 2016).

Land use/land cover change, which is an important aspect of global change, is always accompanied by changes in plant communities and strongly influences soil properties, particularly SOM (Sevink et al., 2005; Smith, 2008). Thus, land use/land cover change has important consequences for the biogeochemical cycles of essential macronutrients such as N, P, K, S, etc. (Zalba et al., 2017; Liu et al., 2018). Yet, land use/land cover impact on the geochemistry of essential trace elements has received relatively less exploration (Ivezic et al., 2015; Elrashidi et al., 2016; Jiang et al., 2017). As for Se, only a very limited number of studies have investigated the variation in soil Se concentration under different land use/land cover types (Ghorbani et al., 2015; Xing et al., 2015; Plak and Bartminski, 2017), and there is no consensus yet on the direction and magnitude of land use/land cover change effect on soil Se abundance and bioavailability. For instance, several authors found that Se is more enriched in forest soils than that in agricultural soils (Tolu et al., 2014; Xing et al., 2015), while the opposite pattern was also reported (Tan et al., 2002; Tuttle et al., 2014; Shang et al., 2015). Thus, to better predict human Se nutrition under the scenario of global change, a more robust understanding of land use/land cover impact on soil Se abundance and bioavailability is clearly needed.

Since 1950s, the Chinese government has implemented a series of ecological restoration programs, such as the Grain to Green Project, to guarantee the sustainable development of economy and society. In this context, land use/land cover change, e.g. the conversion of cropland to grassland or forest, is a common phenomenon occurring especially in the karst region of southwest China (Tong et al., 2018). As a result, diverse land use/land cover systems, including active cropland, grassland, shrubland, and secondary forest, coexist across this region owing to different duration after cropland abandonment. Hechi city in Guangxi, southwest China has been defined as a longevity region and a Se-enriched area as well (Lv et al., 2011). So far, the impact of land use/land cover conversion on soil Se abundance and bioavailability have not been well evaluated at a regional scale. The primary objectives of the current study were (i) to examine the influences of land use/land cover change on the total concentration and potential bioavailability of Se in topsoils in a karst Se-enrich area, and (ii) to identify the environmental factors that control the level and bioavailability of soil Se. Since our previous studies have shown that SOM content increased significantly following cropland abandonment and subsequent vegetation succession (Li et al., 2017a), we hypothesized that the abundance and potential bioavailability of Se in topsoils would also exhibit an increasing tendency after cropland abandonment, owing to Se retention by SOM.

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2. Materials and methods

2.1. Study area

This study was conducted in a typical karst area in the northwest of Guangxi Zhuang Autonomous Region, southwest China, including Huanjiang county and adjacent Duan county (23°40′N–25°25′N, 107°35′E–108°30′E). Most of the soils across this region are developed on carbonate rocks. The study region belongs to subtropical monsoon climate, with mean annual air temperature (MAT) ranging from 17.8 to 22.2 °C and mean annual precipitation (MAP) ranging from 1346 to 1640 mm.

2.2. Field sampling

Field sampling was conducted from the end of March to early June 2015. The sampling sites were selected based on land use/land cover types, including active cropland, grassland, shrubland, and secondary forest (Fig. 1). More detailed information about the geographic location and climate condition of each sampling site is presented in Table S1, in which MAP and MAT were obtained from the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (http://www.resdc.cn). The history of land use was obtained by interviewing local people. The active croplands were managed under corn-soybean rotation each year, and applied N, P, and K at rates of about 150, 60, and 120 kg ha^{-1} yr^{-1} in the form of compound fertilizer. Grassland, shrubland and secondary forest were converted from cropland under corn-soybean rotation and received no fertilization after cropland abandonment. Roughly, grassland, shrubland, and secondary forest were 5–10 years, 10–20 years, and 30–50 years since cropland abandonment, respectively.

