



Relationships between phosphorus fractionations in sediments and phosphorus in overlying water in a constructed wetland: impact of macrophytes

Yonglan Tian, Huayong Zhang*, He Hao, Songbo Cui, Luyi Zhang, Lei Zhao, Xiang Xu

Research Center for Engineering Ecology and Nonlinear Science, North China Electric Power University, Beijing 102206, China, Tel. +86 10 61773936, Fax: +86 10 61773936; emails: rceens@ncepu.edu.cn (H. Zhang), woaimokegege@126.com (Y. Tian), haohe333@126.com (H. Hao), 657469071@qq.com (S. Cui), luyizhang@hotmail.com (L. Zhang), leizhao@ku.edu (L. Zhao), xuxiang229@163.com (X. Xu)

Received 12 January 2017; Accepted 30 June 2017

ABSTRACT

Studies on phosphorus (P) distributions comparing planted and unplanted systems often lead to controversial results regarding the importance and growth seasons of plants. In the present study, the distribution and mobility of phosphorus fractionations (PFs) in eutrophic water and sediments were investigated in the absence or presence of two macrophytes, that is, reed and cattail, in a constructed wetland in autumn and spring. The removal efficiencies of total P in water, soluble reactive phosphorus and total dissolved phosphorus were 51.85%–63.75%, 74.52%–95.96% and 72.23%–83.02%, respectively. PFs mainly presented in permanent forms in sediments. In the absence of macrophytes in autumn, P in the overlying water was related to the mobile iron-bound phosphorus (Fe-P) in the sediment. In the presence of macrophytes, P in the overlying water was related to stable occluded phosphorus (O-P) in sediments which reduced the possibility of P release. In summary, macrophytes contributed to the stability of P in sediments and limited the release of P from sediment to overlying water, especially in spring.

Keywords: Phosphorus distribution; Macrophytes; Overlying water; Phosphorus release; Constructed wetland

1. Introduction

Anthropogenic eutrophication is one of the major water pollution problems in industrialized countries as well as in the developing world. Phosphorus (P) has been regarded as the common limiting factor responsible for eutrophication [1–7]. Based on literature review, P is accumulated in sediments, which are considered as net sinks and sources of many organic and inorganic compounds that include P [6,8–11]. Therefore, P transformation, bioavailability and exchange between sediment and the overlying water have been extensively studied [12–14]. Sequential extraction procedures were developed to elucidate the chemical nature of P in sediments [2,13]. These fractionations are P-forms binding to metals and to organic matter, measured

by different sequential extraction schemes, so-called phosphorus fractionations (PFs) [15].

Macrophytes in aquatic ecosystems were believed to exert great influence on the release of P from sediments; however, there has been a long debate on whether this influence is positive or negative [16–18]. Many previous studies focused on the effects of algal and submerged macrophytes on removing P from water or distributions of PFs [17,19–21]. Few studies have investigated the influences of emergent macrophytes on P transformation. The emergent macrophytes reed (*Phragmites australis*) and cattail (*Typha latifolia*) have become two of the most frequently used and widely distributed species in the rapidly developed constructed wetlands [22–26]. Previous studies have indicated that reed or cattail performed very well in removing pollutants from wastewaters, including metals [22,24] and P [27]. Moreover, reed proved to be efficient in the prevention of contaminant release, including N and P [23]. The influences

* Corresponding author.

of reed and cattail on the phosphorus cycle were studied in degraded, inundated peat soils without considering the different PFs [26]. However, the accumulation and speciation of PFs in sediments may differ among vegetated and unvegetated sediments [27]. Studies comparing planted and unplanted systems often lead to controversial results regarding the importance of plants [28–31]. Moreover, plants use P for growth but release P back into the water column due to decomposition and nutrient leaching from plant litter in different growth seasons [32]. Therefore, as two of the most common wetland plants, the impacts of reed and cattail on the removal of P and the release of P from sediments are of significance for managing constructed wetlands. Nevertheless, there is lack of knowledge on the different impacts of those two plants, especially taking seasonal differences into account.

