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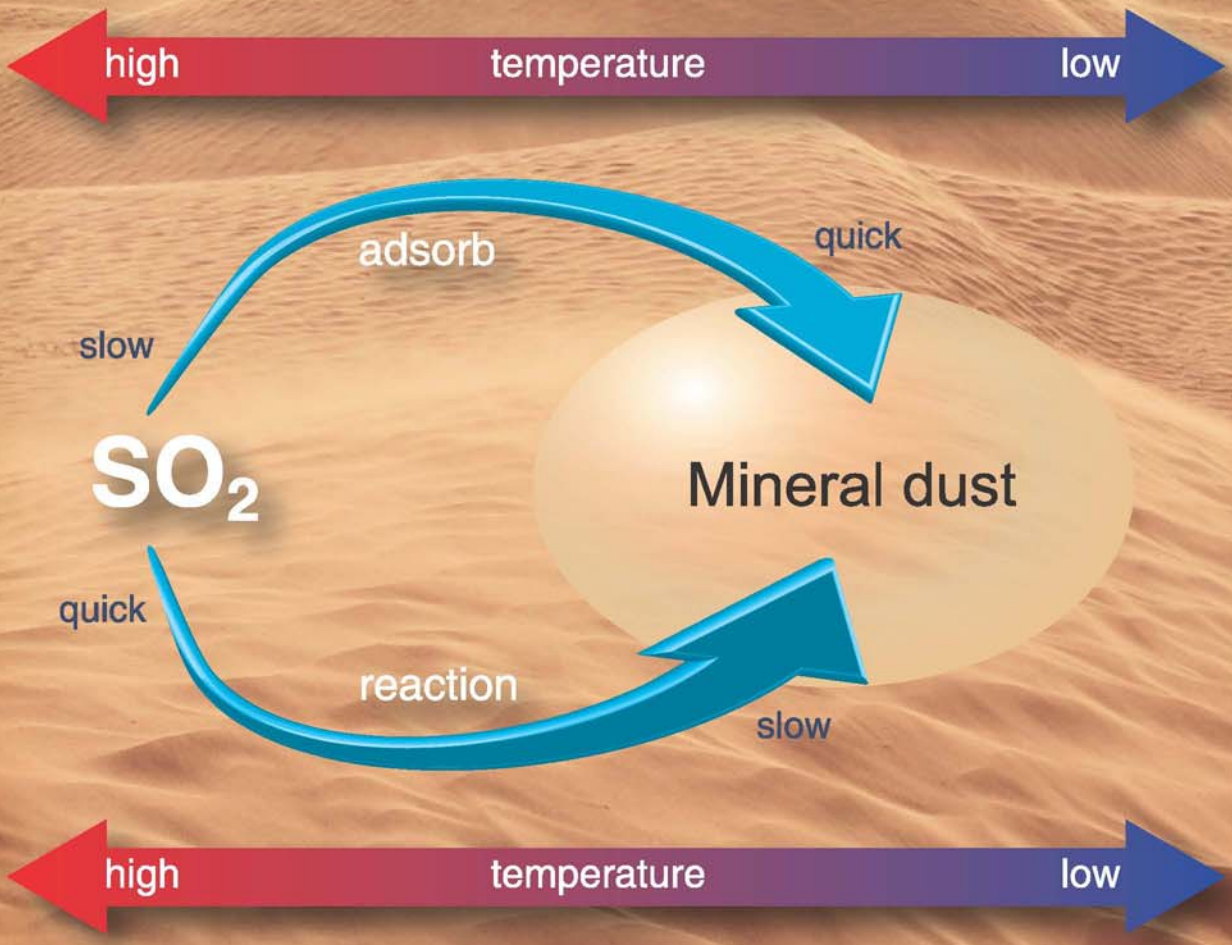

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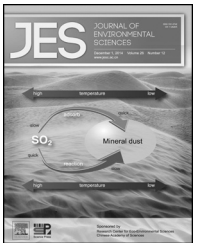

Influence of the inherent properties of drinking water treatment residuals on their phosphorus adsorption capacities

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ABSTRACT

Batch experiments were conducted to investigate the phosphorus (P) adsorption and desorption on five drinking water treatment residuals (WTRs) collected from different regions in China. The physical and chemical characteristics of the five WTRs were determined. Combined with rotated principal component analysis, multiple regression analysis was used to analyze the relationship between the inherent properties of the WTRs and their P adsorption capacities. The results showed that the maximum P adsorption capacities of the five WTRs calculated using the Langmuir isotherm ranged from 4.17 to 8.20 mg/g at a pH of 7 and further increased with a decrease in pH. The statistical analysis revealed that a factor related to Al and 200 mmol/L oxalate-extractable Al (Al_{ox}) accounted for 36.5% of the variations in the P adsorption. A similar portion (28.5%) was attributed to an integrated factor related to the pH, Fe, 200 mmol/L oxalate-extractable Fe (Fe_{ox}), surface area and organic matter (OM) of the WTRs. However, factors related to other properties (Ca, P and 5 mmol/L oxalate-extractable Fe and Al) were rejected. In addition, the quantity of P desorption was limited and had a significant negative correlation with the (Fe_{ox} + Al_{ox}) of the WTRs ($p < 0.05$). Overall, WTRs with high contents of Al_{ox}, Fe_{ox} and OM as well as large surface areas were proposed to be the best choice for P adsorption in practical applications.
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Introduction