In total, 125 sites were selected, with 27 croplands, 29 grasslands, 36 shrublands, and 33 secondary forests therein. At each site, a 20 × 20 m sampling plot was established and surface soil samples (0–15 cm) were collected with a stainless steel auger (5 cm in diameter). We sampled 10–15 soil cores randomly in each plot and then mixed it to form a composite sample. After transporting to the laboratory, the soils were air-dried at room temperature, disaggregated, and then passed through a 2 mm mesh sieve, with plant residues and stone fragments removed by hand-sorting. The 2 mm sieved soil samples were used to measure phosphate extractable Se, soil pH, size fractions, exchangeable Ca, Mg, Fe and Al. An aliquot of 2 mm sieved samples were further ground and passed through a 0.15 mm mesh sieve to measure total soil Se and soil organic carbon (SOC).

2.3. Chemical analyses

Total soil Se (Se_{total}) was analyzed 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 mixture of concentrated acid (10 ml, HNO_3:$HClO_4$ = 4:1 vol/vol) and allowed to stand overnight at room temperature. Then, the mixture was heated in an electrothermal furnace first at 60 °C for 30 min, and then at 120 °C for 30 min, and afterwards 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 quality 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.

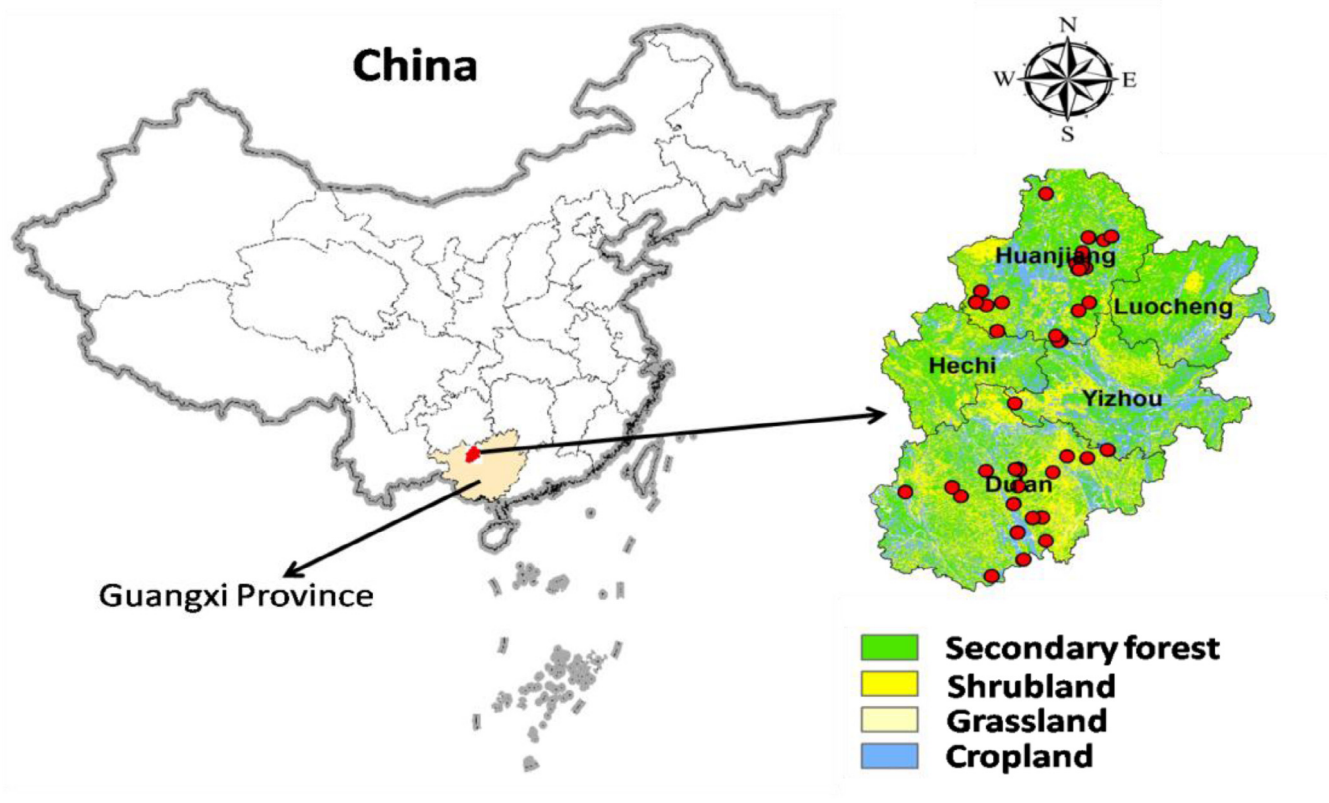

Fig. 1. Distribution of the sampling sites according to land use/land cover types.

