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# Phosphorus removal of rural wastewater by the paddy-rice-wetland system in Tai Lake Basin

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#### ABSTRACT

A field experiment was conducted to remove the potential eutrophication effect of P from rural wastewater (RW) during the whole rice growing season of 2007. The experiments consisted of five treatments, namely black water (BW), domestic wastewater (DW), grey water (GW), surface lake water (SW) and surface lake water without P application as a check (CK), with three replicates in a randomized block design. Commercial fertilizer and RW were applied to furnish 40 kg Pha<sup>-1</sup> except CK. Results showed total P (TP) concentration had significantly declined after P application, from October 15 there were no significant increases in TP concentration in the floodwater. TP removal rates from RW was significantly higher ( $P \le 0.05$ ) than those from fertilizer. TP load was in an overall gradual decline, whereupon it became approximately steady on October 1. The percentage of TP load from wastewater decreased, whereas that from fertilizer continued to increase. Meanwhile, the yield for CK was significantly less ( $P \le 0.05$ ) than SW, GW, DW, and BW, with the yield of BW significantly greater ( $P \le 0.05$ ) than other treatments. It is feasible to remove P from RW by the paddy-rice-wetland system and can be widely used to improve the yield of rice.

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#### 1. Introduction

Agricultural drainage from agricultural land and RW from rural life are major sources of the nonpoint-source pollution, including P which can accelerate the eutrophication of water bodies. The potential for P loss in RW has been increased in many areas with the improvement in living standards [1]. According to Zhu [2], over 85% of the water bodies in China show a continuing increase in eutrophication and P losses from RW are contributing to this decline in water quality. Statistics indicate that the discharge capacity of RW reaches  $2.3 \times 10^4$  t day<sup>-1</sup> in Tai Lake Basin, but the treatment capacity is below 40%, so that annual average P export from RW is up to about 8.35 t, and more than 90% of the water body is eutrophic [3]. Particularly, in May 2007, an outbreak of a large blue-green algae cover occurred in Tai Lake, and the safety of drinking water in Wuxi City was seriously threatened. Therefore, seeking a cost-effective way to remove P from RW is necessary to improve water quality in this area.

Actually, DW is the total of all types of wastewater from rural people's living activities. Its main components are organic metabolic waste, foodstuff residues, physiological excrement (e.g., manure, night soil, urine), kitchen, laundry, bath wastewater, and so on. It can be further classified into BW and GW, the former is composed of human excretory urine and toilet flashing water [3,4], and the latter includes the other living wastewater (e.g., laundry, kitchen and bath wastewater). Moreover, P content and water quantity of GW are much higher than that of BW [5.6]. Due to a large amount of nutrient P contained in RW, it is a P resource waste for RW drainage. But how can RW be treated and P in RW can be reused simultaneously? Constructed wetland is a method used in RW treatment in the recent years [7-10]. But there are some problems (e.g., filler blocking, hydraulic load ununiformity, difficult maintenance) which need to be deal with. In fact, the paddy-rice-wetland system is a kind of constructed wetland, but environmentally friendly, ecologically healthy, and utilization sustainable, compared with conventional constructed wetland systems [11,12]. Although preliminary results on the removal of P from RW by constructed wetlands were reported [13,14]. However, there have been few studies on the mechanisms involved in the removal of P from RW (including the changes of TP concentration and TP load, rice yield response with various P treatment) in the paddyrice-wetland system.

The aim of this study is to help to remove the potential eutrophication effect of P from RW by incorporating the P into irrigation water as a nutrient source for rice production in the paddy-ricewetland system, thereby obtaining benefits of decontaminated RW as well as reduced irrigation water usage, due to substitution of RW, and reduced fertilizer costs while maintaining a favorable yield.

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#### 2. Materials and methods

#### 2.1. Site description, soil properties

The field experiment is located at Yuhang Agricultural Research Station (30°30'N, 120°18'E) in Tai Lake Basin of southeast China which is a very intensive rice-producing area. This area has a subtropical monsoon climate with 1500–1600 mm annual rainfall, the actual annual evapotranspiration of about 1500 mm. The annual average air temperature is 20.3 °C; with the main rainy season lasting from April to September. The dominant soil type in this basin is a *Mollic Endoaquoll* (a blue–purple paddy soil) [15,16]. Also, the soil texture in the top-layer (0–15 cm) of the experimental site is a clay loam; soil properties sampled at the start of the experiment showed: pH, 6.9; CEC, 16.8 cmol kg<sup>-1</sup>; available N, 172.3 mg kg<sup>-1</sup>; available P, 4.5 mg kg<sup>-1</sup>; and organic C, 34 g kg<sup>-1</sup>.