Excessive phosphorus (P) in water bodies is one of the main causes of eutrophication (Edwards and Withers, 2007). Chemical adsorption can remove P effectively. Common adsorbents include natural minerals, engineering materials and industry by-products (Pant et al., 2001; Liu et al., 2007). Drinking water treatment residuals (WTRs) are inevitable by-products of drinking water treatment plants, which have attracted substantial attention in recent years due to their high P adsorption capacity. The reuse of WTRs to remove P is a win–win technology for waste management and water environment restoration. Several applications for WTRs have been developed (Agyin-Birikorang et al., 2007; Zhao et al., 2009; Wang et al., 2012a). Therefore, a comprehensive understanding of the P adsorption capacity of WTRs is crucial to their effective utilization.

The factors affecting the P adsorption capacity of WTRs are mainly derived from two categories. One is solution chemistry, including pH, temperature, dissolved organic carbon, low molecular weight organic acids and P species (Razali et al., 2007; Wang et al., 2012b; Gao et al., 2013). The pH has been considered the most important factor, exhibiting a negative correlation with the adsorption capacity of WTRs (Wang et al., 2011a). The other category is the inherent properties of the WTRs, such as their surface area, particle size and contents of Fe, Al, P, Ca and other elements (Dayton and Basta, 2005; Makris et al., 2005). The physicochemical characteristics of WTRs are related to the source

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water, the chemicals used for treatment and the strategy of operations. However, limited data are available regarding the influence of the inherent properties on the P adsorption of WTRs.

Five types of WTRs were collected from different regions in China. Their physicochemical properties were determined. Batch experiments were carried out to investigate the P adsorption and desorption on different WTRs. Afterward, the integrated relationship of the inherent properties (contents and fractions of elements, pH and surface area) and P adsorption characteristics of the WTRs was calculated using statistical analysis. This work could be beneficial toward understanding P adsorption on WTRs and promoting their practical application.

1. Materials and methods

1.1. Preparation and characterization of the WTRs

The BJ1-WTRs and BJ2-WTRs were sampled from the Beijing NO. 9 Water Treatment Plant in 2011 and 2012, respectively. This plant used a combination of $FeCl_3$ and polymeric aluminium (PAC) as a coagulant, while supplying 60% of the water requirement for Beijing. Active carbon was used as an adsorbent for advanced treatment. The HZ-WTRs and LZ-WTRs were sampled from the Hangzhou Yuxi Water Treatment Plant and Lanzhou Water Treatment Plant, respectively. Both plants used PAC as a coagulant. The SD-WTRs sampled from the Shandong Yuxing Water Treatment Plant used PAC as a coagulant and $Ca(HCO_3)_2$ as a softening agent. The five WTRs were air-dried and sieved (<2 mm) to create homogeneous sub-samples before analysis.

The total Fe, Al and Ca contents of the WTRs were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES, Jobin Yvon, Paris, France) according to USEPA-METHOD 3051. Total P was determined using ICP after digestion by a mixture of HNO_3, $HClO_4$ and HCl. The 200 mmol/L oxalate-extractable Fe (Fe_{ox1}), Al (Al_{ox1}), and P (P_{ox1}), and the 5 mmol/L oxalate-extractable Fe (Fe_{ox2}), Al (Al_{ox2}), and P (P_{ox2}) were determined by ICP after extraction at a 1:60 solid:solution ratio following the procedures described in a previous study (Schoumans, 2000). The phosphorus saturation index (PSI) was determined using the $P_{ox1}/(Al_{ox1} + Fe_{ox1})$, where the units of P_{ox1}, Al_{ox1} and Fe_{ox1} are mol/kg (Elliott et al., 2002). The Mehlich 3 extractable Ca (Ca_M) was determined as described in the literature (Mehlich, 1984). The total carbon was determined using the Dumas method with a 1500 series dry combustion analyzer (Crilo Erba, Milan, Italy). The organic matter (OM) was determined using a potassium dichromate oxidation method (Nelson and Sommers, 1982). The pH was determined in a 1:2 WTRs to 0.01 mol/L $CaCl_2$ solution. The electrical conductivity (EC) was determined in a 1:2 WTRs to deionized water solution. The SEM, XRD and surface area were analyzed to determine the structure of the different WTRs. The tests were repeated twice, and the average values are reported.

1.2. Adsorption characteristics

For adsorption experiment, P working solutions with initial P concentrations (P_0) at eight common levels (5, 10, 15, 20, 30, 40, 50 and 100 mg/L) were prepared by dissolving pre-determined amounts of KH_2PO_4 in a 0.01 mol/L KCl solution.

The batch experiments were conducted by pouring 0.5 g of the WTRs and 50 mL of the P working solutions with different P_0 into 100 mL plastic bottles. The pH values of the P working solutions were adjusted to 5.0, 7.0 and 9.0 with 0.01 mol/L HCl and 0.01 mol/L NaOH. The mixed samples were shaken at 200 r/min for 48 hr to attain equilibrium (Wang et al., 2012b). Afterward, the samples were removed from the shaker and filtered using a 0.45-μm millipore membrane filter to separate the solid from the liquid. The residual P in suspensions was measured using an ammonium molybdate spectrometry method. The amount of P adsorbed (Q) was determined using Eq. (1):

$$Q = \frac{(P_0 - P_e)V}{m} \quad (1)$$

where, P_0 (mg/L) and P_e (mg/L) are the initial P and final P concentrations, respectively; V (L) is the solution volume, and m (g) is the mass of WTRs.

