



## Immobilizing soil exogenous lead using rock phosphate

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### Abstract

The effect of rock phosphate (PR) on exogenous forms of Pb in yellow cinnamon soil (S1) and latosol soil (S2) were studied by Tessier's sequential extraction procedure after PR was applied into the two soils at four levels of P (0, 50, 500 and 2000 mg kg<sup>-1</sup> soil). The results indicated that the content of exchangeable Pb in two soils decreased significantly with the rising of PR amount, which reached the minimum of 2.95 mg kg<sup>-1</sup> for yellow cinnamon soil and 3.57 mg kg<sup>-1</sup> for latosol soil under the treatment of P 2000 mg kg<sup>-1</sup> soil (PR3). The content of carbonate-bound Pb changed marginally. With the increase of PR amount in the two soils, the content of Fe-Mn oxide-bound Pb finished while the content of organic compound-bound Pb increased. The residual fraction of Pb in the two soils reached the maximum under the treatment of P 2000 mg kg<sup>-1</sup> soil, 38.5 mg kg<sup>-1</sup> for yellow cinnamon soil and 66.1 mg kg<sup>-1</sup> for latosol soil. After the two soils being applied with testing PR, the content of exogenous exchangeable Pb decreased and residual fraction was augmented conversely for both soils. In conclusion, the results reflected that Pb was immobilized effectively by PR in the two soils.

**Key words:** Rock phosphate, lead forms, soil.

### Introduction

The heavy metal contamination is one of the primary types of soil pollution. According to statistics, more than an average of 20 million tons of heavy metals is discharged in the world annually, among which Pb runs up to 5 million tons<sup>1</sup>. In general, there are three typical measures to renovate soil heavy metal pollution, namely physical, biological and chemical methods. The physical method is easy to operate but costly, therefore, only available in narrow field. Microbial and botanic amending approach is cheap and friendly to environment, but it takes a long time to put into effect since the organisms as the object contain small quantity of biomass, and as a result, methods is unsuitable for farmland soil. The *in situ* chemical remediation is reasonable for large area owing to easily-manipulative and little-costly advantages, although it cannot remove heavy metals in the soil. In addition, the priority is to choose the appropriate amendment for chemical method.

During the past dozen years, some researchers proposed phosphorus as the amendment to renovate lead-contaminated soil. This technique is of particular interest because of its cost-effectiveness and it is less disruptive to soil structure compared to the common method of removing pollutants from contaminated soil<sup>2</sup>. The resources reserve of Chinese phosphorus ore is abundant, but most of it is low-grade and also difficult for mineral processing. Hence, most of the phosphorus ore cannot play a role in the phosphate fertilizer industry<sup>3</sup>. It is a practical and feasible method to apply the middle or low-grade rock phosphate to amend a heavy metal contaminated soil directly<sup>4,5</sup>.

Lead is a highly toxic heavy metal, which can be released into the environment via numerous routes<sup>6</sup>. Anthropogenic activities,

primarily associated with industrial processes, manufacturing and the disposal of domestic and industrial waste materials, are the major source of lead contamination of soils<sup>7</sup>. Studies have shown that the bioavailability of lead depends on the form and solubility of lead in the contaminated soil<sup>4</sup>. The form of lead in the contaminated soil is an important factor which can affect the toxicity and also can decide the mobility and bioavailability of lead. Lead phosphates, particularly pyromorphites, are one of the most stable forms of Pb in soils under a wide range of environmental conditions<sup>8</sup>. The formation of Pb phosphates in soils contaminated with both Pb and P may be responsible for immobilizing Pb and thereby reducing its bioavailability<sup>2,9</sup>. Pb phosphates have low solubility, and they are several orders of magnitude less soluble than the analogous carbonates and sulphates<sup>10</sup>. Knowing of lead form is vital, not only to predict its mobility and bioavailability, but also in the assessment of risk to living organisms because insoluble forms of lead cannot easily be absorbed by biota<sup>11</sup>.