#### 2.2. Materials

Rice variety is *XiuShui 80* (*Oryza sativa* L.), which is a predominant local late-rice variety and provided by Yuhang Agricultural Research Station. Water used as CK and SW are both taken from an irrigation reservoir near Liangzhu Village. GW and BW are sampled from six local farmers' houses and mixed thoroughly into a uniform composite sample; before the use of BW, a 2-week fermentation period is employed to disinfect the samples of BW, and the concentration of total coliform (TC, in CFUL<sup>-1</sup>) and fecal coliform (FC, in CFUL<sup>-1</sup>) in BW decreased from 43,000 and 29,000 to 8900 and 5200, respectively, and is below the limits for the agriculture use (10,000 CFUL<sup>-1</sup>). Field experimental DW is collected near the village of Liangzhu in a rivulet, where farmers could directly deposit DW at their discretion.

#### 2.3. Experimental design

For this field experiment a randomized complete block design with three replicates was used. There were five treatments, namely a control (CK), surface reservoir water (SW), grey water (GW), domestic wastewater (DW), and black water (BW). In May 2007, 15 plots  $(4.0 \text{ m} \times 5.0 \text{ m})$  were constructed in two parallel rows (Fig. 1). Ridges, 25–30 cm wide at the base and 20 cm high, were covered with plastic sheeting, which was inserted into the soil plow layer to a depth of 15 cm to hydrologically isolate the plots. Then, an irrigation ditch, 40 cm wide and 20 cm deep, was excavated between the two parallel rows of plots. On the boundary adjacent to the irrigation channel, inlets to individual plots, 20 cm in width, were made within 50 cm of the centre of each plot. Directly opposite the irrigation inlets, outlets were connected with runoff collecting barrels placed in the drainage ditch. Both the inlets and the outlets to the plots could be opened and closed as required. To minimize disturbances and edge effects on the experimental plots, five non-experimental guard plots were established at the ends of the rows

First, N, P, and K in the irrigation water was determined so that urea, superphosphate, and KCl could be added (excluding the CK plots) as needed to ensure a basal dressing of  $180 \text{ kg N ha}^{-1}$  (as urea),  $40 \text{ kg P ha}^{-1}$  (as superphosphate), and  $50 \text{ kg K ha}^{-1}$  (as KCl), which was the typical rate of fertilization in this area for one rice season. TP concentration for the original irrigation water (in mg L<sup>-1</sup>) was 0.13 (CK), 0.13 (SW), 0.59 (GW), 0.41 (DW), and 0.21 (BW). Thus, measured P for each treatment (in kg P ha<sup>-1</sup>) was: CK of 5 from SW; SW of 5; GW of 20, DW of 15, and BW of 10. To these, P was added to attain  $40 \text{ kg P ha}^{-1}$ .

With N, fertilizer application included: for CK,  $15 \text{ kg N ha}^{-1}$  was present in SW and no other N fertilizer was added; for SW,  $15 \text{ kg N ha}^{-1}$  was present in SW and  $165 \text{ kg N ha}^{-1}$  was added; for

GW,  $30 \text{ kg N} \text{ ha}^{-1}$  was present in GW and  $150 \text{ kg N} \text{ ha}^{-1}$  added; for DW,  $60 \text{ kg N} \text{ ha}^{-1}$  was present in DW and  $120 \text{ kg N} \text{ ha}^{-1}$  added; and for BW,  $90 \text{ kg N} \text{ ha}^{-1}$  was present in BW and  $90 \text{ kg N} \text{ ha}^{-1}$  was added. Finally, K in the irrigation water was negligible ( $0.02 \text{ mg L}^{-1}$ ), so K came solely from KCl fertilizer.

On June 28, 2007, all 15 plots in the paddy-rice-wetland system were irrigated with CK and SW plots receiving SW as irrigation water and GW, DW, and BW plots receiving the appropriate wastewater. These plots were then maintained at a depth of about 8 cm, so the soil was thoroughly soaked. On July 1, the necessary P and K fertilizers were introduced into each plot and incorporated into the surface 15 cm soil layer with a ploughing and leveling operation. Meanwhile, N fertilizer was top-dressed twice at 10 and 40 days after transplanting.