The desorption was facilitated by adding 50 mL of 0.01 mol/L KCl solution to 0.5 g of P-loaded WTRs from adsorption test that had been washed with ethanol three times. The initial pH was maintained at 7. Afterward, the samples were shaken at 200 r/min for 48 hr. The P concentrations of the suspensions were measured. The amounts of P desorbed (Q_d) were also determined using Eq. (1).

The Langmuir isotherm was used:

$$Q_e = \frac{Q_{max} b P_e}{1 + b P_e} \quad (2)$$

where, Q_e (mg/g) is the mass of P adsorbed per unit mass of WTRs, b is an empirical constant related to the entropy, Q_{max} (mg/g) is the maximum adsorption capacity, and P_e (mg/L) is the equilibrium P concentration. It has been proved that the P adsorption by WTRs fits the Langmuir isotherm well (Dayton and Basta, 2005; Wang et al., 2011a). The Q_{max} was calculated by fitting Eq. (2) to the experimental data.

1.3. Statistical analyses

The statistical analyses were performed using SPSS software version 20 (SPSS Inc., Chicago, Illinois, USA). The measured inherent characteristics of the five WTRs were analyzed using rotated principal component analysis. This procedure reduced the number of independent variables from 12 individual abiotic variables to three or four principal component factors, which were independent linear combinations of the 12 original variables. The resulting factor scores and pH values of the P working solution were used as independent variables during the multiple regression analysis while using the P removal properties of the five WTRs as dependent variables. Afterward, the relationship between the inherent properties of the WTRs and their P adsorption capacities was obtained (Arias et al., 2001).

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2. Results and discussion

2.1. Characteristics of WTRs

The five WTRs had variable physicochemical properties (Table 1). The weight percentage values ranged from 2.5%–9.7% for Fe, 4.2%–9.4% for Al, 0.8%–13% for Ca, 2.9%–10.8% for C, 0.1%–0.3% for P and 2.6%–6.8% for OM. The HZ-WTRs, LZ-WTRs and SD-WTRs had a higher weight percentage of Al than Fe. The weight percentages of Ca were low, except that the percentages in the LZ-WTRs and SD-WTRs were 5% and 13%, respectively. The Fe and Al of the WTRs depended on the species and dose of the coagulants during water treatment. The Ca and C were derived from the Ca and suspended solids in the raw water, respectively. Furthermore, the Ca content increased significantly when $Ca(HCO_3)_2$ was used as a softening agent (Gibbons and Gagnon, 2011).

The range of P contents in five WTRs was similar to the result of a former study encompassing 21 WTRs in Oklahoma utilities (0.2–4.0 mg/g) (Dayton et al., 2003). The inherent P of the WTRs was higher than that of common soil because the P in the raw water accumulated after treatment (Elliott et al., 2002). The Fe_{ox1}, Al_{ox1} and P_{ox1} of five WTRs accounted for 14%–84%, 36%–94% and 14%–80% of the total Fe, Al and P, respectively. The PSIs of the five WTRs were relatively low (≤2.69%). Moreover, the contents of Fe_{ox2}, Al_{ox2} and P_{ox2} were limited: 0.01–0.11 mg/g for Fe_{ox2}, 0.03–0.33 mg/g for Al_{ox2} and 0.01 mg/g for P_{ox2}. The Ca_M was approximately 42%–92% of the total Ca, and the SD-WTRs had the highest value. The oxalate-extractable Fe and Al were closely related to the structure of the WTRs. The amorphous structure of the WTRs was affirmed by the high fraction of Fe_{ox1} and Al_{ox1}. Moreover, the Fe_{ox2} and Al_{ox2} were uncorrelated with the Fe_{ox1} and Al_{ox1}. For instance, the Fe_{ox1} of the BJ2-WTRs was higher than that of the HZ-WTRs, while the Fe_{ox2} was lower. Similarly, the Al_{ox1} of the LZ-WTRs was lower than that of the SD-WTRs, while the Al_{ox2} was higher. The lack of correlation indicated that the Fe_{ox2} and Al_{ox2} may be dependent on other factors, such as the aging effect (Agyin-Birikorang and O'Connor, 2009). In addition, the average PSI of the five WTRs (<1.64%) was a useful tool to characterize P saturation, and suggested that the inherent P content was far from the saturated adsorption capacity. Consequently, it was concluded that the five WTRs had a large potential for P adsorption.

The ECs of the five WTRs were 0.50–1.25 mS/cm, making them well below the 4.00 ms/cm associated with the salinity of soil for salt-sensitive plants (Brady and Weil, 1996). Moreover, a leaching test was conducted and the results showed that the five WTRs were non-hazardous (Table S1). The pH values of the five WTRs were moderate at 7.23–7.90. The surface areas of the BJ1-WTRs (74 m^2/g) and BJ2-WTRs (61 m^2/g) were larger than the others (21–52 m^2/g) because the activated carbon residuals had an expanded surface area. In addition, the XRD and SEM analyses showed that the five WTRs were amorphous and had rough surfaces (Appendix A Fig. S1), making P easily transportable to the micropores. Therefore, the physical properties of the WTRs were beneficial for adsorbing P.