In our previous study, reed and cattail were proved to affect the PFs distributions by influencing the sediment compositions, including organic matter, alkaline phosphatase activity, active Al and active Fe [33]. Here, the contributions of P in sediments to P in the overlying water in the presence of reed and cattail were studied and compared with unplanted areas in a constructed wetland in autumn and spring. The objectives of this study were to (1) investigate the spatial distribution of P in overlying water of open water, reed and cattail community areas in different seasons and (2) analyze the influences of reed and cattail on the mobility of P in sediments and its contribution to the P concentrations in the overlying water. The aim of this research is trying to find the mechanism of the effect of macrophytes on the distribution of P and to provide references for species selection and management of constructed wetlands.

2. Materials and methods

2.1. Study area

This study was conducted in a newly constructed wetland in two seasons, that is, autumn (end of October 2012) and spring (end of May 2013). The wetland is in Tengzhou, Shandong province, China. The study area lies in the south of a warm temperate zone, in a semi-humid region. The warmest month is July with an average temperature of 26.9°C while the coldest month is January with an average temperature of -1.8°C. The average annual precipitation in the region is 773.1 mm. This wetland was constructed in 2009 from a sunken coal mine. The total study area was around 0.2 km² (Fig. 1). The inlet of the wetland came from the Cheng River with characteristics shown in Table 1. The study area was the same as described in [33].

2.2. Sampling strategy

In each season, seven transects were set following the flow direction for sampling the sediments (0–5 cm of the sediment surface) and overlying water (-30 cm under water surface) (Fig. 1). Three kinds of surface sediments, that is, sediments in open water areas (OWA, S1, S2... S7), sediments in reed community areas (RCA, R1, R2... R7) and sediments in cattail community areas (CCA, C1, C2... C7), were collected

in each transect. Three replicate samples were collected and mixed together at each site. In total, 252 samples were taken from both sediment and water from three areas (cattail, reed and open water) during two seasons (spring and autumn), over seven transects that represented various flow conditions. The detailed descriptions for each sampling site can be found in [33].

Water samples (1 L for each sample) were collected with a columnar water harvesting device (WB-PM, China). Sediments samples (~50 g) were collected with a stainless steel grab bucket (Van Veen, the Netherlands). All the samples were kept in a dark cryogenic box (2°C–8°C) and brought back to the laboratory to be measured as soon as possible. In the laboratory, each bag with sediment was homogenized manually to mix all three samples for each quadrant. Any visible root or plant material was manually removed prior to homogenization.

2.3. Analytical methods

The total phosphorous in water (TP_w), soluble reactive phosphorus (SRP), total dissolved phosphorous (TDP) and total nitrogen (TN) were determined spectrophotometrically after digestion with potassium persulfate [34]. TP_w was measured directly while TDP and SRP were determined after filtration through a 0.45-μm cellulose acetate filtration membrane. Briefly, 25 mL of the sample was digested in an autoclave after adding 5% potassium persulfate. After cooling down, 1 mL of 10% ascorbic acid and 2 mL of molybdate solution were added to each of the sample. The samples were measured at 700 nm using an UV-visible spectrophotometer (Shanghai Qinghua Co. Ltd., China) after 15 min. The particulate phosphorus (PP) was calculated by the difference between TP_w and TDP, that is, PP = TP - TDP.

The air-dried sediments for measuring the PFs were ground into powder and passed through a 100-mesh sieve. The classified extraction of PFs in this study was based on Pierzynski's method [35] combined with Bao method [36]. The process of extraction is shown in Fig. 2. The PFs included exchangeable phosphorus (Ex-P), aluminum-bound phosphorus (Al-P), iron-bound phosphorus (Fe-P), occluded phosphorus (O-P), calcium-bound phosphorus (Ca-P) and organic phosphorus (Org-P). The total phosphorus in sediments (TPs) was obtained from the sum of the six PFs. Moreover, permanent phosphorus (Per-P), semi-permanent phosphorus (Semi-Per-P) and reactive phosphorus (Rea-P) were determined as follow: Per-P = Ca-P + O-P; Semi-Per-P = Al-P + Fe-P + Org-P and Rea-P = Ex-P.