Transplanting the late rice crop, which for this part of southeast China was at the beginning of July, was done on July 13, fifteen days after irrigation, with rice seedlings (25-days old, *XiuShui 80*) placed at 15 cm  $\times$  15 cm. Except for the first irrigation, local farmers only irrigated with the reservoirs water, where TP content was very low (0.05 mg L<sup>-1</sup>) and considered negligible; this was done only if there was no rainfall for a long time in order to keep the floodwater level at approximately an 8 cm depth. Also, in this area, Cao et al. [17,18] and Xing et al. [19] found that generally through the whole season there were no runoff events from excess rain. Finally, a single drainage of the paddy-rice-wetland system was accomplished 1 week before the rice harvest on November 13, and the rice was harvested on November 20.

#### 2.4. Sampling and analytical methods

Before the plots were established, soil samples from the surface 0-15 cm layer of the experimental site were taken. These were airdried and sieved (<2 mm) prior to chemical analysis. For P samples  $(in mg L^{-1})$  from paddy water, first 0.25 mL of 4.0 mol mL<sup>-1</sup> HCl was injected into each flask to reduce the sample pH to 2. Then, an irregular interval sampling was conducted according to the growth habit of the rice. Sampling was conducted at 3-day intervals on days 1, 4, 7, and 10 in the early stage (from July 13 to July 22), then at intervals of about 10 d in the middle stage (from July 22 to September 1), and of about 15 days in the late stage (from September 1 to November 13). In addition, samples were collected after the two rainfall events on July 20 and August 18. For each plot, each sample was composed of six to eight random extractions from the floodwater using a syringe and then combined to form a composite plot sample (about 150 mL). Simultaneously, floodwater depth in each plot was measured with a ruler fixed to and extending from a brick that was buried level with the soil surface. The last sampling was taken during drainage on November 13.

Water samples were frozen  $(-6 \,^{\circ}\text{C})$ . Then, prior to analysis, they were thawed. Water samples were digested by  $K_2S_2O_8$ , TP was determined on unfiltered samples using the spectrophotometric method with a continuous-flow automated analyzer (AA3, BRAN+LUEBBE, Germany) at the wave length of 700 nm. After water samples were oxidized with  $K_2S_2O_8$ , TN was measured by double wave length (220 and 275 nm) ultraviolet spectrophotometric method. The determination of K in the water samples by the flame atomic absorption spectrophotometry was carried out at the wave length of 700 nm. TC and FC were determined by the multi-tube zymolytic method, according to the standard methods [20].

For the soil analyses, pH was measured using a pH meter with soil:distilled water = 1:5; CEC was determined with an Un-buffered Salt Extraction Method. Redox potential (Eh) was obtained using a pH meter with a platinum electrode, whereas organic carbon was determined using the Walkley and Black Method. Total N and available N were measured with the Kjeldahl Method, total P and



Fig. 1. Layout of paddy-rice-wetland system plots showing five P treatments with three replicates.

available P with the Molybdenum Method, and available K was extracted by un-buffered ammonium chloride then tested with an atomic absorption spectrophotometer according to the methods described in Sparks et al. [21].

The rice plants at different growing stage were sampled, purified by deionized water, and air dried, then its roots, shoots, leaves and grains were digested with  $H_2SO_4-H_2O_2$  and measured by colorimetric analysis on atomic absorption spectrophotometry.

#### 2.5. Statistical analysis of data

In Tai Lake Basin, there have been several thousand years for the rice planting, and the concentration of P in the rainfall is very low  $(0.03 \text{ mg L}^{-1})$  and negligible. Meanwhile, due to P content in the evaporation is below the detection limit, the evapotranspiration can be neglected in this experiment. So the input of P in the paddyrice-wetland system comes from RW or P fertilizer, and the output of P includes the rice, the paddy soil, and the drainage on November 13.

TP load is the P amount of the floodwater in the paddy-rice-wetland system per unit area (kg ha<sup>-1</sup>) and can be calculated with Eq. (1):

$$TP \text{ load} = \frac{10CV}{A} \tag{1}$$

where C: P concentration of the floodwater in each plot  $(mgL^{-1})$ , A: the area of each plot  $(m^2)$ , V: the volume of the floodwater for each plot  $(m^3)$ , and calculated by multiplying the area  $(m^2)$  of each plot by the depths (cm) of the floodwater of each plot, and 10: the coefficiency of the equation.