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Phosphate exchangeable Se ($Se_{phosphate}$) was extracted by 0.1 mol L^{-1} (pH = 7.0) KH_2PO_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 mixture of concentrated acid (10 ml, HNO_3: $HClO_4$ = 4:1 vol/vol) was added to a 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-Qwater 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 $Se_{phosphate}$ concentrations were finally presented as μg Se kg^{-1} dry soil. The total Se concentration of the standard reference material was 651 ± 83 μg Se kg^{-1} (the certified values of 640 ± 140 μg Se kg^{-1}).

Soil gravimetric water content (GWC) was determined 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 determined by wet oxidation with potassium dichromate redox colorimetric method. Soil total phosphorus (TP) was digested with an H_2SO_4 + $HClO_4$ solution. Soil available phosphorus (AP) was extracted with 0.5 M $NaHCO_3$. TP and AP were determined using the molybdenum colorimetric method. Size fractions, i.e. clay (<2 μm), silt (2–50 μm), and sand (50–2000 μm) were analyzed on a laser diffraction particle size analyzer after decarbonation (Mastersizer, 2000, Malvern, UK). Exchangeable calcium (Ca_{ex}) and magnesium (Mg_{ex}) were measured after extracting with 1 mol L^{-1} ammonium acetate at pH 7.0 with inductively coupled plasma-atomic emission spectroscopy (ICP-AES). Fe and Al oxides 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 quantified by 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. One-way ANOVA with Tukey's post hoc test was used to examine the differenes in Se_{total} and $Se_{phosphate}$ among various land use/land cover types at $P < 0.05$ significance level. Pearson's correlation analysis was used to examine the relationships between environmental factors and Se_{total}, $Se_{phosphate}$, and the $Se_{phosphate}/Se_{total}$ ratio. Stepwise multiple linear regression approach was used to identify the strongest explanatory variables for Se_{total}, $Se_{phosphate}$ and the $Se_{phosphate}/Se_{total}$ ratio.

3. Results

3.1. Total soil Se concentration (Se_{total})

Se_{total} for all samples ranged from 220 to 1820 μg kg^{-1}, with an arithmetic average value of 676 ± 24 (mean ± SE) μg kg^{-1}. There was significant difference in Se_{total} among land use/land cover types ($P < 0.01$, Fig. 2). The average value of Se_{total} was highest in the secondary forest (818 ± 44 μg kg^{-1}). Furthermore, no significant difference in Se_{total} was found between grassland and shrubland, as well as between cropland and grassland (Fig. 2).

3.2. Phosphate extractable Se ($Se_{phosphate}$)

$Se_{phosphate}$ for all samples ranged from 1 to 257 μg kg^{-1}, with an arithmetic mean value of 79 ± 5 μg kg^{-1}. Land use/land cover had significant influences on $Se_{phosphate}$ ($P < 0.01$, Fig. 2). The average $Se_{phosphate}$ was significantly higher in the secondary forest (105 ± 10 μg kg^{-1}) as compared to the grassland (68 ± 8 μg kg^{-1}) and the cropland (55 ± 8 μg kg^{-1}). There was a strong linear relationship between $Se_{phosphate}$ and Se_{total} ($P < 0.001$, Fig. 3). On average, the proportion of $Se_{phosphate}$ to Se_{total} was 13%, ranging between 11% and 14% across the four land use/land cover types (Table S3).

3.3. Dependence of Se on environmental variables

Se_{total} was positively correlated with MAP, MAT, GWC, SOC, C:N ratio, TP, pH, Ca, Mg, Fe-Al oxides and sand contents, while was negatively correlated with AP. Similarly, $Se_{phosphate}$ was positively correlated with GWC, SOC, TP, pH, Ca and Mg contents, while was negatively correlated with AP. In addition, unlike Se_{total}, $Se_{phosphate}$ did not correlate with Fe-Al oxides and sand contents, but negatively correlate with MAP, MAT, and clay content (Table 1). Furthermore, $Se_{phosphate}/Se_{total}$ ratio was positively related to GWC, SOC, C:N ratio, Ca_{ex} and silt contents, and was negatively related to MAP, MAT, AP, and clay contents (Table 1).

