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The distribution of phosphorus from recycled fertilizers to different soil fractions determines the phosphorus availability in soil

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Abstract

Recycling of agricultural wastes to reduce mineral fertilizer input, in particular phosphorous (P), plays crucial role in sustainable agriculture production. Understanding the transformation of phosphorous (P) fractions and their bioavailability following soil application of different renewable P-contained fertilizers is very important for improving P use efficiency and reducing environmental risks. In this study, the effects of mineral P-fertilizer superphosphate and recycled P-fertilizers, i.e., poultry manure, cattle manure, maize straw and cattle bone meal, on their distribution to different soil P fractions, their transformation and the availability of soil P were determined by soil P sequential fractionation and ^{31}P solution nuclear magnetic resonance (NMR). The results showed that addition of mineral P fertilizer, poultry manure and cattle manure increased P fixation in a red soil more than that in a fluvo-aquic soil. In both fluvo-aquic and red soils, cattle manure out-performed all other recycled P sources used in improving soil P availability. The concentration of Olsen-P in fluvo-aquic and red soils supplemented with cattle manure were increased by 41 %-380 % and 16 %-70 % than the other recycled P sources. A structural equation model (SEM) explained 95 % and 91 % of Olsen-P variation in fluvo-aquic and red soils, respectively. Labile P fractions had positive effects on Olsen-P of fluvo-aquic and red soils. ^{31}P-NMR study

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showed that amount of orthophosphate was the main factor affecting the availability of P from different P sources. In summary, cattle manure was found to be a superior renewable source of P in improving bioavailable P in soil, and its use thus has considerable practical significance in P recycling.

Keywords: phosphorus source; soil pH; P sequential fractionation; ^{31}P-NMR

Introduction

Sufficient supply of plant available phosphorus (P) in soil is critical for optimal growth and high yields of crops (Haslam et al., 2019; Zhang et al., 2019). Application of P fertilizer is an important measure to supplement soil P in most agricultural regions. However, excessive application of P fertilizer is common (Withers, 2019; Campbell et al., 2017; Kalkhajeh et al., 2021) and it causes accumulation of P in soil and the attendant environmental and crop quality issues (Cui et al., 2021; Liu et al., 2016; Lucas et al., 2021). Recovering and recycling P in agricultural wastes such as manure, straw, animal bone meal, etc. could reduce P inputs globally (Qaswar et al., 2020; Guan et al., 2020; Ylivainio et al., 2008; Kaikake et al., 2009; Mortola et al., 2019). To this end, understanding the transformation of soil P fractions and its bioavailability following agricultural waste recycling is particularly important for improving crop P utilization, P fertilizer management and reducing environmental risks.

A series of physico-chemical transformations (dissolution, precipitation, adsorption and desorption) occurs when P-containing recycled fertilizers are applied to the soil, which are regulated by soil pH, organic matter content and soil biology (Yuan et al., 2021; Lemming et al., 2020). The addition of mineral P (superphosphate, SSP) leads to an initial spike in P availability, followed by P adsorption and precipitation occurring over time, and culminating in decreased P availability in soil (Tiessen and Moir, 1993). Compared with mineral P, organic inputs are beneficial to the conversion of moderately labile inorganic P to available P (Chen et al., 2021). Organic fertilizers contain a variety of P compounds, including a large proportion of orthophosphate (Lin et al., 2015; Liang et al., 2017). It also affect soil P dynamics by changing its P adsorption capacity (Gatiboni et al., 2019; Barnett, 1994). The concentrations of labile and moderately labile P fractions in soil were significantly increased after applying organic fertilizer for four consecutive years (Negassa and Leinweber, 2009). Compared with mineral P, cattle manure application increases the content of moderately labile P, microbial biomass and microbial activity of soil, which facilitate the provision of plant available P for a longer period (Braos et al., 2020; Neufeld et al., 2017; Zhang et al., 2018). Also, soil application of organic fertilizer was found to increase the proportion of labile organic phosphate (Po) and inositol hexaphosphate (IHP), and decrease stable Ca-P content (Yan et al., 2018). Due to the compositional variation in animal feed and the characteristics of digestive system, different livestock manures are likely to have different effects on P distribution in soil (Garcia-Albacete et al., 2012; Freiberg et al., 2020). For instance, 87 % of P in cattle manure was water-soluble, while it was just 69% in poultry manure (Li et al., 2014; Pagliari and Laboski, 2013). The fractions of P from different P-containing fertilizers, and their effects on soil P availability are very complex. Thus, it is necessary to identify and quantify P fractions from different P sources and their distribution in soil P fraction to determine the potential bioavailability and the environmental impact of P from various sources.

The relative contents of inorganic and organic P in soil is greatly influenced by soil type, land use and the type of organic amendment applied (Zhang et al., 2020; Borno et al., 2018; Pizzeghello et al., 2011). A comparative study of P fractions in a typical red soil (low pH) and a fluvo-aquic soil (slightly alkaline pH) will help reveal their transformation mechanism and relationship with edaphic condition. The improved Hedley fractionation divides soil P fractions into labile P, moderately labile P, sparingly labile P and non-labile P fractions (Negassa and Leinweber, 2009; Tiessen et al., 1984). This approach is widely used to study transformation of P fractions (Zhang et al., 2021). However, information on soil P transformation following organic fertilizer application remains limited. A previous study reported no change in soil organic P fractions following application of organic fertilizers for 62 consecutive years, while another study found an opposite trend of increased contents of soil organic and inorganic P fractions with long-term application of organic fertilizers (Annaheim et al., 2015; Lu et al., 2020).

Although soil P sequential fractionation defines soil P fractions according to the solubility of P component in the extract, it provides only limited information on the biogeochemical processes and plant availability of P. Hence ^{31}P solution nuclear magnetic resonance (NMR) has been widely used to study soil P transformation, which affords more opportunities for a better understanding of organic P species. For example, Wang et al. (2019) used ^{31}P-NMR to characterize the transformation of organic P compounds during the formation of organic soils in an alpine forest in Bavaria over 1500 years (Wang et al., 2019). And, it was also used to explore the source, translocation and transformation of P reservoir in agricultural soil, and to further understand the accumulation of residual P in soil (Joshi et al., 2018). The combination of the classical P sequential fractionation methodology and the advanced P speciation analysis methodology allows a more powerful approach to study P turnover in soil and related substances.