2.2. Characteristics of adsorption

The residual P concentration in the suspended solution was too low to be detected when P_0 was below 50 mg/L (Appendix A Table S2); therefore, only the adsorption capacities for P_0 of 50 and 100 mg/L are displayed (Fig. 1). The results show that the five WTRs exhibited varying removal efficiencies for P. For a P_0 of 50 mg/L, the BJ2-WTRs had the highest removal efficiency: approximately 100% at pH 5, followed by HZ-WTRs (99%), BJ1-WTRs (88%), SD-WTRs (77%) and LZ-WTRs (74%), while the P removal efficiencies at pH 9 decreased to 97%, 95%, 70%, 58% and 54% respectively. Moreover, the adsorption capacities of the five WTRs for P_0 of 100 mg/L (Q_2) exceeded those for P_0 of 50 mg/L (Q_1). The (Q_2–Q_1) values at pH 5 were 4.76, 4.06, 2.94, 1.85 and

Table 1 – General physicochemical properties of the five drinking water treatment residuals (WTRs).

Properties	BJ1-WTRs	BJ2-WTRs	HZ-WTRs	LZ-WTRs	SD-WTRs
Fe (mg/g)	80.10	97.10	28.13	39.54	25.60
Al (mg/g)	42.20	74.25	94.48	49.79	47.27
Ca (mg/g)	8.21	16.54	4.87	49.65	129.83
C (mg/g)	107.74	106.33	41.41	28.77	62.54
P (mg/g)	1.41	1.31	2.86	1.21	1.91
OM (mg/g)	65.72	68.24	40.50	26.25	44.41
Fe_{ox1} (mg/g)	58.80	82.00	8.90	5.70	6.16
Al_{ox1} (mg/g)	39.69	62.00	70.00	18.00	26.73
P_{ox1} (mg/g)	0.34	0.19	2.29	0.61	0.81
PSI (%)	0.44	0.16	2.69	2.57	2.36
Fe_{ox2} (mg/g)	0.11	0.02	0.05	0.01	0.01
Al_{ox2} (mg/g)	0.33	0.03	0.46	0.04	0.03
P_{ox2} (mg/g)	0.01	0.01	0.01	0.01	0.01
Ca_M (mg/g)	7.55	14.03	3.76	40.28	40.01
EC (ms/cm)	0.74	1.25	0.73	0.72	0.50
Surface area (m^2/g)	74	61	52	34	21
pH	7.23	7.30	7.40	7.90	7.60

BJ1-WTRs: samples collected from Beijing in 2011; BJ2-WTRs: samples collected from Beijing in 2012; HZ-WTRs: samples collected from Hangzhou; LZ-WTRs: samples collected from Lanzhou; SD-WTRs: samples collected from Shandong; OM: organic matter; PSI: phosphorus saturation index; EC: electrical conductivity; ox1: 200 mmol/L oxalate extractant; ox2: 5 mmol/L oxalate extractant; M: Mehlich 3 extractant.

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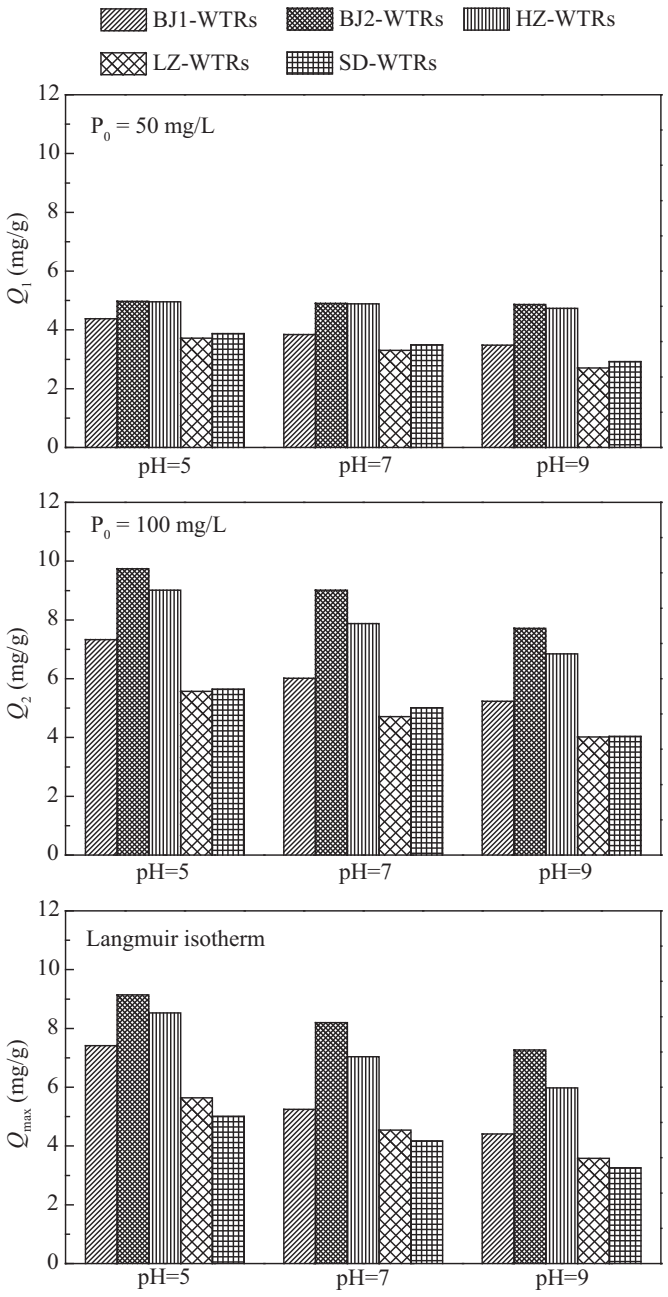

Fig. 1 – P adsorption of the five WTRs at different pH levels.