TP removal rates in paddy-rice-wetland system are calculated with Eq. (2):

TP removal rate = 
$$\frac{C_0 V_0 - C_1 V_1}{C_0 V_0} \times 100\%$$
 (2)

where  $C_0$ ,  $C_1$ : the initial and final P concentration and of the floodwater in each plot (mg L<sup>-1</sup>),  $V_0$ ,  $V_0$ : the initial and final volume of the floodwater for each plot (m<sup>3</sup>), and calculated the same as in Eq. (1).

## 304 **Table 1**

Rice grain yields in different treatments, mean  $\pm$  SD, n = 3.

Treatments <sup>a</sup>	Rice grain yield <sup>b</sup> (kg ha <sup>-1</sup> )
CK (control)	6921.5 ± 733.9c
SW (surface reservoir water + fertilizer)	$8121.9 \pm 423.6b$
GW (grey water + fertilizer)	$9324.3 \pm 272.9a$
DW (domestic water + fertilizer)	$8647.9 \pm 358.1 ab$
BW (black water + fertilizer)	$8353.3\pm242.8ab$

<sup>a</sup> CK:  $5 \text{ kg P ha^{-1}}$  given by surface reservoir water and no P fertilizer; SW:  $40 \text{ kg P ha^{-1}}$  given by surface reservoir water and superphosphate; DW:  $40 \text{ kg P ha^{-1}}$  given by domestic wastewater and superphosphate; BW:  $40 \text{ kg P ha^{-1}}$  given by black water and superphosphate; GW:  $40 \text{ kg P ha^{-1}}$  given by grey water and superphosphate.

<sup>b</sup> Means  $\pm$  standard deviations within the same column followed by the same letter are not significantly different (*P*>0.05) by Duncan's multiple range test.

Soil percentage (%) of TP load reflects the contribution degree of the paddy soil to TP load in the floodwater, and is expressed with Eq. (3):

soil percentage of TP load = 
$$\frac{C_{S1} - C_{S0}}{C_{S0}} \times 100\%$$
 (3)

where  $C_{S0}$ ,  $C_{S1}$ : the initial and sampled soil total P concentration in each plot (mg kg<sup>-1</sup>).

Fertilizer percentage (%) of TP load represents the contribution degree of the applied fertilizer to TP load in the floodwater, which can be calculated with Eq. (4):

fertilizer percentage of TP load = 
$$\frac{C_{\text{sample}} - C_{\text{CK}}}{C_{\text{CK}}} \times 100\%$$
 (4)

where  $C_{\text{sample}}$ ,  $C_{\text{CK}}$ : the sampled treatment and CK treatment TP concentration after the fertilizer addition (mg L<sup>-1</sup>). So the wastewater percentage (%) of TP load means the contribution degree of the added wastewater to TP load in the floodwater, which can be calculated with Eq. (5):

wastewater percentage of TP load

$$= \left(1 - \frac{C_{S1} - C_{S0}}{C_{S0}} - \frac{C_{sample} - C_{CK}}{C_{CK}}\right) \times 100\%$$
(5)

The parameters have the same meaning as the aforementioned ones. A one-way ANOVA was conducted to test the effects of P from RW in the paddy-rice-wetland system on rice yield, TP concentration, and TP load. Statistical differences between treatments were determined using Duncan's multiple range test at the 0.05 probability level.

#### Table 2

TP concentration and removal rate in the paddy-rice-wetland system.

#### 3. Results

#### 3.1. Rice yield

Yield for CK was significantly less ( $P \le 0.05$ ) than SW, GW, DW, and BW with yield of GW significantly greater ( $P \le 0.05$ ) than all treatments (Table 1). Meanwhile, there were no significant differences (P > 0.05) between yields of RW treatments GW and DW, which were significantly higher than that of SW. Also, the mean yield of CK was about 6.8% lower than the local average yield (7500 kg ha<sup>-1</sup>) for 2007.

#### 3.2. TP concentration in the floodwater

On July 13, the day of P application, mean TP concentration in the floodwater of SW, BW, DW, and GW was significantly greater  $(P \le 0.05)$  than CK (Table 2). On day 7, July 19, mean TP concentration in mg L<sup>-1</sup> had declined from 7.94 to 2.10 for SW, from 7.27 to 1.45 for GW, from 6.77 to 1.22 for DW, and from 6.24 to 0.84 for BW (Fig. 2). Although it rained 20 mm on July 20 and 14 mm on August 18, both events produced no runoff, and P concentration of the rainwater (0.03 mg L<sup>-1</sup>) was negligible. Also, for all treatments there was no significant decrease (P > 0.05) in TP concentration measured after each event (Table 2, Fig. 2). From October 15 to final drainage on November 13, for each treatment there were no significant increases (P > 0.05) in TP concentration in paddy water; over this time all treatments ranged from 0.04 to 0.4 mg L<sup>-1</sup>.

