



## Effect of biogas slurry application rate on colloidal phosphorus leaching in paddy soil: A column study



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### ABSTRACT

Colloidal phosphorus ( $P_{\text{coll}}$ ) in soil solutions and subsurface drainage has attracted increasing attention as an additional mobile form of P. A column study was conducted to investigate the effect of biogas slurry (BS) on  $P_{\text{coll}}$  leaching in paddy topsoil (0–20 cm) at rate of 0; 225  $\text{m}^3 \text{ha}^{-1}$  (55  $\text{kg N ha}^{-1}$ , 17  $\text{kg P ha}^{-1}$ ); 673  $\text{m}^3 \text{ha}^{-1}$  (165  $\text{kg N ha}^{-1}$ , 52  $\text{kg P ha}^{-1}$ ); 1350  $\text{m}^3 \text{ha}^{-1}$  (330  $\text{kg N ha}^{-1}$ , 104  $\text{kg P ha}^{-1}$ ) of BS. The colloids particles were separated by microfiltration and ultra-centrifugation processes. Dissolved phosphorus (DP) in leachate ranged from 0.04 to 0.09  $\text{mg P L}^{-1}$  and contributed between 56 and 76% to the total P leaching.  $P_{\text{coll}}$  in leachate ranged from 0.002 to 0.03  $\text{mg P L}^{-1}$  in leachate and contributed between 3 and 26% to TP leaching. Results clearly showed that BS increased  $P_{\text{coll}}$  leaching at medium (673  $\text{m}^3 \text{ha}^{-1}$ : 165  $\text{kg N ha}^{-1}$ , 52  $\text{kg P ha}^{-1}$ ) and high application rate of BS (1350  $\text{m}^3 \text{ha}^{-1}$ : 330  $\text{kg N ha}^{-1}$ , 104  $\text{kg P ha}^{-1}$ ). In contrast, the smallest tested BS application of 225  $\text{m}^3 \text{ha}^{-1}$  (55  $\text{kg N ha}^{-1}$ , 17  $\text{kg P ha}^{-1}$ ) did not affect  $P_{\text{coll}}$  concentrations in leachates. Significant positive correlations was found between  $P_{\text{coll}}$  and colloidal aluminum ( $\text{Al}_{\text{coll}}$ ), colloidal calcium ( $\text{Ca}_{\text{coll}}$ ), total organic carbon ( $\text{TOC}_{\text{coll}}$ ), and with pH, indicating that Al/Ca minerals - organic matter may play a major role as carriers for  $P_{\text{coll}}$  leaching and these processes were pH-dependent. This study confirms that high level of BS application could result in the mobilization of  $P_{\text{coll}}$  from paddy topsoil by leaching.

### 1. Introduction

The mobilization of colloidal phosphorus ( $P_{\text{coll}}$ ) through provision of P in fertilizer can have important environmental consequences (Siemens et al., 2004). Thus, the addition of P in form of mineral fertilizer and manure to agricultural soils has often exceeded the rate of P removal by harvested crops (Slaton et al., 2004; Tunney et al., 2003). Consequently, the resultant buildup of soil P can be large, saturation of P sorption sites and greater potential for P desorption into soil solution and the risk of P losses by transport mechanisms such as leaching (McDowell and Sharpley, 2001). Kleinman et al. (2005) found significant increase in the concentration of leachate P after manure application. Manure amendment increased the output of  $P_{\text{coll}}$  and particulate P through runoff loss (Liang et al., 2016). Previous studies have also reported that colloidal particles in manure fertilizers will preferentially facilitate the vertical migration of P to deeper depths through soil macropores (McGechan, 2002). It has been reported that organic colloidal particles in manure fertilizers will also promote the

accumulation and migration of  $P_{\text{coll}}$  in undisturbed soil columns (Zang et al., 2011; Zang et al., 2013). Moreover, these authors found a > 25% increase in colloid-facilitated P transport in soils receiving manure over those with no manure addition. Other studies have also observed the importance of manure addition in the formation of soil colloids (Makris et al., 2006; Zang et al., 2011). Artificially increasing the number of water-dispersible colloids and adding manure amendments to soils were both found to increase colloidal inorganic P losses (Makris et al., 2006). Therefore, fertilization has a significant effect on the distribution, migration and mobilization of  $P_{\text{coll}}$  through soil profile.