Stepwise multiple linear regression analysis further demonstrated that SOC, MAP, and C:N ratio were the best three predictors for Se_{total}, explaining 55% of Se_{total} variation (Table 2). SOC, MAP, GWC, and MAT were the major explanatory variables for $Se_{phosphate}$ and explained 65% of $Se_{phosphate}$ variation (Table 2). In addition, MAP, MAT, SOC, and elevation were the main explanatory variables for the proportion of $Se_{phosphate}$ to Se_{total} and explained 57% of the variation in $Se_{phosphate}/Se_{total}$ ratio (Table 2).

4. Discussion

4.1. Soil Se abundance and bioavailability in the studied region

In the present study, Se_{total} for all samples (220–1820 μg kg^{-1}) is within the reported range of topsoil Se concentration across China (5–79080 μg kg^{-1}) (Dinh et al., 2018). The average Se_{total} in this region (676 ± 24 μg kg^{-1}) is 1.7 and 2.8 times greater than the global average value (400 μg Se kg^{-1}; (Fordyce, 2005)) and the national average value of China (239 μg Se kg^{-1}; (Tan et al., 2002)), respectively. For the cropland, the average Se_{total} is 531 ± 32 μg kg^{-1} (262–1070 μg kg^{-1}), which is almost 2 times higher than the national average value (269 μg Se kg^{-1}) for the cultivated soils in China (Tan et al., 2002). The result of frequency distribution showed that nearly 90% of the soil samples had Se_{total} concentrations over 400 μg kg^{-1} and none below 175 μg kg^{-1} (Fig. 1S). Therefore, the study area can be classified as a Se-rich area, according to the reference criteria proposed by Tan et al. (2002) for evaluating Se deficiency and Se excess in soil, i.e. Se-deficient (<125 μg kg^{-1}), Se-marginal (125–175 μg kg^{-1}), Se-sufficient (175–400 μg kg^{-1}), Se-rich (400–3000 μg kg^{-1}), and Se-excessive (>3000 μg kg^{-1}). Moreover, the proportion of potentially bioavailable Se, i.e. $Se_{phosphate}$, to total Se in the studied region is on average 12.9 ± 0.8%, which is higher than that reported for Enshi soils (<5%) (Yuan et al., 2012) and slightly lower or close to that reported for agricultural soils of Finland (~18%) (Keskinen et al., 2009; Keskinen et al., 2011), but much lower than that in Jainpur of Punjab, North-West India (>50%) (Bajaj et al., 2011).