TP removal rates for all treatments ranged from 75.2% to 98.2% with CK significantly less ( $P \le 0.05$ ) than other treatments (Table 2). Also, the TP removal rates from RW (GW, DW, and BW), ranging in the order of GW > DW > BW from 60.3% to 71.4%, was significantly higher ( $P \le 0.05$ ) than that from superphosphate fertilizer (SW), ranging from 26.8% to 36.7%.

#### 3.3. TP load in the floodwater

On July 13, TP load in paddy water, except for CK, was at its maximum (Fig. 3). Then, TP load together with TP concentration declined with time and could be characterized with nonlinear regression equations (Table 3), whereupon it became approximately steady from October 1 until drainage on November 13. There were no significant nonlinear relationships between the P loads and time for the CK treatment. Two days after the moderate rainfall events of July 20 and August 18, TP loads were not significantly different (P > 0.05) from before the rainfalls with SW significantly greater ( $P \le 0.05$ ) than the RW plots (Fig. 3).

From soil, wastewater, and fertilizer of the floodwater on July 13 just after P fertilization (which did not include the CK plots), TP load for fertilizer (ranging from 49.3% to 81.1%) in SW, GW, DW, and BW was significantly higher ( $P \le 0.05$ ) than that of wastewater (rang-

Treatments <sup>a</sup>	TP concentration (mg	g L <sup>-1</sup> )			TP removal rate (%)		
	July 13 <sup>b</sup>	July 20	August 18	November 13	Wastewater	Fertilizer	Total
СК	0.13d <sup>c</sup>	0.15d	0.06c	0.03d	75.20a <sup>d</sup>	0d	75.20b
SW	7.94a	1.92a	0.83a	0.26a	56.73d <sup>d</sup>	40.05a	96.78a
GW	7.27ab	1.13b	0.61b	0.22ab	71.36ab <sup>e</sup>	26.81c	98.17a
DW	6.77bc	0.97bc	0.48b	0.18b	65.43bc <sup>e</sup>	31.85bc	97.28a
BW	6.24c	0.70c	0.34b	0.11c	60.29cd <sup>e</sup>	36.75ab	97.04a

<sup>a</sup> CK: 5 kg P ha<sup>-1</sup> given by surface reservoir water and no P fertilizer; SW: 40 kg P ha<sup>-1</sup> given by surface reservoir water and superphosphate; DW: 40 kg P ha<sup>-1</sup> given by domestic wastewater and superphosphate; BW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by grey water and superphosphate. <sup>b</sup> July 13, rice seedlings transplanting; July 20, the first rainfall; August 18, the second rainfall; November 13, draining.

<sup>c</sup> Means within the same column followed by the same letter are not significantly different (*P*>0.05) by Duncan's multiple range test.

<sup>d</sup> Surface reservoir water (SW).

e Rural wastewater.



Fig. 2. Temporal changes of TP concentration in the paddy-rice-wetland system floodwater in different treatments. The vertical bars are standard deviations of the means, *n* = 3.



Fig. 3. Variations of TP load in the paddy-rice-wetland system floodwater in different treatments. The vertical bars are standard deviations of the means, n = 3.

ing from 10.8% to 46.1%) and soil (ranging from 4.5% to 8.1%), which made fertilizer the main P source in these treatments (Table 4). For CK the opposite order (soil, followed by wastewater, and no fertilizer) was observed. From July 13 to August 18, wastewater and fertilizer TP load sources generally followed a gradual increase as soil TP loads decreased. Then from August 18 to November 13, except for CK, the percentage of TP load from wastewater decreased (ranging from 24.9% to 48.2%), whereas the percentage for fertilizer continued to increase (ranging from 73.1% to 89.8%).

#### 4. Discussion

#### 4.1. Rice yield response for different P treatments

Compared to the four treatments receiving P additions, the significantly lower rice grain yield for the CK treatment (Table 1) indicated that supplemental P was necessary in this area for normal rice production. This differs from studies of dry–wet alternate irrigation systems that do not add P and yet still have high yield

Table 3

Regression equations relating TP concentration and TP load in the paddy-rice-wetland system floodwater with respect to time (*t*, days) from July 13 to the drainage on November 13.