1.77 mg/g for BJ2-WTRs, HZ-WTRs, BJ1-WTRs, SD-WTRs and LZ-WTRs, respectively. The Langmuir isotherm was utilized to fit the experimental data. The maximum P adsorption capacities (Q_{max}) for the five WTRs at pH 5 were 5.01–9.14 mg/g. The variations in Q_{max} followed a trend similar to Q_1 and Q_2, exhibiting an average decrease of 30% when the pH increased from 5 to 9. Overall, the Q_1, Q_2 and Q_{max} of the five WTRs showed a consistent trend (BJ2-WTRs > HZ-WTRs > BJ1-WTRs > LZ-WTRs > SD-WTRs) at the three pH levels.

The mechanism of P adsorption by WTRs was complicated, including ligand exchange, hydroxide exchange, surface complexation, and co-precipitation. Among them, the ligand exchange between P in solution and reactive functional groups (*i.e.*, –OH, –Cl, $–SO_4$ and humic substances) on the WTRs' surface was reported to be the dominating pathway (Yang et al., 2006). Thus with the increase of solution pH, the change in surface potential and competitive adsorption between the P and –OH reduced the P adsorption capacity (Guan et al., 2007). In addition, the higher adsorption capacity for higher P_0 can be attributed to the increased concentration gradient between the aqueous solution and solid phase, resulting in more contact and diffusion. Overall, the P adsorption process of WTRs was more favorable under acidic conditions.

The desorption of the five P-loaded WTRs at three pH levels followed a similar pattern of adsorption. The P desorption for P_0 < 50 mg/L was also below the detected levels

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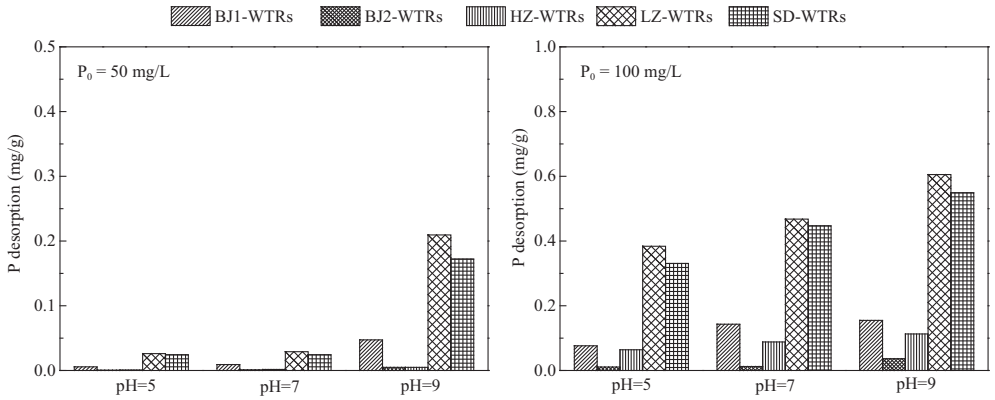

Fig. 2 – P desorption of the five WTRs at different pH levels (P_0: the initial P concentrations).

for BJ1-WTRs, BJ2-WTRs and HZ-WTRs (Table S3); so only the data for P_0 of 50 and 100 mg/L are shown (Fig. 2). For P_0 of 50 mg/L, the LZ-WTRs desorbed the highest proportion of adsorbed P (0.70%) at pH 5, followed by the SD-WTRs (0.62%), BJ1-WTRs (0.13%), HZ-WTRs (0.10%) and BJ2-WTRs (0.10%). An inverse trend was found in the P adsorption and desorption of the five WTRs. The LZ-WTRs desorbed the greatest amount of P while adsorbing the least amount of P. Furthermore, P desorption increased with increasing solution pH and P_0. For instance, for P_0 of 50 mg/L, increasing pH from 5 to 9 increased the P desorption for HZ-WTR from 0.7×10^{-3} to 4.8×10^{-3} mg/L. The P desorption increased further to 0.11 mg/L at P_0 of 100 mg/L.

The result was similar to a former study that reported a minimal percentage of desorbed P (<0.20%), even after a long-term desorption over 211 days (Ippolito et al., 2003). The reason may be that P in WTRs was mainly in the form of Fe/Al-bound P, while the loosely bound P only accounted for a small proportion (Babatunde and Zhao, 2009). Moreover, larger quantities of P desorbed at higher pH values because the Fe/Al-bound P is easily desorbed under alkaline conditions (Christophoridis and Fytianos, 2006). In addition, the P release from the P-saturated WTRs could be divided into three stages: an initial rapid P desorption stage, followed by a P re-adsorption stage, and a P desorption balance stage (Wang et al., 2012c). Overall, the limited desorption suggested a low risk of P release from saturated WTRs, favoring the development of real applications for WTRs, such as the restoration of lake sediment and soil amendments.

