Research Article

Sorption and Environmental Risks of Phosphorus in Subtropical Forest Soils

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Phosphorus (P) is one of the key limiting factors for the growth of forests and their net primary productivity in subtropical forest ecosystems. Phosphorus leaching of the forest soil to the catchment and groundwater in karst region is the main source of water eutrophication. Strong P sorption capacity of minerals is generally assumed to be a key driver of P leaching in subtropical ecosystems which varies among different soil types. Here, we estimated P adsorption capacity of the O/A and AB horizon in both limestone soil and red soil of subtropical forests by fitting the Langmuir and Freundlich isotherm to investigate the potential environmental risks of P. The maximum P sorption capacity (Q_m), P sorption constant (K_L), P sorption index (PSI), degree of P saturation (DPS), and maximum buffer capacity (MBC) were calculated. The results indicate that Q_m of the O/A horizon in both soils were similar. Comparing these two soils, the red soil had a higher K_L and MBC in the AB horizon; Q_m of limestone soil was larger but K_L was lower, indicating that the adsorption capacity of limestone soil was weaker and MBC was lower. There was no significant difference in PSI between the two soils. The DPS values of both soils were below 1.1%, indicating that P saturation is low in both subtropical forest soils due to the lack of marked anthropogenic disturbance. In the O/A horizon, P saturation associated with available P (DPS_{M3} and DPS_{Olsen}) and that associated with P in the Fe-Al bound state ($DPS_{citrate}$) were higher in the red soil than in the limestone soil. DPS did not differ significantly in the AB horizon, except for higher DPS_{M3} and $DPS_{citrate}$ in the red soil. The findings highlight the influence of the soil types on P adsorption. The P adsorption and buffering of red soils were higher than those of limestone soils, indicating a lower risk of P leaching in red subtropical forest soils.

1. Introduction

Phosphorus is one of the key limiting factors for the growth of forest trees and their net primary productivity in natural forest ecosystems, and it exists mainly in the form of organic P (P_o) and inorganic P (P_i) in primary and secondary minerals. Phosphorus that can be directly absorbed and used by plants is mainly phosphate in the ionic state [1]. Due to the sorption and fixation of P by soil minerals, phosphate accounts for only a few fraction of the soil P pool, resulting

in a generally low availability of P in soils [2, 3]. This is especially the case in subtropical forest ecosystems, due to a high degree of soil weathering and more pronounced fixation of P by iron and aluminum oxides. Most of P in soil is fixed in a stable form that is difficult to be absorbed and utilized by plants, so P limitation in subtropical forest ecosystems is more severe than in other ecosystems [4]. The input of P in forest ecosystems is mainly from slow weathering of the parent bedrock [5] and in a shorter time scale from the decomposition of litter and mineralization of organic

TABLE 1: Physiochemical properties in soil of O/A and AB horizons of the red soil and limestone soil.

	Red soil		Limestone soil	
	O/A	AB	O/A	AB
pH_{H2O} (1 : 2.5)	4.46 ± 0.12^{aB}	4.33 ± 0.06^{aB}	6.75 ± 0.31^{aA}	6.65 ± 0.24^{aA}
TOC (g·kg^{-1})	37.64 ± 2.19^{aA}	13.14 ± 0.61^{bB}	46.58 ± 7.61^{aA}	24.77 ± 5.95^{aA}
TN (g·kg^{-1})	2.74 ± 0.17^{aA}	1.11 ± 0.06^{bA}	4.61 ± 0.67^{aA}	2.71 ± 0.59^{aA}
TP (mg·kg^{-1})	353.32 ± 17.25^{aB}	299.06 ± 15.04^{aB}	602.73 ± 79.36^{aA}	457.93 ± 24.29^{aA}
Ca (g·kg^{-1})	0.07 ± 0.01^{aB}	0.06 ± 0.01^{aB}	2.19 ± 0.06^{aA}	2.40 ± 0.02^{aA}
Mg (g·kg^{-1})	0.24 ± 0.08^{aA}	0.25 ± 0.09^{aA}	0.48 ± 0.03^{aA}	0.50 ± 0.03^{aA}