Treatments <sup>a</sup>	TP concentration $(y, mg L^{-1})$		TP load (y, kg ha <sup>-1</sup> )				
	Regression equation	$r^2$	Regression equation	r <sup>2</sup>			
СК	$y = 0.2316t^{-0.3653}$	0.1662	$y = 0.1906t^{-0.398}$	0.1264			
SW	$y = 11.663t^{-0.8017}$	0.8623**	$y = 8.7008t^{-0.7975}$	$0.6295^{*}$			
GW	$y = 12.8384t^{-0.8559}$	0.9808**	$y = 5.0196t^{-0.8498}$	0.9563**			
DW	$y = 7.1905t^{-0.7387}$	0.9297**	$y = 5.4991t^{-0.7507}$	0.9202**			
BW	$y = 8.0469t^{-0.7064}$	0.9035**	$y = 5.8466t^{-0.7074}$	0.8951**			

<sup>a</sup> CK: 5 kg Pha<sup>-1</sup> given by surface reservoir water and no P fertilizer; SW: 40 kg Pha<sup>-1</sup> given by surface reservoir water and superphosphate; DW: 40 kg Pha<sup>-1</sup> given by domestic wastewater and superphosphate; BW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg Pha<sup>-1</sup> given by grey water and superphosphate.

\* Significant at the 0.05 probability level by Duncan's multiple range test.

\*\* Significant at the 0.01 probability level by Duncan's multiple range test.

## 306 **Table 4**

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Percentage (%) of IP load in the	naddy_rice_wetland syst	em floodwater from various so	ircec
i cicciitage (70) of it ibad in the			urces.

Treatments <sup>a</sup>	July 13 <sup>b</sup>			July 20			August 18			November 13		
	Soil	Wastewater	Fertilizer	Soil	Wastewater Fertili		Soil	Wastewater	Fertilizer	Soil	Soil Wastewater	
СК	77.26a <sup>c</sup>	22.74c <sup>d</sup>	0e	63.87a	36.13c <sup>d</sup>	0e	52.76a	47.24b <sup>d</sup>	0e	54.18a	45.82a <sup>d</sup>	0e
SW	8.11b	10.78d <sup>d</sup>	81.11a	-13.72b	26.53d <sup>d</sup>	87.19a	-16.22b	28.14e <sup>d</sup>	88.09a	-14.72b	24.94d <sup>d</sup>	89.78a
GW	4.54b	46.14a <sup>e</sup>	49.32d	-20.83d	53.92a <sup>e</sup>	66.91d	-23.93e	52.84a <sup>e</sup>	71.09d	-21.34d	48.23a <sup>e</sup>	73.11d
DW	6.67b	32.85b <sup>e</sup>	60.48c	-18.47c	44.86b <sup>e</sup>	73.61c	-19.89d	43.73c <sup>e</sup>	76.16c	-18.45c	39.89b <sup>e</sup>	78.56c
BW	7.34b	21.52c <sup>e</sup>	71.14b	-17.28c	37.23c <sup>e</sup>	80.05b	-18.17c	38.23d <sup>e</sup>	79.94b	-17.81c	32.35c <sup>e</sup>	85.45b

Negative values mean the P absorption of the paddy soil.

<sup>a</sup> CK: 5 kg P ha<sup>-1</sup> given by surface reservoir water and no P fertilizer; SW: 40 kg P ha<sup>-1</sup> given by surface reservoir water and superphosphate; DW: 40 kg P ha<sup>-1</sup> given by domestic wastewater and superphosphate; BW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by grey water and superphosphate.

<sup>b</sup> July 13, rice seedlings transplanting; July 20, the first rainfall; August 18, the second rainfall; November 13, draining.

<sup>c</sup> Means within the same column followed by the same letter are not significantly different (*P*>0.05) by Duncan's multiple range test.

<sup>d</sup> Surface reservoir water (SW).

<sup>e</sup> Rural wastewater.

[22,23]. It also differs from an irrigated paddy agricultural system receiving fertilizer and P from manure [24,25], where soil P is very high and the soil can release sufficient P. At the same time, yields with standard fertilizer applications for this area (SW) were significantly lower ( $P \le 0.05$ ) than that of RW treatments (GW, DW), which were significantly less ( $P \le 0.05$ ) than that of BW. This meant that irrigating with RW in the paddy rice field could reduce the amount of commercial P fertilizer used as well as the total irrigation water, thereby decreasing operational costs. Meanwhile, the significantly greater output of BW would mean greater revenues than the normal farmer-applied-fertilizer scheme of SW, whereas GW and DW would have higher revenues than SW, but lower costs. This would make RW a feasible alternative to irrigation water when considering costs and revenues.