Recycling P, nitrogen and other elements from organic waste is an important and necessary step for a green and sustainable agriculture and clean environment (Liu et al., 2020b; Almeida et al., 2019; Powers et al., 2019; Withers, 2019; Zaccheo et al., 1997). Fluvo-aquic and red soils are found extensively in agricultural lands of China.

Compared with fluvo-aquic soil, red soil readily adsorbs P and reduces its bioavailability, necessitating high P fertilizer use in agriculture. However, little is known about soil P dynamics in red soils from a fertilizer management perspective, especially in relation to organic fertilizer use. The research reported here is part of our effort to reduce mineral P fertilizer consumption by replacing it with manure, and to facilitate maximum recycling of P for food production and environmental sustainability. In this study we (1) evaluated the impact of applying different P fertilizers, including organic fertilizers, on soil P composition in fluvo-aquic and red soils, and (2) studied the relationship between various soil P fractions and available P concentration, and how it is affected by various types (mineral and organic) of P fertilizers.

2 Materials and methods

2.1 Experimental material and characteristics

The experiment was carried out in a plant growth chamber located in the Department of Plant Nutrition, College of Resources and Environment, Southwest University, Chongqing, China. Soil samples were collected from calcareous fluvo-aquic soil in Quzhou, Hebei Province and red soil in Shilin County, Yunnan Province in June 2020. The fluvo-aquic soil properties were: 8.8 $mg \cdot kg^{-1}$ Olsen-P, 914.7 $mg \cdot kg^{-1}$ total P, pH 7.9 (water: soil ratio 2.5: 1), 13.2 $g \cdot kg^{-1}$ organic matter, 1.9 $mg \cdot kg^{-1}$ NH_4^+-N, 24.3 $mg \cdot kg^{-1}$ NO_3^--N, and 26.1 $mg \cdot kg^{-1}$ exchangeable potassium. The properties of red soil were: 28.2 $mg \cdot kg^{-1}$ Olsen-P, 1083.7 $mg \cdot kg^{-1}$ total P, pH 5.7 (water: soil ratio 2.5: 1), 34.9 $g \cdot kg^{-1}$ organic matter, 2.4 $mg \cdot kg^{-1}$ NH_4^+-N, 42.7 $mg \cdot kg^{-1}$ NO_3^--N, and 78.2 $mg \cdot kg^{-1}$ exchangeable potassium. Before the experiment, both soils were dried and sieved (2 mm), then re-moistened, and pre-incubated in the dark at 25 °C for 7 d, with 30% WHC.

Five P sources including single superphosphate, poultry manure, cattle manure, maize straw and cattle bone powder were used in the experiment. The total N-P-K contents of poultry manure, cattle manure, maize straw and cattle bone powder were 16-20-32 $g \cdot kg^{-1}$, 7-5-12 $g \cdot kg^{-1}$, 7-5-8 $g \cdot kg^{-1}$, 37-93-1 $g \cdot kg^{-1}$, respectively.

2.2 Experiment design

The experiment was set up with fluvo-aquic and red soils, each with five different P sources and a control. The treatments were mineral P (single superphosphate, SSP), poultry manure (PM), cattle manure (CM), maize straw (MS), cattle bone meal (CB) and without P (CK). The air-dried soil and different recycled organic fertilizers were sieved with 2 mm stainless steel sieve, and the soil was mixed with fertilizers to supply 120 mg total P per kg soil. The total amount of N ($Ca(NO_3)_2 \cdot 4H_2O$) and K (KCl), 200 and 325 $mg \cdot kg^{-1}$ of soil, respectively, was normalized for all treatments. The specific amount of fertilizer for each treatment is shown in Table 1. In all treatments, an experimental unit (a replicate) constitutes 100 g of soil enriched with an inorganic or organic fertilizer, or with no P fertilizer for control, sampled into a 200 ml plastic bottle. All replicates were kept at 25℃ in an incubator for 70 days. The soil moisture content (MC) was maintained at 30% throughout the experiment by weighing. A total of 432 experimental units (2 soil types×6 treatments×36 repetitions) were used in this experiment. Large number of replicates allowed destructive sampling on each sampling date. Soil samples were taken for Olsen-P analysis at 0, 3, 7, 14, 21, 28, 35, 42 and 70 days after adding the fertilizer. We analyzed the P fractions of day 70 soil samples.

2.3 Sample analysis

Olsen-P was determined by phosphomolybdate method using soil extract prepared with 0.5 mol·L^{-1} $NaHCO_3$, pH 8.5 (180 RPM, 25°C) and 1:20 soil (W/V) (Olsen, 1954). After acid digestion with ammonium paramolybdate-vanadate reagent, total phosphorus was determined by colorimetry (Olsen and Sommers, 1982, Koenig and Johnson, 1942). Soil total organic carbon content was determined following a wet oxidation method with $K_2Cr_2O_7$ and concentrated H_2SO_4 (Schumacher, 2002). The sequential extraction procedure proposed by Tiessen and Moir (1993) was used to obtain different soil P fractions: Resin-P, $NaHCO_3$-P, NaOH-P, HCl-P and Residual P. Organic P (Po) in different extracts ($NaHCO_3$-P, NaOH-P, conc.HCl-P) was determined by ammonium persulfate digestion method (Tiessen and Moir, 1993). Phosphorous content of extracts was quantified colorimetrically (Shen et al., 2011). The concentration of Po was calculated as the difference between total P and inorganic P. Fig. S1 shows the detailed analysis process.