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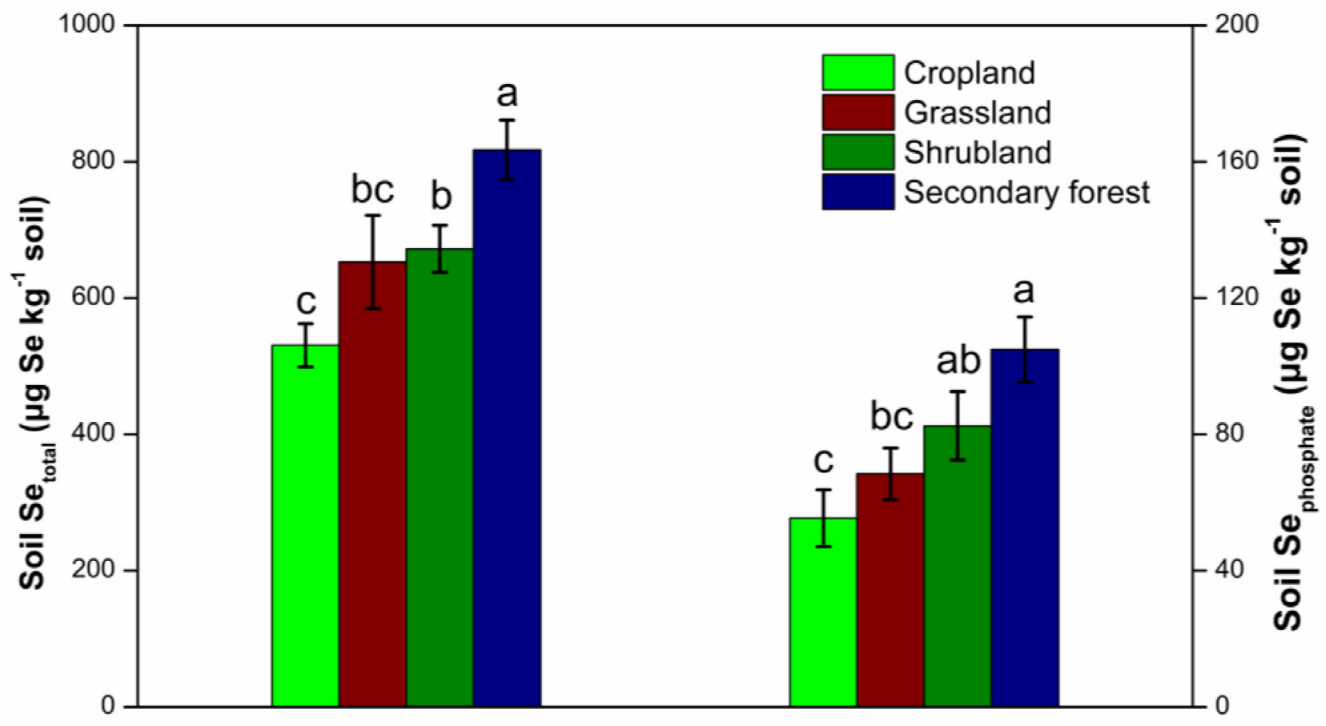

Fig. 2. Effects of land use/land cover on soil Se_{total} and $Se_{phosphate}$. The bars represent mean ± SE. Sample sizes are 27, 29, 36, and 33 for cropland, grassland, shrubland, and secondary forest, respectively. The bars with different letters denote significant difference among land use/land cover types at $P < 0.05$ (Tukey's post hoc test). More detailed information about the data is presented in Table S2.