Note: (1) average ± standard error (SE, $n = 3$); (2) different lowercase letters indicate significant differences ($p < 0.05$) between the different soil horizons within one site. Different uppercase letters indicate significant differences ($p < 0.05$) of the same soil horizon between the red soil and limestone soil.

matter. The P output is mainly from the sorption and utilization of P by plants, P loss due to leaching, and surface loss due to surface runoff and soil erosion. Soil P fractions and available P are controlled by processes including precipitation-solubilization of phosphate, mineralization-immobilization by microorganisms, and adsorption-desorption of adsorbed P_i, which in turn affects the supply of soil available P and the risk of P leaching. Therefore, P transformation in forest soils is an important process in the P cycle, which has strong ecological and environmental implications. In different soil types, P fixation capacity is regulated by environmental physicochemical factors, and the surplus P may be leached into water with runoff, causing water eutrophication [6, 7]. Phosphorus speciation in soil determines P availability and P leaching. Soil DPS is an important predictor of soil P mobility and effectiveness, which integrates the soil P fixation capacity and P content, and can be applied to assess the soil P environmental threshold as well as to determine the risk of soil P loss [8, 9]. The larger the DPS, the higher the soil contains dissolved state, the lower the P sorption capacity, and the higher the risk of soil P leaching.

In this study, we investigated the P adsorption and potential environmental risk in limestone and red soils which are the main soil types in subtropical forests of China. The Langmuir equation and Freundlich equation were used to fit the P sorption curve, and Q_m, MBC, and PSI of bulk soil were obtained. The fitted correlation parameters with the experimental data combined with DPS and other indicators were adapted to predict the potential environmental risk of P leaching of limestone and red soils.

2. Materials and Methods

2.1. Study Area.
The study area is located in Guilin City, South China, which is located at low latitude and belongs to the subtropical monsoon climate zone with mild climate, abundant rainfall, and basically the same season of rain and heat; the average annual temperature is about 19.1°C, the average annual rainfall is about 1887.6 mm, the average annual relative humidity is about 76%, and the average annual sunshine duration is about 1447.1 hours. In this study, Ludiyan (25°13′18.2″N, 110°14′53.3″E) and Guilin National Forest Park (GNFP, 25°19′06.6″N, 110°15′19.9″E) were selected for sampling, which represent karst lime-

stone soil and acidic red soil, respectively. The linear distance between the two sites is about 11 km. Ludiyan is mainly a mixed evergreen broad-leaved forest, and the soil type is limestone soil (LS) with pH range of 6.65~6.75. Guilin National Forest Park is mainly a mixed coniferous forest, and the soil type is typical strongly leached acidic red soil (RS) with pH range of 4.33~4.46 (Table 1).

2.2. Sample Collection.
Three plots within 20×20 m were randomly selected at each of the two sites for soil sample collection, and three replicated soil samples were taken at each site. At each site, soil was collected in the O/A horizon (0-10 cm and 0-12 cm for Ludiyan and GNFP, respectively) and AB horizon (10-22 cm and 12-33 cm for Ludiyan and GNFP, respectively) according to the natural stratification of the soil profile. The collected soil samples were air-dried, passed through a 2 mm sieve to remove the roots, prior to determination of the physical and chemical properties.

2.3. Soil Phosphorus Isothermal Adsorption.
Eight soil samples of 2.0 g were weighed into 50 mL centrifuge tubes, and 25 mL of 0.01 mol·L^{-1} CaCl$_2$ solution containing 0, 10, 20, 40, 80, 120, 150, and 200 mg P·L^{-1} of P was added, along with 2 drops of toluene to inhibit microbial activity. After shaking at room temperature for 24 h at 25°C, the samples were separated by centrifugation at 4000 r·min^{-1} for 15 min, and the supernatant used for analyzing the concentration of P was filtrated and aspirated. To develop the blue color, molybdenum blue reagent was added, and the volume was made up with deionized water. After 30 min, the absorbance was read at 880 nm using an enzyme-labeled instrument [10].

