影响磷矿粉在淹水水稻土中溶解和有效性因素研究

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摘　要：通过对在采自浙江省金华市的 12 种不同性质的水稻土添加江西省吴村磷矿粉（WPR）进行淹水培养试验，研究淹水条件下影响磷矿粉溶解和有效性的土壤因素。结果表明，磷矿粉在土壤中的溶解量与土壤 pH（KCl）、交换性钙离子含量以及土壤粉粒含量呈极显著或显著的负相关，而与土壤粘粒含量呈显著的正相关。经逐步回归发现，土壤 pH（KCl）是影响磷矿粉溶解的第一因素，土壤 pH（KCl）和土壤磷吸附常数 K 共同解释了 78%的磷矿粉溶解。加入磷矿粉后土壤有效磷的增加量与土壤的 Olsen-P 含量呈极显著的正相关。施用磷矿粉后土壤有效磷增加量占溶解量的百分比与土壤 Olsen-P 含量呈极显著性正相关，而与土壤粘粒含量、最大吸磷量呈显著负相关。

关键词：有效磷；溶解；水稻土；磷矿粉

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Factors Affecting the Dissolution and Availability of Phosphate Rock in Flooded Paddy Soils

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Abstract: Two P fertilizer of varying solubility, namely, Wucun phosphate rock (WPR), monocalcium phosphate (MCP), were incubated with twelve flooded paddy soils to quantify factors affecting the dissolution and availability of PR, the twelve paddy soils were sampled from Jinhua, Zhejiang province. Results showed the amounts of WPR dissolution (ΔNaOH$-$P) was negatively correlated with soil pH ($P<0.01$), exchangeable calcium and soil silt content ($P<0.05$), positively correlated with soil clay content ($P<0.05$). Stepwise multiple regression indicated that KCl extractable pH was the soil property that most affected PR dissolution, soil pH and adsorption parameter K together accounted for 78% of the variation in PR dissolution. The increase of soil available P (ΔResin$-$P) after WPR application was positively correlated with soil Olsen$-$P ($P<0.01$). Percentage of ΔResin$-$P by ΔNaOH$-$P after WPR application was positively correlated with soil Olsen$-$P ($P< 0.01$), negatively correlated with soil clay content and Xm ($P<0.01$). Stepwise multiple regression indicated that soil Olsen$-$P and Xm were the soil properties that mainly affected percentage of ΔResin$-$P by ΔNaOH$-$P after WPR application.

Key words: Available P; dissolution; paddy soil; phosphate rock.

0 Introduction

Soil properties affecting dissolution of phosphate rock (PR) are soil acidity, cation exchange capacity, exchangeable calcium, exchangeable magnesium, soil P status, P retention capacity and soil organic matter[1-2]. The soil properties that favor the dissolution of PR are

low pH (less than 5.5), low solution concentration of Ca ions, low P fertility levels and high organic-matter content. Soil factor affecting dissolution and availability of PR under upland condition has been extensively studied[3-4]. But there were little know about flooded conditions[5-7].

Dissolution of PR under flooded condition is not more extensively study because many factors determine the conditions under which their use is profitable. When soil is placed under anaerobic conditions, usually changes of soil physicochemical properties such as solubility of Fe, Mn and Al minerals brought about by a change in the redox potential may lead increase of soil solution P and decrease of P adsorption capacity[8-9], which may have negatively influence on PR dissolution. However, P sorption capacity can increase in some case. It was found that P sorption increased in anaerobic soils which pH rose to around 6.4 or more because of increased adsorption on freshly precipitated amorphous ferrous hydroxides[10-11]. After study effects of flooded conditions on P sorption and release of 18 paddy soils with contrasting chemical and physical properties, Gao et al[12] found that the P sorption capacity and maximum buffering capacity and standard P requirement of most of the soils increased while P release from all soils decreased. At the same time, flooding may tend to increase diffusion coefficient of phosphate twofold or threefold times compared with soils at field capacity[13-14]. The increased diffusion coefficient may accelerate the dissolving ions away from the PR surface, and thus may favor dissolution of PR.