Quantitation of soil P species variation by ^{31}P-NMR assay

Po analysis was also performed by NaOH-EDTA extraction followed by ^{31}P-NMR analysis (Xu et al., 2012; Li et al., 2015). For the ^{31}P-NMR analysis, soil samples at the end of 70-day incubation were ground into powder and sieved through a 100-μm mesh. They were then extracted with a solution of 0.25 mol·L^{-1} NaOH and 0.05 mol·L^{-1} EDTA for 16 h at room temperature at a sediment and extract ratio of 1:10 (Shafqat et al., 2009; Li et al., 2017). Adjust the solution pH to 9.0±1.0, stable for 30 minutes, centrifuged at 12000 g (20 °C) for 30 minutes. The NaOH-EDTA solution was frozen and lyophilized for ^{31}P-NMR analysis. This extract was then re-dissolved in 2 mL of 1 mol·L^{-1} NaOH solution for 2 h by vortex shaking, and the suspension centrifuged at 12,000 g (20 °C) for 30 min (Ding et al., 2010). An aliquot (940 μL) of the supernatant was transferred into 5-mm NMR tube, and added with a deuterated aqueous solution of methylenebisphosphonic acid-P, P′-disodium salt (MDP, Epsilon Chimie, Brest) as internal standard (δ = 16.62 ppm), to reach a final 2.65 mM concentration. Three replicate pots for each treatment were prepared for NMR analyses (Li et al., 2017).

Solution ^{31}P-NMR spectra were obtained using a Bruker 600-MHz spectrometer (Bruker, AVANCE III,

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Switzerland) operated at 242.93 MHz at a temperature of 25 °C. A power-gated decoupling pulse, a relaxation delay of 2s, an acquisition time of 0.67 s and 4000 scans was applied in the measurement. Chemical shifts were recorded relative to an 85 % H_3PO_4 standard ($\delta = 0$ ppm). All ^{31}P spectra were baseline corrected and processed by MestReC software (v. 4.9.9.9). Signal areas were calculated by integrating the individual peaks resulting from a deconvolution process. The MDP internal standard was contained in the solution used to dissolve samples for NMR analyses and served also to calibrate the frequency axis, standardize data and perform a quantitative assessment of P forms. Signals were assigned to P compounds based on data in the literature (Cade-Menun, 2005; Turner, 2008; Cade-Menun and Liu, 2014; Mclaren et al., 2020; Hill and Cade-Menun, 2009). The solution ^{31}P NMR spectra of NaOH-EDTA extracts reflected the different P sources in Fig. S2.

2.4 Statistical analysis

NMR data were processed using the MestReNova package (V8.1.4 Mestrelab Research, Spain). All experimental data were analyzed by one-way analysis of variance (ANOVAs) with SPSS 13.0 (SPSS Inc., Chicago, IL, USA) software. When ANOVAs were significant, treatment means for independent variables were compared using Fisher's LSD Tests. All significance levels were set to $P < 0.05$. The structural equation model (SEM) was used to identify the potential driving factors of transformation of different P fractions following different fertilizer application in the two experimental soils using IBM SPSS AMOS 22.0 (IBM Corporation 2013). Root-mean square-error of approximation (RMSEA) (<0.08), chi-square (χ^2) (χ^2/df < 2), and the P value of χ^2 (P > 0.05) were used to evaluate the model fitting.

3 Results

3.1 Cattle manure was far superior than other common organic sources to increase soil Olsen-P

In both soils, the Olsen-P fluctuated with incubation time (Fig. 1). Based on the effect size, treatments could be grouped into (1) those that strongly improved Olsen-P and (2) those slightly or not improved Olsen-P, following soil enrichment with different P sources. The highest concentration of Olsen-P was found in SSP treatment (Fig. 1). During 0-70 days of incubation, the Olsen-P concentration of SSP, PM and CM enriched fluvo-aquic soil has increased by 538 %, 236 % and 374 % compared with CK in average, respectively. In SSP, PM, and CM enriched red soil, the Olsen-P concentration was increased by 80 %, 41 % and 63 % compared with CK in average during 0-70 days of incubation, respectively. Among the organic P sources used, cattle manure outperformed other sources in improving soil Olsen-P. In both fluvo-aquic and red soil, the Olsen-P content of soil supplemented with cattle manure was increased respectively by 41 % to 380 % (p<0.05) and 16 % to 70 % (p<0.05) compared with all other recycled P sources used in this study in average. In the fluvo-aquic soil, the concentration of Olsen-P in MS and

CB was not significantly different from CK. However, in red soil, Olsen-P concentration in CB significantly increased by 11.67 % ($p<0.05$) compared with CK during the 70-day incubation. In the fluvo-aquic soil, Olsen P decreased significantly between day 28 and day 70 after application of superphosphate, while Olsen-P in soils amended with the recycled fertilizers remained stable.

3.2 All recycled P sources increased moderately labile and sparingly labile soil P fractionations

Amendment of fluvo-aquic and red soils with different P sources significantly altered different soil P fractions and increased the total soil P content (Table 2, Fig. 2). In both soil types moderately labile and sparingly labile P fractions accounted for bulk of the P content; however, their relative contribution varied greatly (Fig. 2). In fluvo-aquic soil, moderately labile P was the dominant fraction accounting for 61-67 % of the total P, and the proportion of labile-P, sparingly labile-P and non-labile P was 7 % to 16 %, 15 % to 24 % and 7 % to 8 % of total P, respectively (Fig. 2a). In contrast, in red soil, moderately labile P and sparingly labile P were present in more or less equal proportion, accounting for 32-39 % and 35-45 % of the total soil P, respectively and the labile-P and non-labile P accounted for 16-22 % and 6-7 % of total P, respectively (Fig. 2b).