The phosphorus adsorption was calculated as follows.

$$Q_m = (C_0 - C_b) \times \frac{V}{m}, \quad (1)$$

where Q_m is the adsorption amount (mg·kg^{-1}), C_0 and C_b are the added and equilibrium concentrations of P in solution (mg·L^{-1}), respectively, V is the volume of solution (mL), and m is the mass of soil.

The Freundlich and Langmuir equations were used to fit the soil P adsorption. In general, the Langmuir equation describes the physical sorption of a single molecular layer, while the Freundlich equation assumes that the sorption between the adsorbent molecules and the adsorbent should

be a nonuniform sorption of multiple molecules. Both Langmuir and Freundlich equations are suitable for describing soil P isothermal sorption curves, but the fit varies depending on soil type.

The Freundlich equation is given by

$$q = K_F c^{1/n}, \tag{2}$$

where q is the amount of P adsorbed by the soil ($mg \cdot kg^{-1}$), c is the mass concentration of the sorbent in the liquid ($mg \cdot kg^{-1}$), K_F is the capacity parameter representing the sorption capacity of soil for P ($mg \cdot kg^{-1}$), and $1/n$ is the sorption intensity factor ($L \cdot kg^{-1}$).

The Langmuir equation is given by

$$\frac{C_e}{q} = \frac{1}{Q_m \times K_L} + \frac{C_e}{Q_m}, \tag{3}$$

where the parameter C_e is the concentration of P in the equilibrium solution ($mg \cdot L^{-1}$), Q_m is the maximum sorption of P by the soil ($mg \cdot kg^{-1}$), which is used to describe the size of the soil P pool, q is the sorption of P by the soil ($mg \cdot kg^{-1}$), and K_L is the soil sorption affinity constant, which indicates the strength of P sorption by the soil. K_L indicates that the sorption of P by the soil can proceed spontaneously at ambient temperature. A greater K_L value suggests that the P sorption capacity is stronger. The maximum buffer capacity (MBC) of soil P can be calculated according to the Langmuir equation. The MBC reflects the P sorption of the soil and provides an index for evaluating P supply capacity of the soil; the larger the MBC value, the greater the P fixation capacity of the soil.

The MBC is calculated by the following equation:

$$MBC = K_L \times Q_m. \tag{4}$$

Soil phosphorus sorption index (PSI) is calculated by the following equation [11]:

$$PSI = \frac{X}{\log C}, \tag{5}$$

where the unit of X is the soil P sorption ($mg \cdot kg^{-1}$) and C is the equilibrium concentration of P in solution ($mg \cdot L^{-1}$).

The Mehlich-3 extractant was used to extract P (P_{M3}), iron (Fe_{M3}), and aluminum (Al_{M3}) from the soil to calculate DPS_{M3} [12]. The concentration of the soil-available P (P_{Olsen}), water-soluble P (P_{CaCl2}), calcium-related P (P_{HCl}), and iron-aluminum-related P ($P_{citrate}$) was determined by using different extractants to calculate the corresponding DPS_{Olsen}, DPS_{CaCl2}, DPS_{HCl}, and $DPS_{citrate}$ [12]. P_{CaCl2} is a water-soluble P that can mimic the salt status in soil solution, is positively correlated with bioavailable P in runoff or leachate water, and is an important indicator for assessing P leaching [3, 4]; P_{M3} and P_{Olsen} are both soil bioavailable P forms that are closely related to plant P uptake, and Pizzeghello et al. [13] found a significant correlation between the two P forms, where 58% to 98% of P_{Olsen} can be converted to water-soluble P [14], and the combination of P_{Olsen} and P_{CaCl2} can be used to evaluate P availability in environmental

soil solutions [15]. $P_{citrate}$ is the P associated with Fe and Al and is an important pool for supplying bioavailable P in acidic red soils. Most of the P_{HCl} is associated with calcium phosphate salts mainly including insoluble P such as octacalcium phosphate and decacalcium phosphate. In limestone soils, P sequestration is achieved mainly by precipitation with calcium phosphate or coprecipitation with carbonate [16].