The bioavailability of lead ions can be decreased by complexation with various materials so as to decrease their toxicity<sup>12</sup>. Pb chemical immobilization using phosphate addition is a widely acceptable technique to immobilize Pb from aqueous solution and contaminated soils<sup>5,12</sup>. The goal of this research was to use the low-grade rock phosphate to immobilize exogenous Pb in yellow cinnamon and latosol soils, providing the scientific basis for the comprehensive utilization of middle- and low-grade rock phosphate.

## Materials and Methods

**Test materials:** Two samples were from topsoil (0-20 cm) of yellow cinnamon and latosol soils collected in Zaoyang City of Hubei Province and Chengmai County of Hainan Province, respectively. Their basic physical and chemical properties are presented in Table 1. The tested rock phosphate was purchased from Huji Jinxiang Industry District in Zhongxiang City of Hubei Province. The soil samples were sieved through a stainless steel screen of 0.149-mm mesh. The contents of total phosphorus, calcium, citric acid-soluble phosphorus and lead in rock phosphate were 232 g kg<sup>-1</sup> (P<sub>2</sub>O<sub>5</sub>), 381 g kg<sup>-1</sup> (CaO), 260 g kg<sup>-1</sup> (P<sub>2</sub>O<sub>5</sub>) and 18.9 mg kg<sup>-1</sup>, respectively.

**Sample treatments:** Both yellow cinnamon soil and latosol soil were treated as lead-contaminated soils after addition of 200 mg kg<sup>-1</sup> lead as analytically-pure Pb(NO<sub>3</sub>)<sub>2</sub>. The two soils were adjusted to reach the water content of 20% (weight) and kept for 30 d at room temperature. After application of 0, 50, 500 and 2000 mg kg<sup>-1</sup> rock phosphate (CK, PR1, PR2, and PR3) the soils were undergone static incubation for 2 d, air-dried and ground.

**Analytical methods:** After digestion with HNO<sub>3</sub>-HCl-HClO<sub>4</sub> the total content of Pb in two soils was determined by flame atomic absorption spectrometry (FAAS-240). The forms of Pb were classified by Tessier's sequential extraction procedure, whose specific operating steps are given in Table 2. The determination of basic physical and chemical properties of soil was according to standard methods<sup>13</sup>. Total phosphorus of rock phosphate was dissolved with heated nitric acid at a ratio of 1:1; the soluble phosphorus was extracted by 2% citric acid and determined by vanadium-molybdate-yellow colorimetry. All experimental data was analyzed by using Excel and SAS software.

## Results

**Exchangeable Pb:** The exchangeable Pb content in yellow cinnamon soil was 21.2 mg kg<sup>-1</sup> in the CK treatment and 16.1, 9.11 and 2.95 mg kg<sup>-1</sup> in the PR1, PR2 and PR3 treatments, respectively, exchangeable Pb in these treatments being 24.1, 57.0 and 86.1% less than in CK (Table 3). The differences among the treatments were obvious. In the CK treatment of latosol soil the content of exchangeable Pb was 36.5 mg kg<sup>-1</sup> and in treatments PR1, PR2 and PR3 28.9, 16.4 and 3.57 mg kg<sup>-1</sup>, respectively. Exchangeable Pb in these treatments decreased significantly by 20.8, 55.1 and 90.2% comparing with CK.

With PR amount rising from 50 to 2000 mg kg<sup>-1</sup>, the content of exchangeable Pb in experimental soils decreased noticeably and reached the minimum under the treatment of P 2000 mg kg<sup>-1</sup> soil (PR3). Furthermore, the more PR was applied the more was the decrease of Pb. For the same treatment of PR, the rates of the exchangeable Pb content accounting for the total in latosol soil were higher than those in yellow cinnamon soil, and the lessening gap of exchangeable Pb content in two soils was almost the same.