Biogas slurry (BS), being a liquid fraction from anaerobic digestion of organic waste, is often applied to agriculture fields (Kumar et al., 2015; Nyang'au et al., 2016). In China, the annual product of BS is 0.4 billion tonnes (Cao et al., 2015), and 33% of farmers are using this product as fertilizers (Zhang et al., 2009). However, its application risk to the environmental and ecological security is not yet conclusive (Cao et al., 2015). It has been reported that the application of concentrated BS could bring significant changes to tomato production, including

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increases in organic matter and available N and P (Yu et al., 2010). In addition, Lal et al. (2015) showed that the application of BS as a nitrogen fertilizer stimulated the growth of baby corn in terms of biomass and corn yields. Moreover, Singla et al. (2014) also found that application of biogas digested slurry increased the soluble organic carbon content of the soil compared to the chemical fertilizers treatment. After all, highly concentrated BS may lead to nutrient leaching when the nutrient provided is beyond the crop requirements. However, the influence of BS on  $P_{\text{coll}}$  leaching in soil was not reported to date and requires further and detailed study. BS may contain dissolved organic matter such as humic substances and/or fulvic acids with carboxyl and/or phenol functional groups which may compete with phosphate ions for binding sites, resulting in the release of phosphate ions (Hua et al., 2008; Guppy et al., 2005).

This study investigates the effect of BS on soil  $P_{\text{coll}}$  leaching in paddy soil. Previous studies focused on sandy soils (Ilg et al., 2005, 2008; Siemens et al., 2008; Zhang et al., 2003) with low P-sorption capacity (Luis, 2008). Yet, release of  $P_{\text{coll}}$  leaching in paddy soil was not well clarified. Soil organic carbon (SOC) tends to accumulate in paddy soils as a result of organic amendments and anaerobic conditions (Kögel-Knabner et al., 2010). The increased SOC could enhance the formation of organo-Al (Fe) complexes, which has strong affinity to P (Kang et al., 2009) and, in turn, benefit soil P sorption. In contrast, evidences suggested that organic anions can also reduce P sorption capacity of existing oxides by competing with phosphate ions for binding sites (Hua et al., 2008; Guppy et al., 2005). In addition, phosphate may be released when organic matter-Al (Fe) complexes are solubilized, although some organic matter-Al (Fe)- $PO_4$  complexes that occur in soil solution and leachate waters are soluble (Darke and Walbridge, 2000).

To understand the mechanisms of soil P transport, knowledge of the portioning of individual reactive P species such as phosphate between solid phase and soil solution containing solutes and colloids is required (Shand et al., 2000). Dissolved P (DP) and particle P (PP), both of which play an important role in P migration, denote P species truly dissolved and binding with macro granules in aqueous environment, respectively (Liang et al., 2016). Environmental behavior of  $P_{\text{coll}}$  distinguishes from those of DP and PP, which can be attributed to the special physico-chemical characteristics of colloids (Hens and Merckx, 2002). In general, the fate of P in agricultural soils is mainly related not only to its occurrence in soil solution but largely to mobile  $P_{\text{coll}}$  complexes (Hens and Merckx, 2002; Shand et al., 2000). Previous studies reported that solubility, reactivity and mobility of  $P_{\text{coll}}$  are directly linked to P binding forms (Beauchemin et al., 2003; Liu et al., 2014). The presence and formation of secondary minerals, particularly iron-(Fe-) and aluminum-(Al-) (hydr) oxides, counteract the vertical losses of ortho-phosphate P and organic P from the “P recycling zone” in topsoil and organic layers. This continued formation of these secondary minerals leads to a more effective subsoil P retention by sorption (Kaiser et al., 2001). However,  $P_{\text{coll}}$  may still pass this mineral soil barrier and escape to greater soil depths (Bol et al., 2016).

In this study, a laboratory experiment was conducted with paddy topsoil (0–20 cm) in order to test the hypothesis that application rate of BS increases the leaching of  $P_{\text{coll}}$ .

## 2. Materials and methods

### 2.1. Soil and biogas slurry samples

Paddy soil and BS were collected from Jing Shan town, Zhejiang Province, China. The cropping system in the field site was planted to rice annually (rice – fallow). Compound fertilizer was applied at the same rate of  $53.55 \text{ kg ha}^{-1}$  for N, P, and K as the basal fertilizer. The urea fertilizer was applied with the rate of  $77 \text{ kg N ha}^{-1}$  as the top-dressing, and with the rate of  $56 \text{ kg N ha}^{-1}$  for fertilization for head sprouting. The rice season is from June to October, and the paddy soil was taken at the end of November 2015. > 30 kg topsoil (0–20 cm)

samples were collected from ten points of rice paddy field, composited by mixing together in plastic bag. Then, transported to the laboratory, air-dried at room temperature, and ground to pass through 2 – mm sieve for further use.