Degree of phosphorus saturation (%) was calculated as follows [17–19]:

$$DPS_{M3}(\%) = \frac{P_{M3}}{(Fe_{M3} + Al_{M3})} \times 100,$$

$$DPS_{Olsen}(\%) = \frac{P_{Olsen}}{(P_{Olsen} + Q_m)} \times 100,$$

$$DPS_{CaCl2}(\%) = \frac{P_{CaCl2}}{(P_{CaCl2} + Q_m)} \times 100, \tag{6}$$

$$DPS_{HCl}(\%) = \frac{P_{HCl}}{(P_{HCl} + Q_m)} \times 100,$$

$$DPS_{citrate}(\%) = \frac{P_{citrate}}{(P_{HCl} + Q_m)} \times 100.$$

2.4. Statistical Analysis. The data in the study presented as average ± standard error (SE; $n = 3$) were analyzed using SPSS Statistics 25.0 (V. 2018, IBM, Armonk, NY, USA). One-way ANOVA (Fisher test, $p < 0.05$) was used to compare the mean concentrations of the physiochemical properties between the two soil types. All statistical analysis in this study was performed using the Origin 2018 software for Windows.

3. Results and Discussion

3.1. Soil Phosphorus Isothermal Sorption Curve. The isothermal sorption curves of both soil P were well fitted by Langmuir (r^2 between 0.91 and 0.97) and Freundlich equations (r^2 between 0.91 and 0.98) (Table 1). By comparing the correlation coefficients r^2 of the two equations, it was found that for red soils, the fit with the Langmuir equation was higher than that with the Freundlich equation or both were comparable, while for limestone soils, the fit of the two equations was comparable but tended to perform slightly better with the Freundlich equation. The isothermal sorption curves fitted with the Langmuir equation and Freundlich equation for P in different soil horizons of different soils, respectively, are shown in Figure 1. Soil P sorption increased with increasing P concentration. At low P concentrations, different soil P sorption curves exhibited a larger slope, and the slope of the soil P sorption curve in the O/A horizon of limestone soil was relatively the lowest. Q_m, K_L, and MBC, which characterize the P sorption capacity in soils, were obtained by the Langmuir equation. Q_m represents the size of the soil P pool, and a larger value indicates that more P can be absorbed. The differences in Q_m in red soil (981.87-1134.98 $mg \cdot kg^{-1}$) and limestone soil (1036.41-1110.29 $mg \cdot kg^{-1}$) were not significant, and both showed that the O/A horizon was larger than the AB horizon
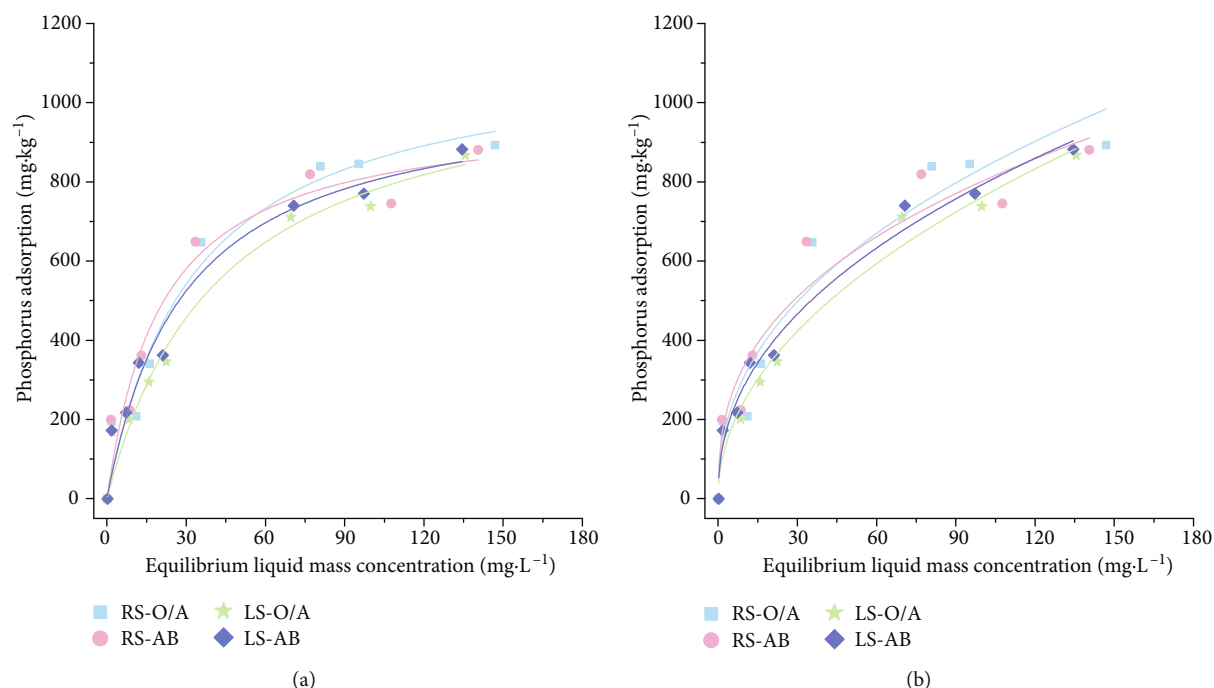
FIGURE 1: Fitting curve of (a) Langmuir and (b) Freundlich model for the O/A and AB horizons of the red soil (RS) and limestone soil (LS).