# 4.2. TP dynamics in the floodwater of the paddy-rice-wetland system

Application of P fertilizer significantly increased ( $P \le 0.05$ ) floodwater P concentration (Fig. 2), and these effects were more pronounced with fertilizer application rate. The increased TP concentrations following P application as the combined application of RW and fertilizer declined more readily than did those resulting from fertilizer only. The combination treatment gave rise to significantly lower ( $P \le 0.05$ ) floodwater P concentration than did the same rate of P applied as fertilizer only about 1 week after application. Two reasons were particularly responsible for the subsequent decrease. Firstly, P was absorbed by the rice plant which had entered the fast growing stage. The relative growth rate of rice reached to 67.2, 55.3, 49.8, and 42.8 mg  $g^{-1}$  day<sup>-1</sup> for GW, DW, BW and SW, respectively, while it was only  $32.4 \text{ mg g}^{-1} \text{ day}^{-1}$  in CK, implying that under the same P fertilizer application level, P from RW can be absorbed by rice plant prior to P from fertilizer [26], because orthophosphate can be easily chelated with Ca<sup>2+</sup>, Fe<sup>3+</sup> of the blue-purple paddy soil and formed to difficult dissolved materials in Tai Lake Basin, but organic phosphorus from RW can be absorbed by the

rice. Secondly, reduced conditions in the cultivated layer were completely developed, which can change organic forms of P to inorganic forms through microbial degradation of organic matter (mainly *Agrobacterium*) [27], and P was easily precipitated [28–30]. Redox potential of seepage waters throughout the rice growing season showed that Eh (approximately 83 mV) was initially positive in the paddy soil to a depth of 20 cm immediately after flooding, then rapidly decreased to highly reduced conditions (about –225 mV).

Although it rained on July 20 (20 mm) and August 18 (14 mm, both with no runoff produced), TP concentration slightly decreased (Fig. 2), implying that some soil-adsorbed fertilizer P was released into the floodwater, since P in the rainwater  $(0.05 \text{ mg L}^{-1})$  was negligible. After September 2, TP concentrations in the floodwater reached a constant value, ranging from 0.04 to 0.4 mg L<sup>-1</sup>, and were not linked to applied P levels. However, several days before the final drainage on November 13, TP concentration rose a little, due to the release P by the paddy soil (Fig. 4b). Results indicated clearly that paddy-rice-wetland system can remove massive P from irrigated RW and P fertilizer applied. The drainage from the rice-paddywetland system before harvest only contained very little P (Fig. 2), far lower than wastewater discharge permission  $(1.0 \text{ mg L}^{-1})$  and data reported in similar researches [16,17], which showed that rice plant absorbed plenty of P, especially at the tillering stage and booting stage, shown as Table 5.

TP removal rate of RW was significantly higher ( $P \le 0.05$ ) than that of superphosphate (Table 2), showing that P of RW could be removed more easily by the paddy-rice-wetland system than P of fertilizer, one possibility could be the inorganic form P in RW would be present as small molecular weight molecules and therefore would be preferentially absorbed by the plants [7,31]. Another reason may be that the sorption of exchangeable P onto fine particulate material was unstable and could be desorbed easily, when the floodwater condition of the paddy-rice-wetland system changed, due to the rainfall or the drainage, just shown as P adsorption experiments (Fig. 4), which was in agreement with the results obtained by Zhang et al. [15,16] in the same soil. In addition, the composi-



Fig. 4. Adsorption and desorption isotherm of soil P.

#### Table 5

P content in different organs of rice during the whole growing season of 2007 (mg kg<sup>-1</sup>).