Dissolution dynamic of PR in soil under flooded conditions related with soil pH changes, the PR dissolution reached equilibrium when soils pH reach neutral[7]. When soils are first placed under anaerobic conditions, flooding usually takes from 2 weeks to one month to tend the soil pH increase to around 7.0[11-14]. A hypothesis of this study is that PR could dissolve in this lag. On the other hands, the dissolution reaction might not occur when the soil pH to stabilize around 7.0[15].

Accordingly, a thirty five day laboratory incubation experiment was conducted to quantify factors affecting the dissolution and availability of PR in flooded paddy soils.

1 Materials and methods

1.1 Soils

Twelve representative top (0-20 cm) paddy soils were taken from seven townships of the Jinhua-Quzhou Basin in central Zhejiang, near Jinhua City (29° 5′ N, 119° 47′ E), and represents a rice-growing area of about 145000 hm^2 in the Jinhua district. The soils belong to typical eduoagulpt of alluvial origin (aquents, aquepts, and aqualfs). Soil samples were air-dried and ground to pass a 2 mm sieve. Soil pH was measured in deionized water and 1 mol/L KCl both with a soil/solution ratio of 1:1. Organic carbon content was measured by method of Walkley[16]. Available P was extracted with 0.5 mol/L (pH 8.5) $NaHCO_3$[17]. Extractable Ca and Mg of the soils extracted by neutral NH_4OAc were determined by atomic absorption spectrophotometer. Efficient cation exchange capacity was determined by the method of Hendershot and Duquette[18]. Percentages of sand (20-2000 μm), silt (2-20 μm), and clay (<2 μm) were determined by the pipette method[19].

The adsorption experiment was carried out using method of Nair et al[20]. This mainly involves addition of six KH_2PO_4 solutions (0, 5, 10, 15, 20 and 30 mg P/L in 0.01 mol/L $CaCl_2$ solution) to the soil samples at soil/solution ratio of 1:25. A few drops of toluene were added to inhibit microbial activity. The solutions were shaken for 24 h at 25ºC, and then centrifuged. P in the supernatant solution was determined. The P adsorption maximum and adsorption parameter K were determined by use of Langmuir equation[21]. Langmuir equation is $X=KXmC/(1+KC)$, where C is the concentration of P in the solution at equilibrium, Xm is the P adsorption maximum and K is a constant presumably related to binding strength of P on the soil. Some relevant properties of the soil samples are given in Table 1.

pH (H_2O), pH extracted with deionized water. pH (KCl), pH extracted with 1 mol/L KCl solution. Exch. Ca, exchangeable Ca. Exch. Mg, exchangeable Mg. Eff. CEC, efficient cation exchange capacity.

1.2 Sources of P

Wucun PR (WPR) was collected from Jiangxi province, China. The PR sample was finely ground to pass 0.15 mm sieve. The properties of WPR were: pH (1: 5H_2O) 8.40, total P 62.3 g/kg, 2% citric acid extractable

Table 1 Some basic properties of the paddy soils used

Soil code	pH (H$_2$O)	pH (KCl)	Organic carbon/%	Olsen-P /(mg/kg)	Exch. Ca /(cmol/kg)	Exch. Mg /(cmol/kg)	Eff. CEC /(cmol/kg)	Sand/%	Silt/%	Clay/%	Xm /(mg P/kg)	K
1	4.94	4.14	2.27	14.46	6.96	0.34	12.30	25.7	39.9	34.4	299.52	0.144
2	4.95	3.96	1.87	8.06	6.58	0.49	12.37	12.0	51.4	36.6	358.28	0.366
3	5.12	4.26	2.07	7.62	7.28	0.32	12.38	25.9	43.8	30.3	256.00	0.161
4	5.00	4.17	1.86	18.04	4.42	0.30	8.27	17.1	52.6	30.3	216.62	0.678
5	7.42	6.88	1.63	8.95	42.20	1.53	20.10	9.7	63.0	27.3	278.26	0.750
6	5.41	4.50	1.72	9.26	13.08	1.01	14.64	19.9	40.5	39.6	253.44	0.902
7	6.33	5.45	2.19	12.09	16.88	1.04	16.30	14.9	46.3	38.8	278.57	0.125
8	6.25	5.39	1.98	60.52	14.57	1.19	15.82	16.5	58.1	25.4	256.41	0.076
9	5.14	4.48	1.84	43.11	6.40	0.73	10.36	26.5	43.4	30.1	212.79	0.090
10	5.59	4.63	1.81	17.78	7.15	0.68	10.94	27.6	42.3	30.1	276.52	0.128
11	5.25	4.61	1.55	11.52	4.12	0.44	7.94	30.3	45.5	24.2	178.74	0.107
12	7.97	7.29	2.76	10.63	53.13	0.60	15.20	26.8	54.0	19.2	141.57	0.544