TABLE 2: Parameter of adsorption isotherm fitting curve and PSI of the O/A and AB horizons of the red soil (RS) and limestone soil (LS).

Soil types	Langmuir equation					Freundlich equation		
	Q_m (mg·kg^{-1})	K_L	r^2	MBC (mg·kg^{-1})	PSI (mg·kg^{-1})	K_F	$1/n$	r^2
RS-O/A	1134.98	0.030	0.96	34.56	412.07	115.46	0.03	0.94
RS-AB	981.87	0.048	0.96	47.32	410.16	141.87	0.05	0.94
LS-O/A	1110.29	0.023	0.97	25.90	407.63	80.21	0.02	0.98
LS-AB	1036.41	0.034	0.91	35.43	415.84	112.11	0.03	0.91

(Table 1). The K_L value of red soil (0.030-0.048) was higher than that of limestone soil (0.023-0.034) and was higher in the AB horizon than in the O/A horizon in both soils (Table 2). The MBC values were higher in red soils (34.56-47.32 mg·kg^{-1}) than in limestone soils (25.90-35.43 mg·kg^{-1}) and showed AB horizon > O/A horizon in both soils. K_F, the sorption parameter obtained from the Freundlich fitted curve, reflects the soil sorption capacity and the magnitude of the adsorption affinity. A larger value of K_F indicates stronger adsorption capacity, and its trend is similar to that of K_L of the Langmuir equation (Table 1). n is a constant, which is related to the homogeneity of soil surface sorption capacity, and the more the $1/n$ value tends to 0, the stronger the soil sorption surface heterogeneity, and $1/n$ values between 0.1 and 0.5 indicate that the bulk soil have strong adsorption capacity [12]. In both soils, $1/n$ values showed AB horizon > O/A horizon, while $1/n$ values were slightly larger (0.02-0.03) in red soils (0.03-0.05) than in limestone soils, indicating stronger spatial heterogeneity of the adsorption surface in red soils than in limestone soils. There was no significant difference in PSI between the two soil types, which varied between 407.63 and 415.84 mg·kg^{-1}.