**Carbonate-bound Pb:** The content of carbonate-bound Pb in yellow cinnamon soil after the treatments of CK, PR1, PR2 and PR3 was 41.5, 40.9, 40.6 and 33.4 mg kg<sup>-1</sup> with marginal reduction (Table 4). The proportion of carbonate-bound Pb accounting for the total content was 18.2, 17.9, 17.7 and 14.6%, respectively. The content of carbonate-bound Pb under the treatment of PR3 was noticeably less than in any other treatment. However, there were no significant differences between the treatments CK, PR1 and PR2.

There were no remarkably different changes for the carbonate-bound Pb content between the treatments CK, PR1, PR2 and PR3 with the increase of PR in latosol soil. The lowest amount (35.5

**Table 1.** Some chemical properties of soils.

Soil	pH	OM (g kg <sup>-1</sup> )	CEC (cmol kg <sup>-1</sup> )	Total (g kg <sup>-1</sup> )	Available P (mg kg <sup>-1</sup> )	Pb (mg kg <sup>-1</sup> )
S1	5.31	18.4	21.0	0.533	20.5	28.7
S2	4.30	20.5	9.0	0.679	3.81	27.2

S1 yellow cinnamon soil; S2 latosol soil; OM soil organic matter; CEC cation exchange capacity; same as follows.

**Table 2.** Tessier's sequential extraction procedures for soil heavy metal forms.

Procedure	Form	Extraction reagents	Reaction condition
I	Exchangeable	16 ml 1 M MgCl <sub>2</sub> , pH 7.0	Continuously shaking for 1 h at 25±1°C
II	Carbonate-bound	16 ml 1 M NaOAc, pH 5.0 (adjusting with HOAc)	Continuously shaking for 1 h at 25±1°C
III	Fe-Mn oxide-bound	40 ml 0.04 M NH <sub>2</sub> OH·HCl (25% HOAc)	Continuously shaking for 6 h at 96±3°C
IV	Organic compound-bound	1) 0.02 M HNO <sub>3</sub> 6 ml, 10 ml 30% H <sub>2</sub> O <sub>2</sub> 2) 6 ml 30% H <sub>2</sub> O <sub>2</sub> 3) 10 ml 3.2 M NH <sub>4</sub> OAc diluted with 20% (v/v) HNO <sub>3</sub> to 40 ml	1) Continuously shaking for 3 h at 85±2°C 2) Continuously shaking for 3 h at 85±2°C 3) Continuously shaking for 30 min at 25±1°C
V	Residual	subtraction method	

**Table 3.** Exchangeable Pb in two soils treated with rock phosphate.

Soil	Application amount of PR (P mg kg <sup>-1</sup> )			
	0(CK)	50(PR1)	500(PR2)	2000(PR3)
S1	21.2a±0.25 (9.2%)	16.1b±0.32(7.0%)	9.11c±0.15 (4.0%)	2.95d±0.36(1.3%)
S2	36.5a±1.04 (16.1%)	28.9b±0.05(12.7%)	16.4c±0.88(7.2%)	3.57d±0.06(1.6%)

Note: Different letters after the data in the same row are statistically significant at P < 0.05, the same below.; The rates of the exchangeable Pb accounted for the total content are presented in the brackets.

mg kg<sup>-1</sup>) was for CK while small increase was responsible for the treatments PR1 and PR2 up to 41.6 and 42.1 mg kg<sup>-1</sup>. Nevertheless, Pb decreased to 38.3 mg kg<sup>-1</sup> for the treatment PR3. The rates of carbonate-bound Pb accounting for the total content were 15.6, 18.3, 18.5 and 16.9%, respectively.

**Fe-Mn oxide-bound Pb:** The contents of Fe-Mn oxide-bound Pb in the treatments CK and PR1 were 89.0 and 87.4 mg kg<sup>-1</sup> in yellow cinnamon soil with no remarkable differences, but both of them were obviously higher than contents in PR2 and PR3 (Table 5). Generally, the content of Fe-Mn oxide-bound Pb in yellow cinnamon soil decreased with increasing of PR.