2.3. Relationship analysis

2.3.1. Relationship between the characteristics of the WTRs and P adsorption capacity

To identify which of the inherent characteristics of WTRs were most related to the P adsorption capacity, principal component analysis was conducted on the characteristics of the five WTRs. These factors can be classified into three groups (Table 2): (1) PC1 is related to pH, Fe, surface area, C and OM (high loadings for pH, Fe, Fe_{ox1}, surface area, C and OM), accounting for 48.80% of the total variance; (2) PC2 is related to Al (high loadings for Al and Al_{ox1}), accounting for 9.92% of the total variance, and (3) PC3 is related to other properties (high loadings for Ca, Ca_M, P, P_{ox1}, P_{ox2}, Fe_{ox2} and Al_{ox2}), accounting for 35.28% of the total variance. Overall, PC1, PC2 and PC3 accounted for 94.00% of the total variance.

The principal components and solution pH were used as independent variables during the multiple regression analysis. The P adsorption capacities (Q_1, Q_2 and Q_{max}) of the five WTRs were used as dependent variables, respectively. The results show that the PC1, PC2 and solution pH were significant in multiple regressions ($p < 0.01$) (Table 3). In the multiple regression of Q_1, PC2 entered the multiple regression first, accounting for 58.0% of the variation in the P adsorption ($p < 0.005$). Afterward, PC1 entered the multiple regression, accounting for

Table 2 – Principal component analysis of the physicochemical properties and contents of the five WTRs.

	Eigenvalue	Proportion of variance (%)	Cumulative proportion of variance (%)
Principle component			
PC1	7.321	48.80	48.80
PC2	1.488	9.92	58.72
PC3	5.293	35.28	94.00
Rotated factor pattern			
	PC1	PC2	PC3
Variable			
pH	**−0.746**	−0.312	−0.469
Surface area	**0.773**	0.136	0.570
Fe	**0.978**	−0.092	−0.038
Fe_{ox1}	**0.999**	0.018	−0.028
Fe_{ox2}	0.364	−0.184	**0.911**
Al	−0.010	**0.991**	0.049
Al_{ox1}	0.406	**0.877**	0.249
Al_{ox2}	−0.064	0.424	**0.903**
Ca	−0.493	−0.396	**−0.513**
Ca_M	−0.523	−0.542	**−0.656**
P	−0.473	0.4	**0.711**
P_{ox1}	−0.604	0.396	**0.689**
P_{ox2}	−0.165	0.501	**0.850**
C	**0.933**	−0.151	0.108
OM	**0.932**	0.026	0.147

Bold values: the highest loadings of principal components on the physicochemical properties and contents of WTRs.

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Table 3 – Results of multiple regression describing the relationship between the principal factors and the P adsorption.

	PC2	PC1	pH	PC3	R_1^2
Multiple regression					
Q_1	58.0%	21.4%	12.8%	Reject	0.924
Q_2	46.1%	31.1%	21.4%	Reject	0.986
Q_{max}	36.5%	28.5%	29.7%	Reject	0.947
	Significance	Equation	R_2^2		
Regression equation					
Q_1	<0.001	$Q_1 = 5.185 + 0.396PC1 + 0.669PC2 - 0.16\ pH$	0.938		
Q_2	<0.001	$Q_2 = 9.822 + 1.11PC1 + 1.419PC2 - 0.472\ pH$	0.971		
Q_{max}	<0.001	$Q_{max} = 9.892 + 1.085PC1 + 1.292PC2 - 0.561\ pH$	0.958		

Q_1 and Q_2: the P adsorption capacity of WTRs under an initial P concentration of 50 and 100 mg/L, respectively; Q_{max}: the maximum P adsorption capacity; R_1^2: R-squared of multiple regression; R_2^2: R-squared of predicted model.

another 21.4% ($p < 0.005$); the solution pH accounted for an additional 12.8% of the variation in P adsorption ($p < 0.001$). However, the PC3 related to other properties was rejected. Therefore, 92.2% of the variation in the P adsorption was explained using PC1, PC2 and solution pH. A similar pattern was also found during the multiple regression analysis of Q_2 and Q_{max}; however, the R_1^2 values of Q_2 and Q_{max} were slightly higher than that of Q_1. In addition, the equations yielded from multiple regressions. The test results revealed good correlations between the predicted and actual values. Therefore, the inherent characteristics of the WTRs had a remarkable influence on their P adsorption capacities.

A previous study indicated that there was a significant linear relationship between Q_{max} and Al_{ox1} (extracted at a solid to liquid ratio of 1:100) for the Al-based WTRs ($R^2 = 0.916$, $p < 0.001$), while adding Fe_{ox1} had no effect on the significance level (Dayton and Basta, 2005). However, that conclusion was not appropriate for the Fe-based WTRs. This study showed that PC1 and PC2 contributed a similar proportion of the various P adsorption capacities; in addition to Al, other inherent characteristics of WTRs (Fe, surface area, pH and OM) also played important roles. The dominant role of Fe and Al was attributed to the ligand exchange. The positive loading of the surface area indicated that a large surface area increased the P adsorption capacity by providing enough area for P attachment (Makris et al., 2004). However, since the pH variation of the five WTRs had a small range (7.23–7.90), the effect of pH on P adsorption needs further study in detail. In addition, the C in the WTRs mainly existed in organic form, which could inhibit the crystallization of Fe and Al and be beneficial to maintaining the high P adsorption capability (Dodor and Oya, 2000), thus the P adsorption was positively affected by the contents of C and OM. Consequently, the factors related to Fe and Al as well as the roles of the surface area and OM should be considered regarding the P adsorption capacity of the WTRs.