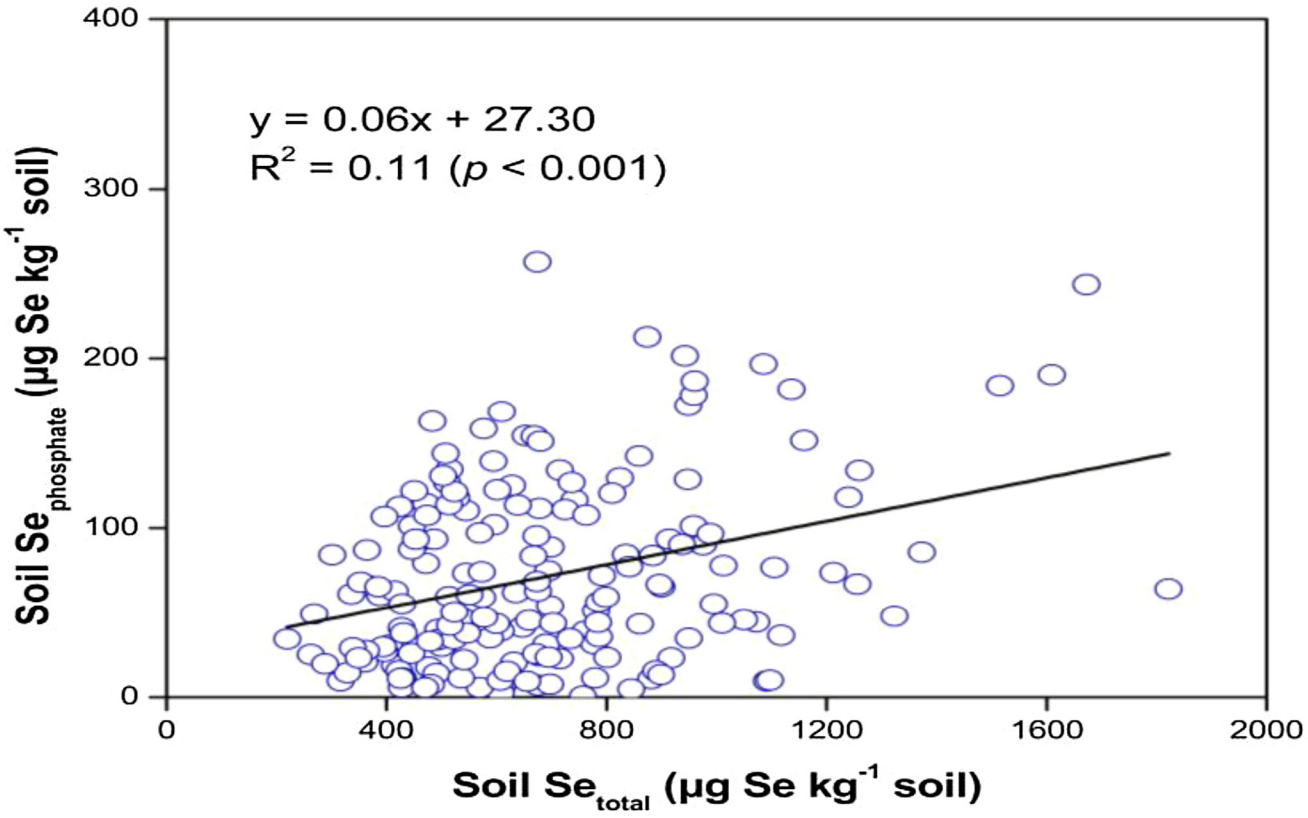

Fig. 3. Relationship between Se_{total} and $Se_{phosphate}$.

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Table 1
Pearson's correlation coefficients between environmental variables and Se_{total}, $Se_{phosphate}$, and the $Se_{phosphate}/Se_{total}$ ratio.

	Se_{total}	$Se_{phosphate}$	$Se_{phosphate}/Se_{total}$ ratio
MAP	0.32**	−0.55**	−0.70**
MAT	0.24**	−0.51**	−0.63**
GWC	0.39**	0.56**	0.31**
SOC	0.53**	0.67**	0.35**
C:N ratio	0.52**	0.46**	0.23**
pH	0.34**	0.26**	0.10
TP	0.37**	0.27**	0.03
AP	−0.26**	−0.25**	−0.19*
Ca	0.38**	0.45**	0.19*
Mg	0.29**	0.24**	0.10
Fe-Al oxides	0.29*	0.12	−0.03
Silt	−0.15	0.17	0.25**
Clay	−0.08	−0.29**	−0.25*
Sand	0.40**	0.16	−0.06

* Correlation is significant at $P < 0.05$.
** Correlation is significant at $P < 0.01$.

4.2. Land use/land cover impact on soil Se abundance and bioavailability

In the current study, both soil Se_{total} and $Se_{phosphate}$ showed a tendency to increase along the sequence of cropland, grassland, shrubland, and secondary forest (Fig. 2), keeping in line with our initial hypothesis. On average, Se_{total} and $Se_{phosphate}$ in the secondary forest were approximately 1.5 and 1.9 times greater than that in the cropland, respectively. This pattern is in line with some studies showing that the woodland soils have higher Se concentrations than the agricultural soils (Zhang et al., 2005; Tolu et al., 2014; Xing et al., 2015), but in contrast with Tan et al. (2002) who reported Se_{total} contents were higher in the cultivated soils than those in the natural soils. Significant variations in Se_{total} and $Se_{phosphate}$ among land use/land cover types can largely be ascribed to land use-induced changes in soil physicochemical properties and related anthropogenic activities. Lower concentrations of Se_{total} and $Se_{phosphate}$ in the cropland relative to the natural soils is partly a consequence of depletion of Se by continuous crop uptake and harvest. In addition, fertilization might be another contributor. This is supported by the findings of significantly higher TP and AP concentrations in the cropland owing to fertilization (Table S4), and a significant negative relationship between AP and $Se_{phosphate}$ (Table 1). Phosphate, which is commonly applied in cropping systems, can decrease selenite adsorption on soil solid surfaces due to competition for binding sites, thus increasing the plant-availability of Se as well as aggravating the potential risk of Se loss by leaching (Eich-Greatorex et al., 2010). Our results thus suggest that P fertilization, other than to increase Se availability, is also likely to accelerate Se resource depletion in agricultural systems in the long term.