In fluvo-aquic soil enriched with SSP, PM, CM and CB, the labile P fraction (Resin-P, $NaHCO_3$-Pi and $NaHCO_3$-Po), was increased by 146.07 %, 94.26 %, 141.24 % and 27.79 % compared with CK, respectively (Table 1). Similarly, in red soil, SSP, PM and CM addition increased labile P by 50.66 %, 38.08 %, 57.93 % and 17.48 % compared with CK, respectively. These results also indicate that among different organic amendments only cattle manure could increase labile soil P fraction similar to that observed with mineral P fertilizer application. Adding different P sources has increased the moderately labile P content in the soil, including NaOH-Pi, NaOH-Po and dil.HCl-P. In fluvo-aquic soils, the moderately labile P fractions of SSP, PM, CM, MS and CB supplemented soils were significantly increased by 2.3 %, 5.5 %, 6.8 %, 1.3 % and 3.5 % compared with CK, respectively. The addition of SSP, PM and MS, but not other sources, in the red soil had a significantly improvement in moderately labile P fraction, by about 7.6 %, 10.5 % and 13.3 % compared with CK, respectively, after 70-day incubation. In fluvo-aquic soil, both MS and CB significantly increased the concentration of soil sparingly labile fraction (conc.HCl-Pi + conc.HCl-Po). The content of sparingly labile P in MS and CB enriched soil was 24.5 % and 44.5 % higher than that in CK, respectively. In red soil supplemented with CM, MS and CB, the concentrations of soil sparingly labile P in treatments were significantly increased by 7.7 %, 8.4 % and 26.5 % compared with CK, respectively.

In both soils supplemented with SSP, PM and CM, the labile P and the moderately labile P fractions mostly accounted for the increase in Pt content (Fig. 3). In contrast, the increase in Pt in soils enriched with MS and CB mainly came from the moderately stable P and the inert P fractions. In the two soils, adding SSP, PM and CM significantly increased soil Resin-P and $NaHCO_3$-Pi concentrations.

3.3 Changes in soil organic P species in response to different recycled P sources

The ^{31}P-NMR spectra recorded peaks in the Po and Pi regions, including monoester P, inorganic orthophosphate, inositol hexakisphosphate, glucose-1-phosphate, DNA P, diester P and Polyphosphates (Fig. 4a, b). In both soils, inorganic orthophosphate was the dominant P species in NaOH-EDTA extracts. In fluvo-aquic soil, the concentration of inorganic orthophosphate ranged from 38 mg·kg^{-1} to 110 mg·kg^{-1} and the peak value of SSP was the highest, followed by CM. In red soil, the concentration of inorganic orthophosphate ranged from 133 mg·kg^{-1} to 367 mg·kg^{-1} and the peak value of CM was the highest, followed by SSP, which was consistent with the soil Olsen-P content and soil active P concentration (Fig. 1 and Table 2). Although the detected signal was weak, compared with SSP, more monoester P and inositol hexakisphosphate signal were detected in PM and CM supplemented both soils (Fig. 4a, b). And more inositol hexakisphosphate was detected of PM supplemented both soils. Compared with fluvo-aquic soil, more inorganic and organic P forms were detected in red soil.

3.4 Structural equation modeling (SEM) revealed contrasting effects of soil pH and total organic carbon (TOC) on P fraction transformation in fluvo-aquic and red soils enriched with different P sources

The relationship of different attributes of P sources on its transformation to various P fractions in fluvo-aquic (Fig. 5a) and red soils (Fig. 5b) were further studied by SEM. SEM explained 95 % and 91 % of Olsen-P variation in fluvo-aquic soil and red soil, respectively. In general, the labile P fractions and the moderately labile P fractions had positive effects on soil Olsen-P of the fluvo-aquic soil and the red soil, while a negative effect was evident with non-labile P fraction.

TOC and pH had different effects on various P fractions in both fluvo-aquic and red soils (Fig. 5). The soil pH turned out to be a major driver of P transformation. For instance, in the fluvo-aquic soil, pH had a positive effect on labile P fraction and moderately labile P fractions, negative effect on sparingly labile P fraction and non-labile P fraction. In the red soil, however, pH positively influenced both the labile and non-labile P fraction, while it impacted the moderately labile and sparingly labile P fractions negatively.

In the fluvo-aquic soil, TOC had a negative effect on the labile P fraction and moderate labile P fraction, but positively influenced the sparingly labile P and non-labile P fractions. In the red soil, TOC had a negative effect on labile P and non-labile P fractions, while it positively affected moderately labile P and sparingly labile P fractions.

Discussion

Large variability for soil P availability in fluvo-aquic and red soil supplemented with different P sources

In this study, we selected Olsen P as well as the labile and moderately labile fractions of the sequential soil extraction as indicators of bio-available P level in soil. The effects of different P sources on soil P availability decreased in the following order: SSP>CM>PM>CB≥MS (Fig. 1). Mineral P, used here as superphosphate, can be readily dissolved in the soil solution and immediately transformed with less labile soil P fractions or get absorbed by plant roots. In contrast, Po present in manure needs to be mineralized first before entering into the soil solution gradually. It then transforms into less labile P fractions and utilized by crop plants. Therefore, the P availability of both soils amended with superphosphate was higher than that of soil with the recycled fertilizers (Braos et al., 2020).

The results demonstrate that cattle manure had innate advantages over other commonly used renewable P sources in increasing soil P availability. In fluvo-aquic soil, the P availability of soil with mineral P-fertilizer decreased significantly at the end of 70 days' incubation, while the available P in the soil with recycled P sources remained flat (Fig. 1). Previous studies have shown that the level of soil available P remains stable following cattle manure application (Mkhonza et al., 2020), but was found decreased when mineral P fertiliser was used (Braos et al., 2020). Some studies suggest that long-term straw retention in the soil improves the content of soil available P (Li et al., 2019; Cao et al., 2021). However, in our study, incorporation of maize straw in the soil did not improve soil P availability in both soils. In long term field studies, the addition of organic matter rather than straw influenced the soil P availability (Huang et al., 2021; Sales et al., 2017). The type of straw and its processing before incorporating into soil may affect its soil enrichment capacity. Thus, future research should consider the management method of straw returning to the field to promote its in situ decomposition and nutrient release to improve soil available P.