P (% total P) 29. Apatite minerals in WPR was identified by Phillips-PW1732 X-diffractometer (XRD) using nickel filter Cu radiator with intensity of scan at 2°/min at electric pressure 40 KV and electric current 20 mA. Based on the measured unit-cell *a*-value by XRD, the empirical formula of apatite mineral in WPR was estimated as Ca$_{9.50}$Na$_{0.102}$Mg$_{0.04}$(PO$_4$)$_{5.50}$(CO$_3$)$_{0.50}$F$_{2.20}$) [1]. In this study, analytical-grade reagent of monocalcium phosphate (MCP), Ca (H$_2$PO$_4$)$_2$·H$_2$O was used to estimate recovery of added P by 0.5 mol/L NaOH extraction.

1.3 Dissolution and availability in acid paddy soils

P treatments included: no P (Control), incorporated P at 1000 mg P/kg with KPR and 100 mg P/kg with MCP. For investigation changes of 0.5 mol/L NaOH and resin extractable P, one gram of soil mixture was flooded with 2 mL of distilled water in 10 mL screw cap tube. All soil mixtures were closed incubated for 35 d at 25ºC with three replications for each extraction method. The details of the extraction procedure were as below.

1.3.1 Dissolved P The dissolution of PR in soil was measured by 0.5 mol/L NaOH extraction method[22]. At the end of incubation, soil mixture was transferred to 100 mL tube with 38 mL 1.05 mol/L NaCl (make sure the final NaCl concentration was 1 mol/L) so that the final soil/solution is 1:40 and shaken 1h for prewashing the soil mixture. After prewashing, the soil was shaken with 100 mL of 0.5 mol/L NaOH for 16 h; the extracts

were filtered for P determination. The P in the 1 mol/L NaCl extractions of soils was also determined, and its amounts were negligible.

The difference in P level (ΔNaOH-P) between the MCP treated and the control samples was used as an estimate of P recovery.

1.3.2 Resin-P Resin in nylon netting resin capsule was purchased from UNIBEST Company USA. Total area per capsule is 11.4 cm^2, cation exchange capacity is 1.2 mmol (＋), anion exchange capacity is 1 mmol (-). To prepare the resin capsule as an ion sink, it was saturated with HCO$_3^-$ by adding capsule into 500 mL 1 mol/L NaHCO$_3$ solution for 10 h. At the end of incubation, soil mixture was transferred to 100 mL tube with 38 mL distilling water; one capsule saturated with HCO$_3^-$ was added to the tube and shaken for 2 h. The recovered resin capsules were then treated with 30 mL of 2.0 mol/L HCl for P desorption from resin for 16 h. All of the P content in the extracts was determined by the method of Murphy and Riley[23].

2 Results and Discussion

2.1 Dissolution of phosphate rock in paddy soils

Most paddy soils used in this study had a clay loam to silty clay soil texture and relatively high soil organic matter content and low to moderate cation change capacity. Small amounts of P (0.034%) in WPR were extracted by 0.5 mol/L NaOH. Recovery of added water soluble P by 0.5 mol/L NaOH extraction was different

from paddy soils. The recoveries of twelve paddy soils ranged from 79.63 to 98.58%. The NaOH extractable P from PR dissolution was, therefore, corrected with P recoveries in the MCP amended soils. The amounts of PR dissolved in twelve paddy soils ranged from 7.51 to 28.75 mg P/kg. The influence of relevant soil properties on the amounts of PR dissolution was investigated by linear regression analysis (Table 2). Results showed that the soil properties that significantly negatively correlated with dissolution of PR were soil pH ($P<0.01$), exchangeable Ca and silt content ($P<0.05$), significantly