Treatments <sup>a</sup>	Seedling stage <sup>b</sup>			Tillering stage			Booting stage				Mature stage					
	Root	Shoot	Leaf	Grain	Root	Shoot	Leaf	Grain	Root	Shoot	Leaf	Grain	Root	Shoot	Leaf	Grain
СК	0.12 <sup>c</sup>	0.054	0.014	_d	0.11	0.048	0.24	_	0.17	0.12	0.13	0.08	0.011	0.007	0.004	0.17
SW	0.17	0.076	0.020	-	0.16	0.064	0.34	-	0.25	0.18	0.16	0.12	0.018	0.018	0.014	0.28
GW	0.31	0.135	0.036	-	0.29	0.113	0.61	-	0.43	0.32	.030	0.21	0.024	0.036	0.026	0.45
DW	0.24	0.105	0.028	-	0.22	0.088	0.47	-	0.33	0.25	0.24	0.16	0.021	0.032	0.021	0.38
BW	0.21	0.092	0.023	-	0.20	0.077	0.42	-	0.31	0.21	0.19	0.15	0.020	0.029	0.020	0.34

<sup>a</sup> CK: 5 kg P ha<sup>-1</sup> given by surface reservoir water and no P fertilizer; SW: 40 kg P ha<sup>-1</sup> given by surface reservoir water and superphosphate; DW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given by black water and superphosphate; GW: 40 kg P ha<sup>-1</sup> given by given b

<sup>b</sup> Seedling stage: from July 13 to 19; tillering stage: from July 20 to August 20; booting stage: from August 21 to October 15; mature stage: from October 16 to November 13. <sup>c</sup> Mean values within nine samples (sampling for three times with three replicates, *n* = 9).

<sup>d</sup> Not detected.

tion of the different types of the wastewater used could affect the TP removal rate, especially nitrogen content and pH. The higher nitrogen concentration of the wastewater was, the lower the TP removal rate was, which perhaps have a relation with the antagonism between N and P content of the wastewater. But the study of pH effect showed that alkaline pH could promote the TP removal rate, and the TP removal rate of GW (pH 9.4) was higher than that of BW (pH 6.2).

# 4.3. TP load dynamics in the floodwater of the paddy-rice-wetland system

On July 13, just after P fertilizer application, TP load in the floodwater, except for CK, was at its maximum (Fig. 3), showing a close relationship between TP load and the P fertilizer application rate, and little P released from the paddy soil. Then, from July 13 to August 18, the soil continuously adsorbed P, so the main TP load sources from wastewater and fertilizer gradually increased as TP loads from soil decreased and showed negative values (Table 4), indicating that the paddy soil adsorbed some P, due to the high P concentration in the floodwater. Another possible contributing factor to the increase in TP load for wastewater and fertilizer during this period was the two rainfall events of July 20 and August 18. Zhang et al. [15,16,32] found that hydraulic erosion (from rainfall) causes a release of particulate P into paddy water. However, in this study, 2 days after the rainfall events, there were no significant differences (P>0.05, not shown) in TP loads. Afterward, the TP load decreasing from August 18 to November 13 for RW and increasing for fertilizer (Table 4), revealed that P from RW could be more easily removed than P from fertilizer.

Since rainfall became the sole water supply to the plots during this period and P in rainwater was negligible, improper water management of floodwater drainage could cause an appreciable loss of P. If this were the case, the order of P loss in runoff would be SW > RW (Fig. 3). Therefore, using GW, DW, or BW instead of SW as an irrigation water source would not only decrease commercial P fertilizer usage and improve P use efficiency, but also reduce the danger of P runoff into surface water bodies minimizing the risk of environmental pollution. This would be ecologically beneficial to the health of the environment.

#### 5. Conclusions

This study describes a P removal experiment from irrigated RW in the paddy-rice-wetland system in Yuhang around the Tai Lake Basin. As noted, supplemental P was necessary in this area for normal rice production and standard fertilizer applications were not significantly different from GW and DW. However, irrigating with RW in the paddy rice field could reduce the costs of fertilizer and irrigation water. Additionally using BW would significantly increase yield revenues. The paddy-rice-wetland system also removed large quantities of P due to irrigation with RW and applied P fertilizer with TP removal rates for RW much higher than for superphosphate fertilizer. Thus, the application of RW instead of P fertilizer could greatly decrease the discharge of P into water bodies. Considering the growing scarcity of water resources and the increasing algal cover due to excess nutrient resources, using RW as irrigation water and as a P source could be a great opportunity to improve the agricultural economy and minimize the risk of environmental pollution, especially potential eutrophication, while fostering a healthy ecosystem. So, the paddy-rice-wetland system can be considered as a new method in RW treatment, especially for the Tai Lake Basin. If the paddy-rice-wetland system can be widely used and managed effectively, the emission of P from RW into Tai Lake can be greatly reduced, and water quality can be improved. But the determination of what exactly is the largest P-removal capacity of the paddy-rice-wetland system per unit and how rice grain quality is affected by RW irrigated the paddy-rice-wetland system still needs future work.

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