For the treatments CK, PR1, PR2 and PR3, the contents of Fe-Mn oxide-bound Pb in latosol soil were 89.7, 96.7, 62.1 and 54.3 mg kg<sup>-1</sup>, respectively, with remarkable differences. The content of Fe-Mn oxide-bound Pb was higher in the treatment PR1 compared to CK in latosol soil, and reached the lowest amount at PR3. Basically, the content of Fe-Mn oxide-bound Pb in latosol soil decreased with increasing amount of PR.

**Organic compound-bound Pb:** With addition of PR, the content of organic compound-bound Pb was lowest (56.1 mg kg<sup>-1</sup>) in response to the treatment PR1 in yellow cinnamon soil, and an increasing trend attained maximum (89.7 mg kg<sup>-1</sup>) under the treatment PR3 consequently (Table 6). Although there was no significant differences between the treatments PR2 and PR3, organic compound-bound Pb was evidently higher than in CK and PR1.

The content of organic compound-bound Pb increased with the rising of PR in latosol soil, and the value ranged from 14.2 to 65.0 mg kg<sup>-1</sup>. There was no significant difference between the treatments PR1 and PR2, but Pb in these treatments was significantly lower than in PR3 and higher than in CK.

**Residual Pb:** The content of residual Pb of yellow cinnamon soil was 0.44 mg kg<sup>-1</sup> in the treatment CK and 28.3, 28.3 and 38.5 mg kg<sup>-1</sup> in the treatments PR1, PR2 and PR3, respectively (Table 7). After application of PR, the residual Pb content was obviously higher than in CK for the three treatments. The content of residual Pb was from 64.3 to 87.5 times that of CK for the treatments PR1, PR2 and PR3. There was no significant difference between the treatments PR1 and PR2, both were significantly lower than PR3 and higher than CK. The content of residual Pb increased significantly in yellow cinnamon soil after application of PR.

The contents of residual Pb under treatments CK, PR1, PR2 and PR3 were 51.3, 9.2, 49.1 and 66.1 mg kg<sup>-1</sup> at latosol soil, showing that it was lower for PR1 than for any other treatment and there was no significant difference between CK and PR2. In PR3 the content of residual Pb reached the maximum, which was 1.29 times the content of CK treatment.

## Discussion

**The relationship between PR application and transformation of lead in soil:** Many relevant studies have manifested that addition of phosphate amendments (including PR, hydroxyapatite, phosphate fertilizers etc.) to the contaminated soil could effectively transform soil Pb from the nonresidual (sum of exchangeable, carbonate, Fe/Mn and organic Pb) to the residual fraction. Particularly, lead immobilization by phosphate amendments showed a long-term stability. Ma *et al.*<sup>14</sup> demonstrated this kind of transformation of lead in soil after the application of PR through related experiments and suggested that lead precipitation as a fluoropyromorphite-like mineral in these contaminated soils was the primary mechanism of reducing Pb solubility and reduction in Pb of the nonresidual fraction. A further study indicated that PR effectively immobilized Pb in Pb-contaminated soils, but its effectiveness was affected by soil pH and extent of Pb contamination, with aqueous Pb reduction

**Table 4.** Carbonate -bound Pb in two soils treated with rock phosphate.

Soil	Application amount of PR (P mg kg <sup>-1</sup> )			
	0(CK)	50(PR1)	500(PR2)	2000(PR3)
S1	41.5a±0.17(18.2%)	40.9a±1.31(17.9%)	40.6a±0.05(17.7%)	33.4b±1.55(14.6%)
S2	35.5a±4.15(15.6%)	41.6a±3.24(18.3%)	42.1a±1.07(18.5%)	38.3a±0.01(16.9%)

Note: The rates of the carbonate-bound Pb accounted for the total content are presented in the brackets.