positively correlated with dissolution of PR was Xm ($P<0.05$). It was also found that the correlation between dissolution of PR and KCl extractable pH was stronger than water extractable pH. This may be explained by that KCl solution may also exchange part of H^+ and Al^{3+} adsorbed on soil colloid, which may have a major influence on PR dissolution. A study of Yampracha et al[7] also found that soil acidity measured by pH and KCl extractable Al were the soil properties that most affected PR dissolution under flooded acid sulfate soils.

Stepwise regression analysis (probability of F to

Table 2 Correlation coefficients between soil properties for the 12 soils used

	WPR	pH (H$_2$O)	pH (KCl)	Organic carbon	Olsen-P	Exch. Ca	Exch. Mg	Eff. CEC	Sand	Silt	Clay	Xm	K
WPR	1.00												
pH (H$_2$O)	-0.77**	1.00											
pH (KCl)	-0.82**	0.99**	1.00										
Organic carbon	-0.23	0.43	0.39	1.00									
Olsen-P	-0.01	-0.03	-0.03	-0.05	1.00								
Exch. Ca	-0.71**	0.96**	0.96**	0.48	-0.17	1.00							
Exch. Mg	-0.40	0.60*	0.59*	-0.23	0.27	0.48	1.00						
Eff. CEC	-0.41	0.74**	0.71**	0.22	-0.02	0.71**	0.82**	1.00					
Sand	0.01	-0.2 2	-0.19	0.20	0.01	-0.21	-0.57	-0.60*	1.00				
Silt	-0.52	0.64*	0.65*	0.01	0.20	0.60*	0.52	0.53	-0.64*	1.00			
Clay	0.61*	-0.52	-0.57	-0.23	-0.25	-0.49	0.03	0.04	-0.36	-0.49	1.00		
Xm	0.41	-0.36	-0.40	-0.25	-0.11	-0.35	0.14	0.24	-0.56	-0.05	0.70*	1.00	
K	0.11	0.30	0.29	-0.11	-0.42	0.44	0.27	0.32	-0.43	0.32	0.10	-0.08	1.00

Correlation coefficient (*r*) significant at $P<0.01$ level (**), $P<0.05$ level (*).

enter ≤0.05, to remove ≥0.10) was employed to determine soil properties that could be used to predict dissolution of PR. The fit equations were listed in Table 3. In regression, the R^2 is often interpreted as the proportion of response variation "explained" by the regressors in the equation[24]. The best fit equation was equation B (Table 3). Our result indicated that soil pH was the first most important factor alone accounted for about 66% of the variation in PR dissolution (equation A), the second most important factor was adsorption parameter K, and these two together accounted for 78% of the variation in PR dissolution. Adsorption parameter K was related to dissolution of PR with an *r*= 0.11 (not significant). However, stepwise multiple linear regression indicated that K positively influenced PR dissolution in the 12 paddy soils and was entered the

Table 3 Stepwise regression equations for predicting the amounts of PR dissolution based on soil properties.

The stepwise regression equation	R^2	S.E.	P
(A) Y=44.84-5.22[pH(KCl)]	0.656	4.31	0. 0014
(B) Y=45.39-5.93[pH(KCl)]+8.76[K]	0.784	3.60	0.0010

Y, amounts of PR dissolved in soil. S. E., Std. Error of the Estimate.

equation B. Soil properties influencing PR dissolution include: soil acidity, capacity of the soil to retain P, capacity of the soil to retain Ca[3,25-26]. Soil pH and adsorption parameter K together explained for 78% of the variation in PR dissolution. In other means, 22% of the variation in PR dissolution still can not be explained by soil pH and P adsorption capacity. It was suggested that capacity of the soil to retain Ca might be account for the residual variation in PR dissolution. Numerous

studies have indicated that solution Ca concentration and exchangeable Ca were important factors controlling the dissolution of PRs in soil[27-29].