3.2. Soil Phosphorus Adsorption Saturation. Extractable P concentrations with different extractants in red and limestone soils are shown in Figure 2. In both soils, the concentration of P_{CaCl2} was low (red soil: 1.56-1.72 mg·kg^{-1} and limestone soil: 2.55-2.58 mg·kg^{-1}) (Figure 1); the P_{CaCl2} concentration in limestone soil was significantly higher than in red soil. In red soils, the concentration of P_{M3} (2.49-7.36 mg·kg^{-1}) was higher than that in limestone soils (0.32-1.32 mg·kg^{-1}); in the O/A horizon, the concentration of P_{Olsen} in red soils (4.99 mg·kg^{-1}) was significantly higher than that in limestone soils (2.84 mg·kg^{-1}), while there was no significant difference between the two soils in the AB horizon. In red soils, $P_{citrate}$ was the dominant P form, and in the O/A horizon, the content of $P_{citrate}$ (12.22 mg·kg^{-1}) was not significantly different from P_{HCl} (11.45 mg·kg^{-1}), but significantly higher than other extractable P contents; in the AB horizon, the content of $P_{citrate}$ (10.08 mg·kg^{-1}) was significantly higher than other extractable P contents. In limestone soils, P_{HCl} was the dominant P form, and its content in the O/A horizon (9.66 mg·kg^{-1}) was significantly higher than that of other extractable P. In both soils, except for $P_{citrate}$ in limestone soils, all other P fraction concentrations showed O/A horizon > AB horizon (Figure 2).
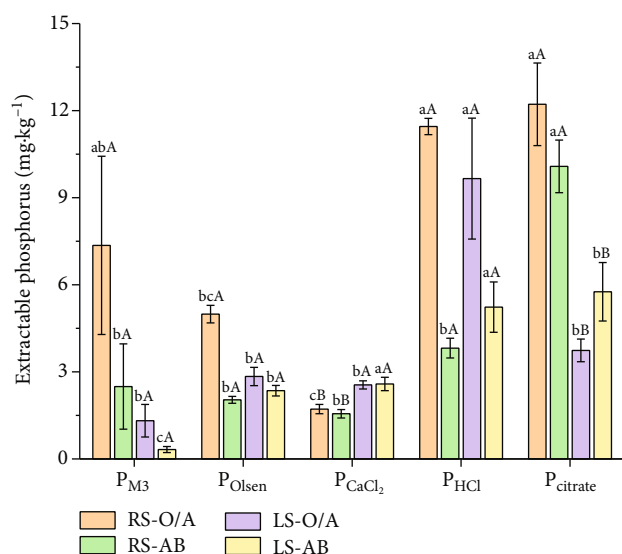
FIGURE 2: Distribution of extractable P in the bulk soils of O/A and AB horizons of red soil (RS) and limestone soil (LS). Different lowercase letters indicate significant differences ($p < 0.05$) among the P fractions from the same horizon of the same soil; different uppercase letters indicate significant differences ($p < 0.05$) of the same P fractions between the same horizon of the red soil and limestone soil. Error bars are standard error ($n = 3$).
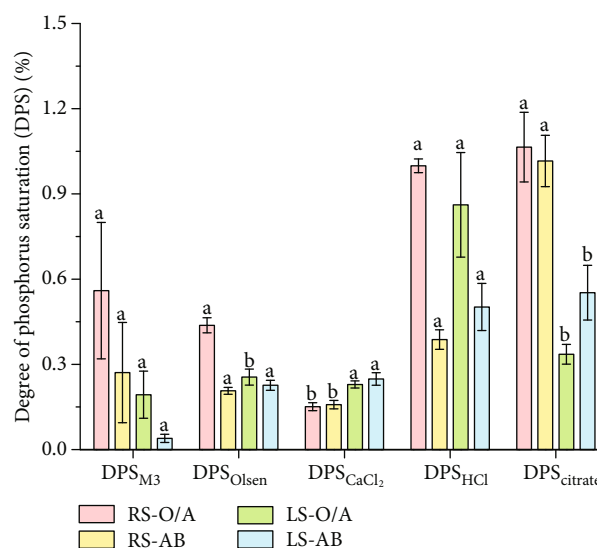

FIGURE 3: Degree of P saturation in the O/A horizon and the AB horizon of red soil (RS) and limestone soil (LS). Different lowercase letters indicate significant differences ($p < 0.05$) between the same soil horizon of the red and limestone soil. Error bars are standard error ($n = 3$).

In the O/A horizon, the values of DPS_{M3}, DPS_{Olsen}, DPS_{HCl}, and $DPS_{citrate}$ in red soils (0.56%, 0.44%, 1.00%, and 1.06%, respectively) were higher than those in limestone soils (0.19%, 0.26%, 0.86%, and 0.26%, respectively), while the values of DPS_{CaCl2} (0.15%) were lower than those of limestone soils (0.23%); in the AB horizon, the values of DPS_{CaCl2}, DPS_{HCl}, and DPS_{Olsen} were higher in limestone soils (0.25%, 0.50%, and 0.23%) than in red soils (0.16%, 0.39%, and 0.21%) (Figure 3).