**Table 5.** Fe-Mn oxide-bound Pb in two soils treated with rock phosphate.

Soil	Application amount of PR (P mg kg <sup>-1</sup> )			
	0(CK)	50(PR1)	500(PR2)	2000(PR3)
S1	89.0a±0.12(38.9%)	87.4a±2.08(38.2%)	63.5b±2.62(27.8%)	64.2b±1.38(28.1%)
S2	89.7b±2.75(39.5%)	96.7a±1.44(42.6%)	62.1c±1.52(27.3%)	54.3d±1.76(23.9%)

Note: The rates of the Fe-Mn oxide-bound Pb accounted for the total content are presented in the brackets.

**Table 6.** Organic compound-bound Pb in two soils treated with rock phosphate.

Soil	Application amount of PR (P mg kg <sup>-1</sup> )			
	0(CK)	50(PR1)	500(PR2)	2000(PR3)
S1	76.6b±0.11(33.5%)	56.1c±3.81(24.5%)	87.3a±2.08(38.2%)	89.7a±2.39(39.2%)
S2	14.2c±0.34(6.2%)	50.8b±1.34(22.4%)	57.6b±2.43(25.3%)	65.0a±4.57(28.6%)

Note: The rates of the organic compound-bound Pb accounted for the total content are presented in the brackets.

**Table 7.** Residual Pb in two soils treated with rock phosphate.

Soil	Application amount of PR (P mg kg <sup>-1</sup> )			
	0(CK)	50(PR1)	500(PR2)	2000(PR3)
S1	0.44c±0.08 (0.2%)	28.3b±0.10(12.4)	28.3b±0.43(12.4%)	38.5a±1.86(16.8%)
S2	51.3b±0.69(22.6%)	9.22c±0.52(4.1%)	49.1b±1.05(21.6%)	66.1a±2.86(29.1%)

Note: The rates of the residual Pb accounted for the total content are presented in the brackets.

ranging from 21.8 to 100%<sup>15</sup>. Ryan *et al.*<sup>16</sup> indicated that addition of hydroxyapatite to the soil caused a decrease in each of the first four fractions of sequential extractable Pb and a 35% increase in the recalcitrant extraction residue. In the study of Cao *et al.*<sup>17</sup>, the residual Pb increased 11-55% for different treatments. Lead immobilization was attributed to the P-induced formation of chloropyromorphite [Pb<sub>10</sub>(PO<sub>4</sub>)<sub>6</sub>Cl<sub>2</sub>], which was identified in surface, subsurface and plant rhizosphere soil<sup>5</sup>. The existence of pyromorphite-like minerals was confirmed by X-ray diffraction (XRD)<sup>17</sup>. Knox *et al.*<sup>18</sup> showed that apatite addition of 50 g kg<sup>-1</sup> of soil decreased the first three least strongly bound Pb fractions (exchangeable, carbonate and Fe-Mn oxide fractions) by 34% and increased the last most strongly bound Pb fraction by 18%. Melamed *et al.*<sup>19</sup> proved that phosphate treatments effectively transformed up to 60% of total soil Pb from the non-residual fraction to the residual fraction relative to the control. In the study of Cao *et al.*<sup>20</sup>, phosphate was more effective in transforming soil Pb (to 53%) from the non-residual to the residual phase than in transforming of soil Zn (to 15%) and Cu (to 13%). This was because Pb was immobilized by P via formation of an insoluble pyromorphite-like mineral in the surface and subsurface soil, whereas no phosphate mineral formation with Zn or Cu was found. Tang *et al.*<sup>21</sup> proved that the slight reduction in water-soluble, exchangeable and carbonate-bound Pb in PR-treated soil is possibly attributed to low solubility of PR compared with other P amendments, thus providing low-soluble P to introduce the formation of pyromorphite. In the study of Chen *et al.*<sup>19</sup> sequential extraction results indicated that the phosphate amendments converted significant amounts of soil Pb non-residual fractions to residual one. The most significant transformation was a reduction in the exchangeable Pb and amorphous Fe and Al oxide-bound-Pb, with a concurrent increase in the residual Pb (35.6-51.9%)<sup>22</sup>. Cao *et al.*<sup>23</sup> found that compared with the control, all P amendments significantly reduced Pb water solubility, phytoavailability and bioaccessibility by 72-100%, 15-86% and 28-92%, respectively.