The long-term transformation of P in WTRs was related to the fraction of Ca, while the formation of soluble calcium phosphate was time dependent. A previous study reported that calcium phosphate began to form and transform in the WTRs after 28 days of a P adsorption experiment (Ippolito et al., 2003). However, the equilibrium time for this study was 2 days, making the contribution of Ca to the P adsorption capacity minor. In addition, Fe_{ox2} and Al_{ox2} had a weak relationship with the P adsorption capacity because Fe_{ox2} and Al_{ox2} represented the most reactive fraction, while the total amorphous Fe and Al had binding capability for P. Furthermore, crystalline Fe and Al could become amorphous under acidic conditions (Wang et al., 2011b). Therefore, the 5 mmol/L oxalate-extractable Fe and Al were unsuitable as an index to assess the P adsorption of the WTRs.

2.3.2. Relationship between the characteristics of the WTRs and P desorption capacity

While using the principal components (PC1, PC2 and PC3) and the solution pH as independent variables during multiple regression analysis, the quantities of P desorbed at P_0 of 50 and 100 mg/L were used as dependent variables, respectively. The R^2 and the significance were low ($R^2 = 0.43$, $p = 0.04$) below P_0 of 50 mg/L, suggesting that multiple regression analysis combined with principal component analysis was not suitable for P desorption. A linear fit was shown for P_0 of 50 and 100 mg/L; the P desorption was strongly negatively correlated with the ($Fe_{ox1} + Al_{ox1}$) of WTRs ($p < 0.05$) (Fig. 3). The R^2 at the three pH levels all exceeded 0.90. Moreover, the slope of the fitted curve decreased when the pH increased from 5 to 9. In addition, the correlations for the P desorption and PSI were weak at the three pH levels. A low correlation ($R^2 < 0.16$) was also apparent for P desorption *versus* ($Fe_{ox2} + Al_{ox2}$). Overall, the ($Fe_{ox1} + Al_{ox1}$) was a key factor that influenced the P desorption capacity of the WTRs.

The release of P from the saturated WTRs can be assessed using their ($Fe_{ox1} + Al_{ox1}$). The negative relationship occurred because the P desorption from the WTRs was prevented by the formation of stable P–Fe/Al complexes. Additional P–Fe/Al complexes were formed under acidic conditions; thus, the slope increased with a decrease in pH (Yang et al., 2006). Moreover, PSI was used as an index to assess the P leaching risk of soil (Kleinman and Sharpley, 2002). However, the PSI of the WTRs was far below that of common soil and was not a reliable index of P desorption. A former study found no significant correlations between the ($Fe_{ox1} + Al_{ox1}$) of seven WTRs and their long-term (80 days) P adsorption capacities (Makris et al., 2005). Therefore, ($Fe_{ox1} + Al_{ox1}$) could serve as an index for the P desorption capacity of WTRs, though not the P adsorption capacity.