2.2 Increase of soil available P after P fertilizer addition

Number of studies suggested that the resin P test has potential as a routine soil P test for soils applied with soluble or insoluble PR fertilizers[30-32]. The resin capsule used in this study was a type of mixed cation (Na^+ form) -anion (HCO_3^- form) capsule. The inclusion of Na^+-saturated resin will enhance P release from PR by removing Ca^{2+} associated with PR so that it does not underestimate PR availability but will not excessively dissolve PR to overestimate PR availability[33]. The differences in P level extracted by resin (ΔResin-P) from

soil mixtures with and without P fertilizer application was used to evaluate the changes of soil available P after P fertilizer addition. Increase of resin-P after WPR application ranged from 0.99 to 8.40 mg P/kg, increase of resin-P after MCP application ranged from 5.51 to 35.57 mg P/kg. Increase of resin-P of the same soil was significant higher with MCP application than with WPR application. The influence of relevant soil properties on the increase of resin-P was investigated by linear regression analysis (Table 4). Results showed that increase of soil resin-P after WPR and MCP applications were both significant ($P<0.01$) correlated with soil Olsen-P.

Certain extent of PR dissolution in soil does not

Table 4 Correlation coefficients between soil properties and increase of Resin−P for the 12 soils used

	pH (H_2O)	pH (KCl)	Organic carbon	Olsen-P	Exch. Ca	Exch. Mg	Eff. CEC	Sand	Silt	Clay	Xm	K
WPR	-0.19	-0.17	-0.03	0.85**	-0.32	-0.04	-0.35	0.17	0.05	-0.25	-0.34	-0.39
MCP	-0.09	-0.05	-0.35	0.88**	-0.26	0.33	-0.04	0.00	0.24	-0.29	-0.12	-0.47
ΔResin-P/ΔNaOH-P (%) of PR	0.29	0.33	0.20	0.73**	0.16	0.14	-0.10	0.27	0.24	-0.59*	-0.64*	-0.38

Correlation coefficient (*r*) significant at $P<0.01$ level (**), $P<0.05$ level (*).

imply an equivalent increase in the amount of plant available P. Part of P released from PR may be resorbed by newly formed Fe^{2+} and residual hydrous oxide phases as well as by P precipitation of Fe^{2+} and Ca^{2+} [7,11]. The ratio between ΔResin-P/ΔNaOH-P (%) is proposed to represent the availability of P dissolved from the PR in the soil. The ratio between ΔResin-P/ΔNaOH-P (%) in twelve paddy soils after PR application ranged from 4.48 to 48.03. The ratio between ΔResin-P/ΔNaOH-P (%) in paddy soils after PR application were significantly ($P<0.01$) positively correlated with soil Olsen-P, and significantly ($P<0.05$) negatively correlated with clay content and Xm (Table 4). Multiple stepwise regression were performed to quantify factors responding variation of ΔResin-P/ΔNaOH-P (%), the results showed that soil Olsen-P and maximum P adsorption together accounted for 85% of the variation in ratio of ΔResin-P/ΔNaOH-P (%), soil Olsen-P alone accounted for 54% of the variation, which was the first key factor affecting ΔResin-P/ΔNaOH-P (%) (Table 5).

3 Conclusions

Dissolution and availability of phosphate rock in

Table 5 Stepwise regression equations for predicting ΔResin−P/ΔNaOH−P (%) based on soil properties

The stepwise regression equation	R^2	S.E.	P
(C) $Y1=12.08+0.65$[Olsen-P]	0.535	10.31	0.007
(D) $Y1=49.03+0.59$[Olsen-P]－0.14[Xm]	0.853	6.11	0.000

Y1, ratio between ΔResin-P/ΔNaOH-P (%). Std. Error of the Estimate (S.E.).

flooded paddy soil is a consequence of joint effect of soil properties. Soil acidity was the first most important factor in the dissolution of PR in flooded paddy soils. Soil pH and P adsorption parameter K strongly affected PR dissolution. The increase of resin extractable P after PR and WSP application were significantly correlated with soil available P. The ratio between ΔResin-P and ΔNaOH-P after PR application was affected by soil Olsen-P and P adsorption maximum.

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