With the increasing amount of PR, the exchangeable Pb content was significantly reduced by 24.1-86.1% in yellow cinnamon soil and 20.8-90.2% in latosol soil compared with the control. However, the addition of PR led to merely little change for the content of carbonate-bound Pb and the reduction of Fe-Mn oxide-bound Pb. Moreover, the augment of organic compound-bound Pb and the increasing trend of residue Pb were attributed to the PR treatments. Generally, for two soils, with the largely tantamount changing trends of various forms of lead in them, the application of rock phosphate was responsible for the decrease of exchangeable Pb content effectively and the conversion of Pb to the residual fraction from other forms. In addition, the more PR was applied the more it contributed to lead immobilization, which was consistent with the results.

**Differences and mechanisms of lead transformation in soil after PR application:** After application of rock phosphate on the whole, the changing trend of exchangeable Pb content in the latosol soil resembled that in yellow cinnamon soil. However, the exchangeable Pb content in latosol soil declined much more than in yellow cinnamon soil, possibly on account of the disparity of soil CEC which was larger in yellow cinnamon soil than in latosol soil (Table 1). Meanwhile, the sharp difference of negative charges

between the two soils owing to the distinct components of soil colloid came to the various Pb<sup>2+</sup> adsorption capacity, thereby resulting in the disparate quantities of the exchangeable Pb for two soils. This manifestation was in line with the widely accepted notion that the adsorption capacity of heavy metals in soil increased with the enhancement of soil pH, organic matter and CEC<sup>15</sup>.

The two distinct soils induced little change of carbonate-bound Pb in quantity, which decreased all the way with rising of PR in yellow cinnamon soil, nonetheless, with the trend of going up and then going down afterwards with the consecutive addition of PR in latosol soil. For non-calcareous soil subjected to Tessier's sequential extraction (pH <7), a considerable quantity of carbonate-bound Pb was extracted in favor with the incomplete extraction of exchangeable fraction<sup>24</sup>.

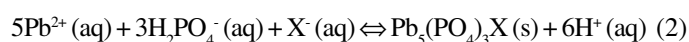
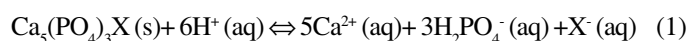
Some studies proved that after application of calcium carbonate in soil, as its amount increased, the soil pH increased while oxide-bound Pb decreased<sup>25</sup>. The whole content of calcium in the tested PR was 381g kg<sup>-1</sup> (CaO), and with the augmentation of PR, the content of calcium carbonate in soil increased, which may lead to reduction in Fe-Mn oxide-bound Pb content. Of course, phosphate can also be adsorbed by or interact with other soil components such as iron and aluminium oxides and clays<sup>26</sup>, which may eventually lead to reduction of oxide-bound Pb. When the largest amount of PR (PR3) was applied to soils, the Fe-Mn oxide-bound Pb content reached the minimum. Moreover, the Fe-Mn oxide-bound Pb content in latosol soil was lower than that in yellow cinnamon soil. The phenomenon should have something to do with the fact that the content of Fe-Al oxide components in latosol soil was higher than that in yellow cinnamon soil. Consequently more opportunities are available for the Fe-Al oxide components to combine with phosphorus but less opportunities to combine with lead. The Fe-Al oxide-bound Pb content reached the maximum under the treatment PR1 in latosol soil, which might be due to the least amount of applied rock phosphate. Meanwhile, Fe and Al contents in latosol soil were higher than in yellow cinnamon soil, where Fe-Al oxide-bound Pb content reached the largest amount. On the other hand, the addition of PR prompted an increase of latosol soil pH, resulting in higher Fe-Al oxide-bound Pb content of PR1 treatment comparing with the control.