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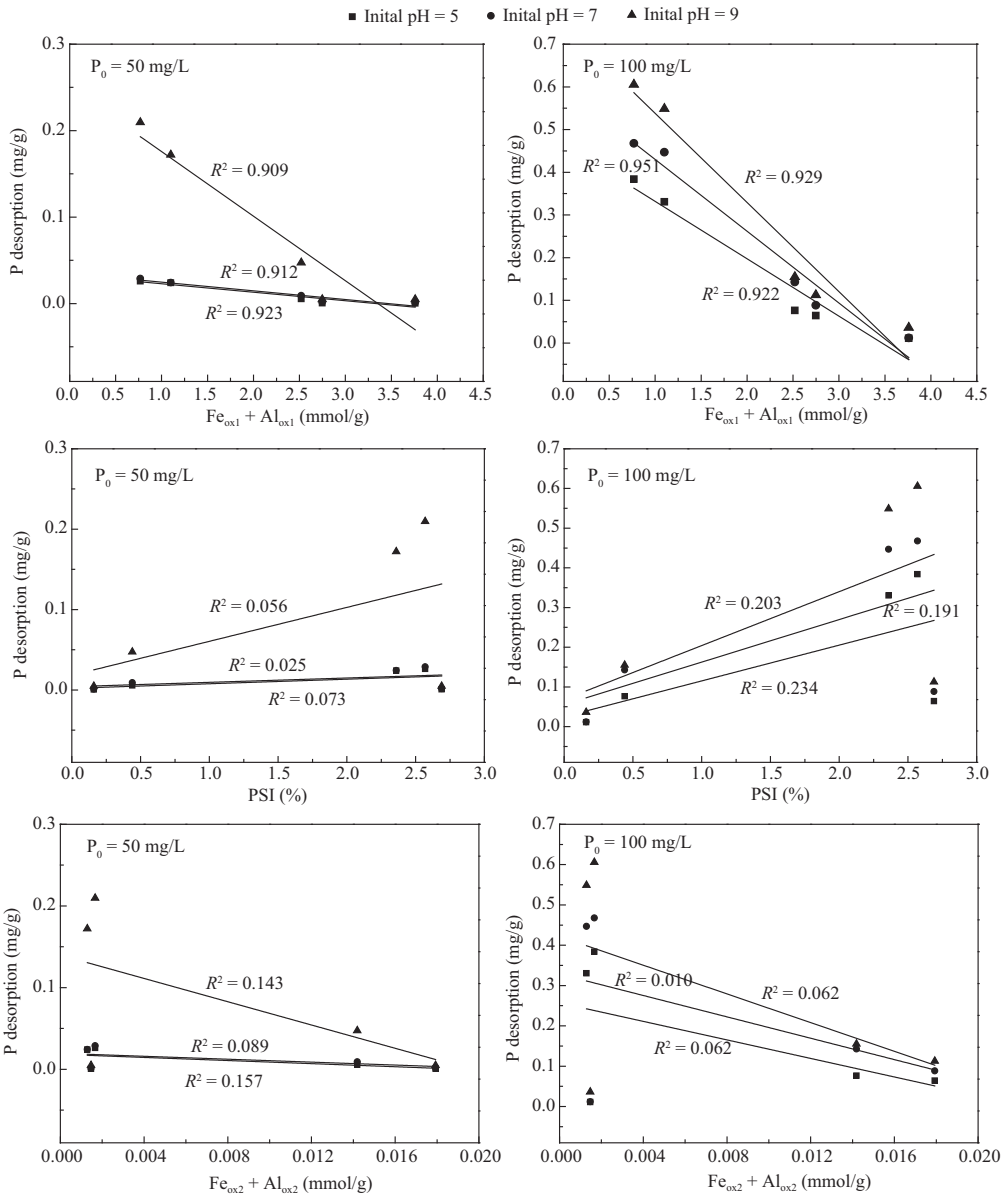

Fig. 3 – Relationship between the P desorption and the inherent properties of the five WTRs at different pH levels.

2.4. *Implications*

The WTRs have been used successfully as substrates for constructed wetlands and lake sediment to control P. A low dose and a high efficiency are preferable and essential for practical applications. However, the varying characteristics of WTRs induce a wide range of P adsorption capacities. Therefore, assessing the P adsorption of WTRs with different physicochemical properties is essential when choosing an application. The P saturation potential has been generally assessed by PSI. When PSI increased, more P was released or leached. However, the P adsorption capacities of 28 soils had a non-significant relationship with their PSIs ($p > 0.05$) (Zhang et al., 2005). A similar result was also found in this study (Appendix A Fig. S2). Therefore, PSI was not suitable as an index when assessing the P adsorption capacity. The P adsorption of Al-based WTRs was positively correlated with the Al and Al_{ox1} (Dayton and Basta, 2005). However, for the Fe-based WTRs and Ca-based WTRs, not only Al and Al_{ox1} but other inherent properties should be considered to improve accuracy. This study shows that the P adsorption capabilities of the five WTRs were remarkably different. An integrated factor (related to Fe, Fe_{ox1}, surface area and OM) played a role similar to Al and Al_{ox1} with variation of Q_{max}. High Fe and Fe_{ox1}, as well as a large surface area, and a high OM yielded a high adsorption capacity for P. The adsorption also increased with decreasing solution pH. In contrast, the influence of the other inherent properties (Ca, Ca_M, P, P_{ox1}, P_{ox2}, Fe_{ox2} and Al_{ox2}) was minor. In addition, the quantity of P desorbed from the saturated WTRs showed a negative correlation with (Fe_{ox1} + Al_{ox1}). Based on the risk of P leaching after the WTRs were saturated, a high (Fe_{ox1} + Al_{ox1}) favored P retention.

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Consequently, the WTRs produced from water treatment plants that used a high dosage of Fe/Al coagulant were more appropriate for this application. However, Ca-based WTRs from water treatment plants that used Ca salts as the main coagulant and softening agents showed a low efficiency for P adsorption.

3. Conclusions

Understanding the influence of the inherent properties of the WTRs on their P adsorption capacities is necessary for their practical application. The physical and chemical properties of five WTRs were determined and classified into three factors using principle component analysis. The relationship between the P adsorption capacities of the WTRs and their inherent properties was analyzed using multiple regression analysis. The factor related to Al and Al_{ox1} contributed the largest proportion (36.5%) of variation for Q_{max}, followed by factors related to Fe, Fe_{ox1}, surface area and OM (28.5%). However, the contribution of the factors related to other inherent properties (Ca, Ca_M, P, P_{ox1}, P_{ox2}, Fe_{ox2} and Al_{ox2}) was minor. Furthermore, the P desorption capacities of the saturated WTRs were negatively correlated with (Fe_{ox1} + Al_{ox1}), suggesting that (Fe_{ox1} + Al_{ox1}) was a useful index when assessing the P leaching risk of WTRs. Therefore, the inherent properties of the WTRs including the Fe, Al, surface area and OM should be considered when evaluating the P adsorption capacity.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found in online version at http://dx.doi.org/10.1016/j.jes.2014.04.002.

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