### 2.2. Leaching experiment

The experiment included four BS levels and three replications were used for each level. Soil columns of 20 cm height and 7.5 cm inner diameter were packed with 700 g of air-dried and sieved paddy soil. The BS was applied at four different treatments, 0 mL (control), 100 mL (Low), 300 mL (Medium) and 600 mL (High) per treatment (column) equaling to 0,  $225 \text{ m}^3 \text{ ha}^{-1}$  ( $55 \text{ kg N ha}^{-1}$ ,  $17 \text{ kg P ha}^{-1}$ );  $673 \text{ m}^3 \text{ ha}^{-1}$  ( $165 \text{ kg N ha}^{-1}$ ,  $52 \text{ kg P ha}^{-1}$ ), and  $1350 \text{ m}^3 \text{ ha}^{-1}$  ( $330 \text{ kg N ha}^{-1}$ ,  $104 \text{ kg P ha}^{-1}$ ) of BS levels respectively. The BS was diluted to 600 mL with deionized water in order to use the same volume and then applied manually on soil column five times at 600 mL each time. The time interval between applications was 24 h after the previous event of treatment. After the BS was applied, a glass bottle was put under the column to collect the leachate samples until no leaching was observed. Then, the leachate solution from each column at each event treatment was collected for analysis.

### 2.3. Soil and leachate analysis

Soil pH was measured at a soil water ratio of 1:5 (w/v) with pH meter (inoLab pH/cond, WTW, Weilheim, Germany) after shaking for 30 min. Soil texture was determined using the pipette method (Kroetsch and Wang, 2006). Soil bulk density was determined on an oven-dry basis using the cylinder method (Page et al., 1982). Soil organic carbon (SOC) was determined by a colorimetric method based on  $105^\circ \text{C}$  digestion with  $K_2CrO_7 - H_2SO_4$  (Sato et al., 2014). Total P was determined by the molybdenum blue method after digestion with  $H_2SO_4 - HClO_4$  at  $300^\circ \text{C}$  for 2 h (Murphy and Riley, 1962). Olsen-P was extracted by shaking 2.5 g of soil and 50 mL of 0.5 M sodium bicarbonate (pH 8.5) solution for 30 min and determined by the molybdenum blue method (Olsen and Sommers, 1982). Ammonium oxalate – extractable P, Al, and Fe ( $P_{\text{ox}}$ ,  $Al_{\text{ox}}$ , and  $Fe_{\text{ox}}$  respectively) were determined by shaking 1: 40 (W/V) soil/ammonium oxalate extracting solution (pH 3.0) for 2 h in the dark (Schoumans, 2000). Total C and Total N were determined using an elemental analyzer (Vario MAX CNX, Elementar, Germany). All analyses were conducted in triplicate.

Leachates were centrifuged at  $3000 \times g$  for 10 min to remove coarse particles and then filtered through 1- $\mu\text{m}$  microporous membrane to capture only particles. The first 5 mL of the filtrate was discarded. An aliquot of the filtrate was ultra-centrifuged at  $300,000 \times g$  for 2 h to remove colloids (Optima TL Beckman, Unterschleißhsoeim, Germany).

The pH and redox potential (Eh) of leachate were determined with pH meter with Ag/AgCl reference electrode (inoLab pH/cond, WTW, Weilheim, Germany). Eh was measured according to the method described by Liu et al. (2013). Briefly, Eh was measured after the leachate was collected, Ag/AgCl electrode was inserted into the solution leachate, and record the numerical value in display screen after it was stable. When the amplitude of fluctuation of the numerical value was < 1 mV in almost 5 min, the value was deemed to be stable. Total P concentration in the filtered and ultra-centrifuged leachate samples was measured colorimetrically after digestion with peroxodisulfate: 5 mL of sample were mixed with 1 mL solution consisting of  $150 \text{ mmol K}_2\text{O}_8\text{S}_2 \text{ L}^{-1}$  and  $180 \text{ mmol H}_2\text{SO}_4 \text{ L}^{-1}$  and autoclaved at  $121^\circ \text{C}$  for 1 h. Afterward 0.7 mL of  $188 \text{ mmol ascorbic acid L}^{-1}$  was added and the samples were boiled again at  $95^\circ \text{C}$  for 1 h (Pagel et al., 2008). Another 5 mL samples were digested with 2 mL concentrated  $HNO_3$  at  $170^\circ \text{C}$  for 20 min in a microwave oven as described by the USEPA method 3015A (USEPA, 1998). The total Fe, Al, Ca and Mg concentrations in the filtered and ultracentrifuged leachate samples were determined by inductively coupled plasma-atomic emission

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