The organic compound-bound Pb content in two soils increased with the addition of PR with higher extent for latosol soil than for yellow cinnamon soil that was in line with soil organic matter content. It was reported that the lead in soil could combine with -SH and -NH<sub>2</sub> groups of organic matter to form a stable complex<sup>27</sup>. Because of the increase in PR amount, the soil pH rose correspondingly; as a result, the odds of Pb<sup>2+</sup> reacting to anions were aggrandized, which may cause the increase of the organic compound-bound Pb content.

After the application of tested PR, the residual Pb content reached the largest amount under the treatment PR3, in this way, Pb was effectively bound in two soils. An increasing trend for the residual Pb content was emerged in yellow cinnamon soil since the increase of PR. However, the variation law of it in yellow cinnamon soil was different from that in latosol soil, possibly for the reason that the available phosphorus was extremely low in latosol soil (3.81mg kg<sup>-1</sup>), and there existed a competitive attraction between Pb<sup>2+</sup> and other cations in soils dwindling in probability of the phosphate combining with Pb<sup>2+</sup> for treatment PR1. In regard

to the pH, which in the soils increased keeping pace with the amount of PR (the pH of yellow cinnamon soil was 5.07, 5.12, 5.20 and 6.39 and the pH of latosol soil 4.16, 4.24, 4.74 and 5.59, for relevant treatments individually). Previous studies have shown that as long as hydroxyapatite is applied to soil as a major source of phosphorus, it does not only provide Ca to the soils but also enhances soil pH<sup>28</sup>. The dissolution of tested PR with calcium content of 381 g kg<sup>-1</sup> (measured in CaO) consumed H<sup>+</sup>, contributing to the increase of pH for the two soils afterwards. The higher pH was in favor of the existence of PO<sub>4</sub><sup>3-</sup>, thereby urging the more residual Pb with the addition of PR in two soils. The residual Pb content of CK was significantly higher than that of PR1 treatment in latosol soil. For all the treatments, except PR1, the residual Pb content in latosol soil was higher than in yellow cinnamon soil likely in virtue of the specific adsorption of Pb<sup>2+</sup> to iron, aluminium and manganese oxides and their hydrates, but the mechanism needs research in depth.

The dissolution of PR and subsequent formation of pyromorphite (the possible mechanism of Pb immobilization in two soils) can be expressed as follows:



where X = F<sup>-</sup>, OH<sup>-</sup> or Cl<sup>-</sup>.

Many previous studies have shown that application of different P amendments caused soil Pb to convert from forms with high activity (exchangeable, carbonate, Fe-Mn oxide, organic matter bound Pb) to the more strongly-bound Pb fractions, sulphide or residual Pb<sup>5, 16, 17, 20, 22</sup>. The increase of Pb in the residual or insoluble fraction in soils results from formation of pyromorphite (Pb<sub>5</sub>(PO<sub>4</sub>)<sub>3</sub>X, where X = F, Cl, Br, OH), the most stable Pb compounds in natural environment within a wide range of pH and Eh conditions<sup>12</sup>.

### Conclusions

Incubation and sequential extraction showed that the content of exchangeable Pb in yellow cinnamon and latosol soils decreased significantly with the increase of PR amount. After application of PR, the content of carbonate-bound Pb in yellow cinnamon and latosol soils changed slightly. With the increase of PR amount in the two soils, the content of Fe-Mn oxide-bound Pb diminished while the content of organic compound-bound Pb enhanced reversely. After application of tested PR, the residual Pb in yellow cinnamon soil rose markedly, and both of the residual Pb in the two soils reached the maximum under the treatment of P 2000 mg kg<sup>-1</sup> soil. To sum up, after the two soils being applied with testing PR, the content of exogenous Pb was immobilized effectively in yellow cinnamon and latosol soils.

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