The extent of soil P fraction variation following the application of organic amendments depends to a great extent on the soil type and soil texture (Braos et al., 2020). Compared with fluvo-aquic soil, the P fixation has increased in the red soil with superphosphate, poultry manure and cattle manure amendments (Fig. 1). A comparison of humic acid-treated soils showed higher Olsen-P concentration in the brown and drab soil than that in red soil (Yang. et al., 2013). This is similar to what we observed in our study, indicating an important role for humic acid on P availability. Further, adding bone meal in the red soil improved soil P availability, but not in the fluvo-aquic soil. This is because P mainly exists in the form of apatite in bone meal, and the release of P from apatite requires H^+ (Ylivainio et al., 2008; Jeng and Vagstad, 2009). This suggests that the application of bone meal in red soil will be more valuable. Bone meal is a very important source of available P for crops (Alotaibi et al., 2013; Jeng and Vagstad, 2009), and thus developing methods to recycle P from bone meal will have a remarkable impact on alleviating P shortage in the future.

Characterization of soil P fractions of fluvo-aquic and red soils indicated that cattle manure is a superior soil amendment than other common P sources

Soil P fraction analysis found rapid integration of most of the mineral P into the labile P fraction (Resin-P, $NaHCO_3$-Pi and $NaHCO_3$-Po) (Table 2). Only a small amount of P was converted into moderately labile P fractions (NaOH-Pi, NaOH-Po and dil. HCl-P). In the short term, there was no significant effect on the sparingly labile P (conc. HCl-Pi and conc. HCl-Po) and the residual P fraction (Residual P). However, the concentrations of moderately labile P and sparingly labile P increased in soil with the recycled fertilizers. Correlation analysis showed that there was a significant positive correlation between soil P availability and soil labile P fraction (Fig. S4), and this was further corroborated by the Structural Equation Model analysis (Fig. 5). Soil Resin-P and $NaHCO_3$-Pi fractions were the most effective forms of P for plant absorption (Negassa and Leinweber, 2010). The provision of P to soil labile P fraction differs among different P sources studied and that became the main reason for the variation in soil available P content in both soils studied. And cattle manure has a significant advantage in improving the availability of soil P over poultry manure, maize straw and cattle bone meal. The percentage of inositol phosphate to total P is about 8 % in cattle manure (Mcdowell et al., 2008; Barnett, 1994), but it is as high as 80 % in poultry manure (Leytem et al., 2008; Yan et al., 2018). Inositol phosphate may complex divalent and trivalent metal elements such as calcium, magnesium, zinc and iron to form extremely insoluble compounds, which can reduce the availability of P (Menezes-Blackburn et al., 2013). This may be an important factor explaining the higher availability of P in cattle manure-enriched soil than that with poultry manure. In contrast, the other organic amendments, the maize straw and cattle bone meal, are difficult to decompose to increase soil nutrients, including available P, in a short period; hence, the relatively low level of soil labile P fraction in their P fractions. Therefore, adding maize straw and cattle bone meal to fluvo-aquic soil will have no significant beneficial effect on soil P availability for some time.

Compared with fluvo-aquic soil, the amount of $NaHCO_3$-Po and NaOH-Pi in red soil supplemented with superphosphate and poultry manure increased, while that of Resin-P decreased (Fig. 3). P adsorption is mainly controlled by iron (Fe) and aluminum (Al) hydroxides and clay minerals in acid soils (Gerard and Frederic, 2016; Jiang et al., 2012; Gérard, 2016). In neutral and fluvo-aquic soils, the influence of $CaCO_3$ and the precipitation of Ca-phosphates is more important (Pizzeghello et al., 2011). The Fe and Al ions and hydroxides in red soil increase the sorption and decrease the decomposition of organic P, thereby inhibiting the conversion of moderately labile P to labile P fraction (Fan et al., 2019). Therefore, P is more easily adsorbed and fixed by amorphous Fe and Al hydroxides in red soil. As a consequence, the accumulation of P in moderately labile P fractions in red soil is higher than that in fluvo-aquic soil, which reduces the availability of P in soil. The change of soil pH caused by fertilization will affect the adsorption and desorption of P in soil. In acidic soils, the increase of pH value and the decrease of extractable aluminum compounds will reduce the adsorption capacity of soil for P (Lopez-Hernandez and Burnham, 1974; Xavier et al., 2009). On the contrary, in fluvo-aquic soil, the precipitation of Ca-phosphates may increase with increasing pH (Adams and Odom, 1985; Gupta et al., 1990). The structural equation model showed that the increase of pH had a significant positive effect on the labile P fraction in red soil, while it has no significant effect on the labile P fraction in fluvo-aquic soil (Fig. 5). In our study, the increment of soil Resin-P concentration in red soil after addition of superphosphate was significantly lower than that in fluvo-aquic soil, but there was no significant difference in the increase of soil Resin-P concentration in two soils with cattle manure (Fig.3). This may be due to the increase in soil pH and the decrease of P adsorption in the red soil following the addition of cattle manure, and the consequent improvement in soil P availability.

^{31}P-NMR analysis of fluvo-aquic and red soil with different P sources

In this study, a large proportion of orthophosphate was found in soils with large amounts of phosphate mineral fertilizer, which is consistent with other previous reports (Li and Marschner, 2019; Appelhans et al., 2020; Li et al., 2020; Liu et al., 2020a). In addition, orthophosphate in SSP-applied soil was significantly higher than in soil applied with recycled P containing fertilizers (Fig. 4), which was consistent with the soil P availability, indicating that orthophosphate was the main driving factor affecting the P availability of different P sources. The addition of poultry manure and cattle manure significantly increased the concentration of monoester P and inositol hexakisphosphate in both soils. Similarly, the addition of cattle manure and poultry manure increased the content of soil phosphate monoester significantly, but poultry manure had a larger effect than that of cattle manure (Yan et al., 2018; Shafqat et al., 2009). However, the mineralization processes of different organic P components in soil and their mechanism are still unclear. In this regard, studying the microbial processes of P transformation and their regulation in fluvo-aquic and red soils enriched with different P sources would provide considerable insights on developing different P management options for these soil types.

Environmental pollution and resource constraints are global challenges and they also impact P use efficiency and to improve P use efficiency (Chowdhury et al., 2017). The low P use efficiency in agricultural production system damages ecosystems beyond the acceptable limit and threatens the future food security (Withers, 2019). Optimizing P input, reducing P loss and recovering P from biological resources are effective measures to increase the utilization efficiency of P fertilizer (Chadwick et al., 2015). The results of this study indicate that animal manure, especially cattle manure, will be a superior renewable P source with increased bioavailability and is expected to play an important role in managing the limitation of P resources in agriculture.