Several mechanisms might be responsible for the significant Se enrichment in topsoils of the secondary forest. First and foremost, significantly higher concentration of SOC accumulates in the secondary forest, relative to the other four land use/land cover types (Table S4). Many studies have indicated that a significant proportion of Se in soil was bound to organic matter, accounting for up to 40–50% of total Se (Gustafsson and Johnsson, 1994; Oram et al., 2008; Qin et al., 2012). In low Se Dutch agricultural soils, over 80% of total Se are presented in organic forms (Supriatin et al., 2015). The possible approaches for binding of Se to organic matter include (1) formation of ternary complexes by Se, organic matter, and Fe-Al oxides (Coppin et al., 2009); (2) Creation of anoxic zones in the presence of organic matter that facilitate reduction processes (Tolu et al., 2014); and (3) soil organic matter provides lots of sorption sites, which promote complexation with Se (Winkel et al., 2012). A positive linear correlation between SOC and Se concentration in soils has been widely documented in the literature (Yamada et al. 2009, Roca-Perez et al. 2010, Tolu et al. 2014, Supriatin et al. 2015, Jones et al. 2017, Li et al. 2017b). Consistently, we also found strong positive correlations between SOC and Se_{total} and $Se_{phosphate}$ ($P < 0.01$, Table 1). Regression models also indicated that SOC was the strongest explanatory variable for variation in Se_{total} and $Se_{phosphate}$ (Table 2). That is, variations in environmental factors influencing the formation and stability of SOM may ultimately lead to changes in Se accumulation and bioavailability.

Second, soils in the secondary forest are much wetter than those in the other three land use/land cover types (Table S4), probably due to higher canopy density and lower air temperature in the secondary forest. Soil moisture is often associated with soil aeration, oxygen availability, and redox status (Sajedi et al., 2012), to a great extent, governing Se speciation and mobility in soils (Jayaweera and Biggar, 1996). Under well-aerated conditions, inorganic Se is mainly present in the form of selenate (SeO_4^{2-}) which is hardly adsorbed on soil solid surface, and is not only readily available to plants but vulnerable to loss by leaching. However, when under the environment of moderate redox, as in the case of the secondary forest in our study, the prevailing form of inorganic Se would be selenite (SeO_3^{2-}) which is firmly adsorbed on clay minerals and sesquioxides, and is not readily available to plants, thus largely retaining in the soil (Elrashidi et al., 1989). Furthermore, field moisture may have an indirect effect on Se enrichment by changing SOC stocks and stability. A recent research by Das et al. (2019) revealed that SOC mineralizability was much lower in high field moisture sites as compared to low field moisture sites, and correspondingly, the mean residence time of SOC was considerably longer in high field moisture sites relative to low field moisture sites. Moreover, biomethylation of Se can be strongly inhibited under reduced conditions, thus reducing Se loss via volatilization (Masscheleyn et al., 1990).

Third, the high humidity environment in the secondary forest enhances carbonate rock dissolution. In the present study, significant correlations were observed between Se_{total} and some bedrock-derived elements such as P, Ca, and Mg (Table 1), indicating that bedrock is an important source of Se in the soil. Increased bedrock dissolution will result in more Se release into the soil. This view is supported by Xie et al. (2010) and Zhang (2011) who found that the carbonate rock dissolution rate in the secondary forest is

Table 2
Results of stepwise multiple linear regression analyses for the dependence of Se_{total}, $Se_{phosphate}$ (μg kg^{-1}), and the $Se_{phosphate}/Se_{total}$ ratio on environmental variables.