3.3. Sorption Characteristics and Release Risk of Soil Phosphorus. In this study, the Q_m values of the two soils were not significantly different, and Q_m and K_L values were slightly higher in the O/A horizon of the red soil than those of limestone soil (Table 2), indicating that the O/A horizon of the red soil has higher P capacity and stronger adsorption capacity. The MBC of red soil was higher than that of limestone soil when Q_m of the two soils were similar, and the P adsorbed by red soil was more easily absorbed by plants and the total P pool of red soil. By researching P sorption in Irish soils, Daly et al. [9] reported that Q_m varied between 467 and 343 mg·kg^{-1} in red soils. Zhou and Li [20] studied P sorption in limestone soils as well as in limestone bedrock and found that Q_m varied between 591 and 5556 mg·kg^{-1}. The total P pool and inorganic P forms other than Fe-P were lower in red soils than in limestone soils, but the supply of P was higher than in limestone soils because of the faster organic P conversion process. In contrast, in the AB horizon, the Q_m values of limestone soils were higher than those of red soils. The K_L values were lower than those of red soils, which may be due to the large amount of calcium carbonate in the parent material of limestone soils that coprecipitate

with P [21]. It indicates that the limestone soils have a large P capacity and weak P adsorption capacity and a potential risk of P leaching.

In the O/A horizon, P_{M3}, P_{Olsen}, and $P_{citrate}$ had higher dissolved states in red soils, and in the AB horizon, limestone soils had higher levels of P_{Olsen}, P_{HCl}, and P_{CaCl2}. P_{Olsen} was at deficient to very deficient levels in both soils, indicating that bioavailable P was low. The DPS values in all soils in this study were less than 1.10%, which is lower than the DPS values (1.40% to 7.20%) in forest soils after revegetation of fire-burned sites [22, 23], indicating that the forest soils in this study are less disturbed by humans and have a lower risk of soil P leaching. Comparing the two soils, the values of DPS_{M3}, DPS_{Olsen}, DPS_{HCl}, and $DPS_{citrate}$ in the red soil O/A horizon were higher than those in the limestone soil. In the AB horizon, all but $DPS_{citrate}$ and DPS_{M3} in the red soil were lower than those in the limestone soil (Figure 3). Combined with the small difference in PSI between the two soils, it indicates that the P fixation capacity of two soils was at a similar level. The DPS is considered as an indicator to identify soils with a high risk of P release [24, 25]; the magnitude of its value is influenced by a combination of soil physicochemical properties [26–28]. The threshold value is not uniform, and some studies have shown a significant impact on the quality of the water environment when the DPS is greater than 15.00% [29]. The others have shown that, when the DPS exceeds 25.00%, soils tend to have high levels of desorbable P and the risk of P leaching is greater [30].

4. Conclusion

Comparing the Q_m, K_L, and MBC values of the subtropical forest soils, the maximum P sorption in the O/A horizon

of two soils was similar. While in the AB horizon, the maximum P sorption of limestone soils was higher, but the adsorption capacity was weaker. Both forest soils had low DPS due to weaker anthropogenic disturbance and have overall weak risk of soil P loss. Comparing the two soils, in the O/A horizon, the red soil DPS_{M3}, DPS_{Olsen}, and $DPS_{citrate}$ were higher than those of the limestone soil, and in the AB horizon, the difference in P saturation was not significant, except for the higher DPS_{M3} and $DPS_{citrate}$ in the red soil. Hence, the risk of P leaching in red soil is lower than that in limestone subtropical forest soil.

Data Availability

All the data used to support the findings of this study are included within the article.

Additional Points

Highlights. Phosphorus (P) sorption and environmental risks of subtropical forest soils differ with lithology. The P sorption and buffering of red soils were higher than those of limestone soils. Compared to limestone soil, red soil has lower potential environmental risk of P leaching.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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