Conclusion

The current study demonstrated that different P sources had different effects on soil P availability. They distributed P differently among different P fractions. Compared with other recycled phosphate fertilizers, cattle manure was found to be a superior source for improving soil P availability in fluvo-aquic and red soils. Olsen-P in soil supplemented with cattle manure was increased by 41.16 %-379.71 % in fluvo-aquic soil and 16.12 %-70.06 % in red soil compared with those received other recycled P-contained fertilizers. The SEM analysis showed that the soil Olsen-P content was mainly affected by the labile P fraction. ^{31}P-NMR study showed that amount of orthophosphate was the main factor affecting the availability of P from different P sources. In addition, soil pH and organic matter content have contrasting effect on soil P transformation process. Better understanding of P use efficiency of different recycled P sources and their impact on yield and environmental impact under crop production conditions should be the next logical step.

Author contribution

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590 **Table 1 The specific amount of fertilizer for different treatments.**

Treatments	Recycled fertilizers ($g \cdot kg^{-1}$)	$Ca(H_2PO_4)_2$ ($g \cdot kg^{-1}$)	$Ca(NO_3)_2 \cdot 4H_2O$ ($g \cdot kg^{-1}$)	KCl ($g \cdot kg^{-1}$)
CK	0.00	0.00	1.69	0.62
SSP	0.00	0.45	1.69	0.62
PM	5.93	0.00	0.89	0.25
CM	26.55	0.00	0.18	0.01
MS	22.06	0.00	0.40	0.30
CB	0.65	0.00	1.29	0.62

591 Note: SSP: $Ca(H_2PO_4)_2$; PM: Poultry Manure; CM: Cattle Manure; MS: Maize Straw; CB: Cattle Bone Meal.

Table 2 Phosphorus fractions in fluvo-aquic soil (FS) and red soil (RS) supplemented with different phosphorus sources as determined by the sequential soil P fraction method (Tiessen and Moir 1993) after 70 days of incubation.

Treatments		Labile P (mg kg^{-1})			Moderately labile P (mg kg^{-1})			Sparingly labile P (mg kg^{-1})		Non-labile P (mg kg^{-1})
		Resin-P	NaHCO$_3$-Pi	NaHCO$_3$-Po	NaOH-Pi	NaOH-Po	dil.HCl-Pi	conc.HCl-Pi	conc.HCl-Po	Residual-P
FS	CK	18.8±1.3de	11.4±1.2cd	36.0±1.0c	28.7±0.7c	34.3±0.7c	546.7±3.0c	134.7±5.2c	33.7±3.8c	70.3±4.3b
	SSP	97.4±2.6a	24.8±0.7b	40.7±1.3bc	40.0±2.9ab	35.0±1.2c	548.7±5.6bc	137.0±4.9cd	30.3±3cd	71.0±1.2b
	PM	60.1±2.2c	25.8±1.1b	42.7±1.2ab	35.7±1.9b	41.0±1.5b	566.7±8.1ab	144.0±4.0de	21.7±3.2de	81.7±2.4a
	CM	70.7±5.3b	45.3±1.5a	43.7±1.5ab	41.7±1.7a	44.3±2.6b	565.3±5.9ab	132.3±3.7e	18.3±0.9e	67.0±1.5b
	MS	14.8±1.3e	9.8±0.8d	43.0±2.6ab	27.0±1.0c	54.0±1.7a	536.7±6.8c	163.0±3.6b	46.7±3.2b	66.3±2.7b
	CB	25.2±0.4d	13.4±0.8c	46.0±1.0a	28.3±1.8c	32.3±2.4c	570.3±2.7a	172.0±3.8a	71.3±3.2a	68.3±1.5b
RS	CK	83.0±2.1d	33.3±2.8b	57.0±0.0c	310.3±1.5c	52.7±3.4bc	43.7±0.9b	372.7±3.5cd	52.7±4.1c	78.3±3.5ab
	SSP	139.7±4.6a	53.7±4.1a	67.7±2.0b	332.7±3.3b	56.0±3.2c	48.7±1.3b	376.3±5.2bc	53.0±2.6c	78.3±0.3ab
	PM	116.0±2.6b	52.0±2.5a	71.3±0.7b	346.0±3.6a	50.3±2.3bc	53.0±1.2b	361.3±3.7d	66.7±3.2b	84.7±2.3a
	CM	137.0±2.6a	53.0±3.2a	83.7±5.0a	304.7±3.9c	44.7±3.8c	47.7±1.8b	389.3±5b	68.7±3.3b	69.3±2.0c
	MS	93.3±2.3c	30.0±1.5b	56.0±3.2c	310.0±1.5c	104.3±1.5a	46.3±1.5b	385.7±3.5bc	75.3±4.3b	70.7±2.2c
	CB	93.3±3.8c	38.3±0.7b	72.0±3.5b	283.7±1.2d	34.7±1.8d	76.7±6.4a	411.0±3.5a	127.3±3.2a	72.3±1.7bc

Note: SSP: Ca$(H_2PO_4)_2$; PM: Poultry Manure; CM: Cattle Manure; MS: Maize Straw; CB: Cattle Bone Meal. Labile P (Resin-P +NaHCO$_3$-Pi +NaHCO$_3$-Po), Moderately labile P (NaOH-Pi +NaOH-Po +1 M HCl-P), Sparingly labile P (conc. HCl-Pi + conc. HCl-Po) and Non-labile P (Residual-P) according to Crews and Brookes (2014) and Ahmed et al. (2019), the same below. Values are means of three replicates ± standard errors (n=3). dil. and conc. HCl indicate diluted and concentrated HCl, respectively. In each column and for each soil type, means followed by a common letter are not significantly different at the 5% probability level according to the LSD test.