Se_{total}				$Se_{phosphate}$				$Se_{phosphate}/Se_{total}$ ratio			
Explanatory variable	Coefficient	Model R^2	*P* value	Explanatory variable	Coefficient	Model R^2	*P* value	Explanatory variable	Coefficient	Model R^2	*P* value
SOC	0.48	0.31	0.000	SOC	0.46	0.46	0.000	MAP	−0.45	0.49	0.000
MAP	0.46	0.52	0.000	MAP	−0.30	0.62	0.000	MAT	−0.54	0.54	0.001
C:N ratio	0.25	0.55	0.009	GWC	0.18	0.64	0.020	SOC	0.15	0.56	0.016
				MAT	−0.15	0.65	0.048	Elevation	−0.28	0.57	0.036

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

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generally higher relative to the cropland and other land use/land cover types in the karst region of southwest China.

4.3. Factors influencing soil Se abundance and bioavailability

Parent bedrock has traditionally been considered the primary source of Se in soil. Furthermore, parent material can also exert great controls on Se biogeochemical cycling by influencing soil physicochemical properties, including pH, texture, ion composition (P, Ca, etc.), metal oxides, and SOM stock (Nakos 1984, Barre et al. 2017). In the study region, Ca that released from carbonate bedrocks can promote the stabilization of SOM (Li et al. 2017a), thus indirectly mediating Se bioavailability. As pedogenic processes progress, the impact of soil parent material on Se distribution weakens gradually, while the role of soil physicochemical properties strengthens gradually. For example, Gabos et al. (2014) found that Se levels in soil were independent of Se occurrence in parent bedrock, but were highly related to soil physicochemical properties such as SOM and Al-Fe oxides.

Atmospheric Se inputs via precipitation is another important source of soil Se, and in some cases it plays a dominant role, over parent bedrocks, in controlling the distribution of Se in topsoils (Blazina et al., 2014; Sun et al., 2016). The level of Se in rainwater of China is usually in the range of 0.1–0.2 μg Se L^{-1}, and Se deposition in subtropical China is estimated up to 160–320 μg m^{-2} yr^{-1} (Sun et al., 2016). In the current study, MAP was significantly and positively correlated with Se_{total} (Tables 1 and 2), confirming the potential contribution of atomospheric Se input to Se accumulation in soil. In addition, climatic variables always have a far-reaching impact on SOC stocks (Callesen et al. 2003), indirectly affecting Se retention in soil. Furthermore, a significant and negative correlation was found between climatic variables (i.e. MAP and MAT) and $Se_{phosphate}$ and its proportion of Se_{total} (Tables 1 and 2), indicating that increases in MAP and MAT would cause a decline in the bioavailability of Se in soils. Similarly, based on results of model simulation, Jones et al. (2017) suggested that the prevalence of Se deficiency would increase under future climate change. Overall, our results highlight the importance of taking climatic conditions into account when studying Se geochemistry especially on large scales.

Clay and Fe-Al oxides has been considered important edaphic variables contributing to Se retention in the soil, due to their relative high surface area and abundant binding sites (Matos et al., 2017). Consistently, we found a significant correlation between Se_{total} and Fe-Al oxides ($P < 0.05$, Table 1). In general, fine grains often have greater exchangeable capacity and stronger adsorption than coarse grains, leading to higher Se retention in clayed soils (Gabos et al., 2014; Shao et al., 2018). On the other hand, no significant correlation was found between Se_{total} and clay content (Table 1), following others that have shown Se_{total} was independent of clay content (Levesque, 1974; Zhang et al., 2008; Sun et al., 2010).

5. Conclusions

Our results demonstrated that land use/land cover change had significant effects on the total concentration and potential bioavailability of Se in topsoils in a karst Se-enriched area in Guangxi, southwest China. Both Se_{total} and $Se_{phosphate}$ were generally higher in the secondary forest than those in the cropland and grassland. SOC was the overriding edaphic variable controlling soil Se abundance and bioavailability. Furthermore, soil Se abundance and bioavailability could be strongly influenced by climatic variables such as MAP and MAT. In addition, high P status, coinciding with lower organic matter content, may be responsible for less Se retention in cropland. Our findings suggest that the abundance and potential bioavailability of Se in topsoil would substantially increase after cropland abandonment in the karst Se-enriched area of southwest China, mainly due to SOM accumulation. In the long run, there is a high risk of accelerated Se resource depletion in intensified agricultural systems, especially for those with high input of P fertilizers.

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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Appendix A. Supplementary data

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