Figure Captions

Figure 1 Changes in Olsen-P concentrations in fluvo-aquic soil (a) and red soil (b) supplemented with different phosphorus sources during 70-day incubation. Values are means ± SE (n= 4). SSP: $Ca(H_2PO_4)_2$; PM: Poultry Manure; CM: Cattle Manure; MS: Maize Straw; CB: Cattle Bone Meal. The same below.

Figure 2 The proportion of different soil P fractions in fluvo-aquic soil (a) and red soil (b) supplemented with different phosphorus sources after 70-day incubation. Orange, green, purple, and grey indicate labile P, moderately labile P, sparingly labile P, and non-labile P, respectively.

Figure 3 Changes in P fractions in fluvo-aquic soil (a) and red soil (b) supplemented with different phosphorus sources after 70-day incubation (the values presented are those measured with addition of different phosphorus sources minus the value of no phosphorus fertilizer control).

Figure 4 Liquid ^{31}P NMR spectra of NaOH-EDTA extracts of a fluvo-aquic soil (a) and a red soil (b) amended with different P-containing fertilizers. In the upper spectrum, the shift positions of the different P compounds are indicated. A: Monoester P (7.19 to 7.58 ppm); B: Inorganic orthophosphate (6.18 to 6.34 ppm); C: Inositol hexakisphosphate (4.38 to 4.49 ppm); D: Glucose-1-phosphate (3.13 to 3.43 ppm); E: DNA P (-0.15 to -0.36 ppm); F: Diester P (-1.73, -2.43 ppm); G: Polyphosphates (-4.63 to -5.83 ppm). Concentrations of P compounds in NaOH-EDTA extracts of fluvo-aquic soil (c) and red soil (d) by ^{31}P-NMR.

Figure 5 Structural equation model (SEM) analysis for the transformation of different P fractions after the addition of different P-containing fertilizers in a fluvo-aquic soil (a) and a red soil (b). Optimal model fitting results under the fluvo-aquic soil (a): $\chi^2 = 0.098$, DF = 1, $\chi^2/DF = 0.098$, P = 0.754, and RMSEA = 0.000; optimal model fitting results under the red soil (b): $\chi^2 = 0.241$, DF =1, $\chi^2/DF = 0.241$, P = 0.623, and RMSEA = 0.000. The number on the arrow represents the standardized path coefficient, the red and blue arrows represent the positive and negative effects, respectively. *, ** and *** indicate significant at P< 0.05, P< 0.01 and P< 0.001, respectively. The black number above each variable is R^2 values, which represent the proportion of variance explained for each variable. The arrow width indicates the strength of the paths. Soil pH and total organic carbon (TOC) data are shown in Supplementary Figure S3

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628 Figure 1

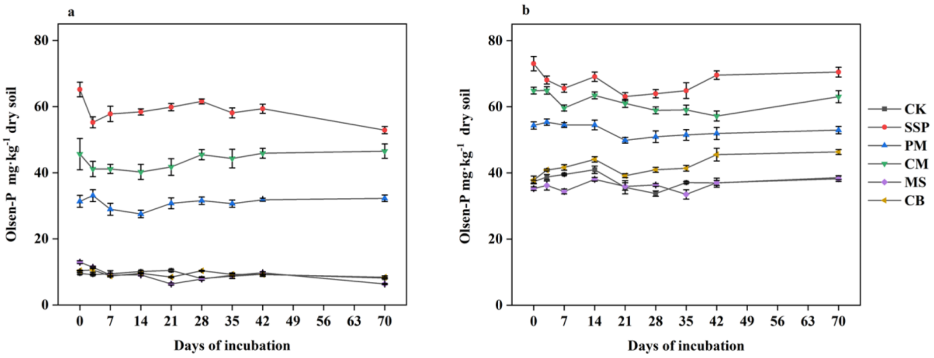

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Figure 2

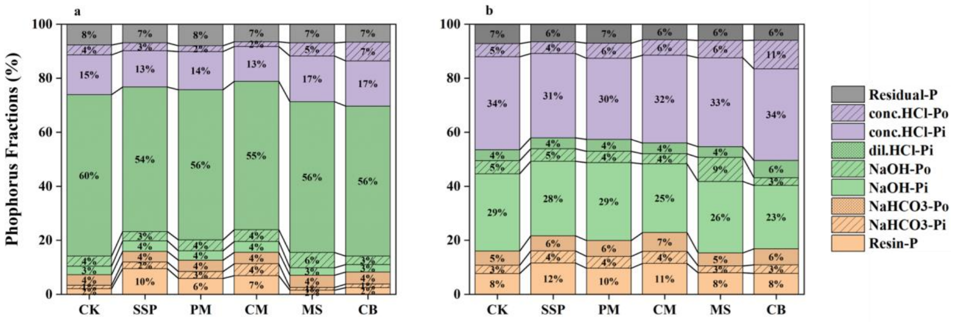

a
Phophorus Fractions (%)
100
80
60
40
20
0
CK
8%
4%
15%
60%
4%
3%
4%
1%
2%
SSP
7%
3%
13%
54%
3%
4%
4%
2%
10%
PM
8%
2%
14%
56%
4%
4%
4%
3%
6%
CM
7%
2%
13%
55%
4%
4%
4%
4%
7%
MS
7%
5%
17%
56%
6%
3%
4%
1%
2%
CB
7%
7%
17%
56%
3%
3%
4%
1%
2%
b
100
80
60
40
20
0
CK
7%
5%
34%
4%
5%
29%
5%
3%
8%
SSP
6%
4%
31%
4%
5%
28%
6%
4%
12%
PM
7%
6%
30%
4%
4%
29%
6%
4%
10%
CM
6%
6%
32%
4%
4%
25%
7%
4%
11%
MS
6%
6%
33%
4%
9%
26%
5%
3%
8%
CB
6%
11%
34%
6%
3%
23%
6%
3%
8%
Residual-P
conc.HCl-Po
conc.HCl-Pi
dil.HCl-Pi
NaOH-Po
NaOH-Pi
NaHCO3-Po
NaHCO3-Pi
Resin-P

Figure 3

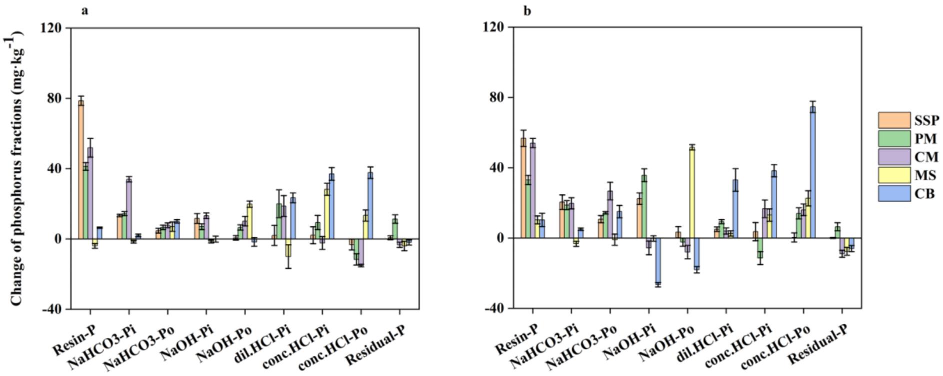

a
b
Change of phosphorus fractions (mg·kg^{-1})
120
80
40
0
-40
Resin-P
NaHCO3-Pi
NaHCO3-Po
NaOH-Pi
NaOH-Po
dil.HCl-Pi
conc.HCl-Pi
conc.HCl-Po
Residual-P
SSP
PM
CM
MS
CB

Figure 4

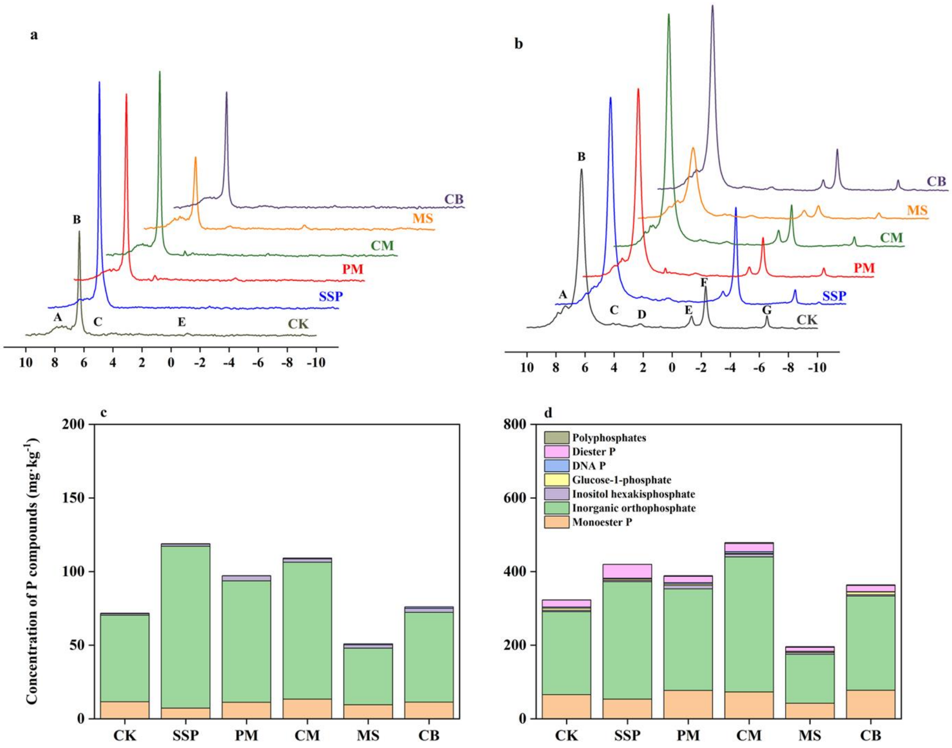
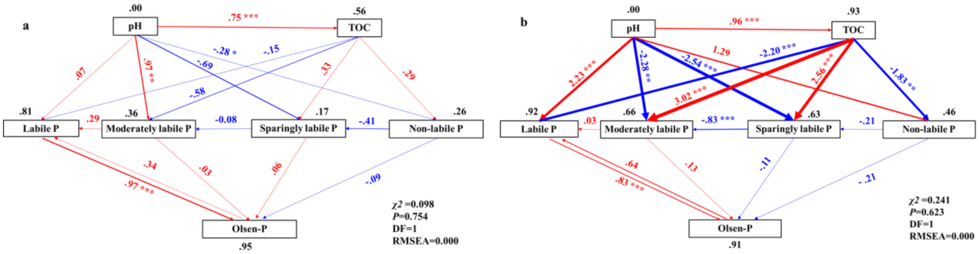

a
b
A
B
C
D
E
F
G
CK
SSP
PM
CM
MS
CB
10 8 6 4 2 0 -2 -4 -6 -8 -10
c
d
Concentration of P compounds (mg·kg⁻¹)
0
50
100
150
200
200
400
600
800
Polyphosphates
Diester P
DNA P
Glucose-1-phosphate
Inositol hexakisphosphate
Inorganic orthophosphate
Monoester P
CK
SSP
PM
CM
MS
CB

Figure 5


a
.00
pH
.75 ***
.56
TOC
.07
.97 **
-.28 *
-.69
-.15
-.58
.33
.29
.81
Labile P
.29
.36
Moderately labile P
-0.08
.17
Sparingly labile P
-.41
.26
Non-labile P
.34
.97 ***
.03
.06
-.09
Olsen-P
.95
χ2 =0.098
P=0.754
DF=1
RMSEA=0.000
b
.00
pH
.96 ***
.93
TOC
2.23 ***
-2.28 **
-2.54 ***
1.29
-2.20 ***
3.02 ***
2.56 ***
-1.83 **
.92
Labile P
.03
.66
Moderately labile P
-.83 ***
.63
Sparingly labile P
-.21
.46
Non-labile P
.64
.83 ***
.13
-.11
- .21
Olsen-P
.91
χ2 =0.241
P=0.623
DF=1
RMSEA=0.000