2.4. Data analysis

Pearson correlation analysis was used to analyze the correlations between PFs in sediment and overlying water at 5% and 1% levels of significance, as indicated by * ($p < 0.05$) and ** ($p < 0.01$), respectively. One-way analysis of variance (ANOVA) was used to analyze the influences of season and presence of reed or cattail on P removal from overlying water. The Pearson correlation analysis and one-way ANOVA were conducted using the Statistical Package for the Social Science software (SPSS 17.0, Chicago, IL, USA). The influence of seasons and presence of reed or cattail on the ratio of nitrogen

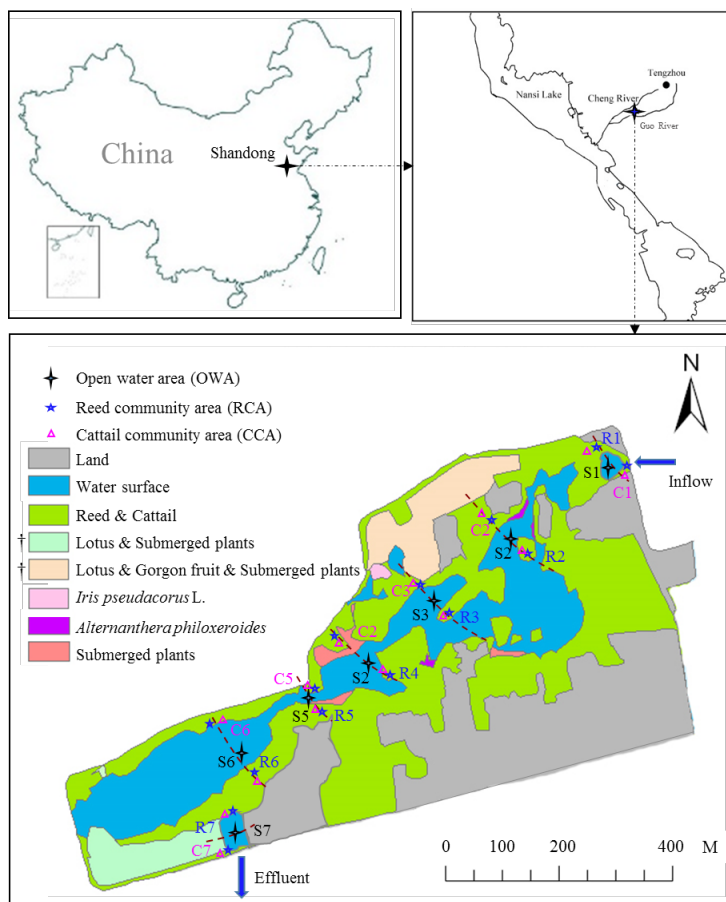


Fig. 1. Study area, vegetation distributions and sampling sites. The study site is in Tengzhou City, Shandong province, China. Different colors represent different plants, and the location and area of each plant was recorded by GPS. S1–S7 (black four-pointed stars), R1–R7 (blue five-pointed stars) and C1–C7 (pink triangles) represent the sites for sampling water and sediments in open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs), respectively.

†Lotus in the wetland only occurred in May and was harvested before October.

Table 1

Climate conditions, inflow characteristics and removal efficiency of the studied wetland, as well as the status of the reed and cattail at the end of October 2012 and May 2013

Parameters	End of October 2012	End of May 2013
Temperature (°C)	8–18	17–26
Daylight (h)	10.5	14.5
Vegetation density (plant/m ²)	Reed 76 ± 26 Cattail 9 ± 4	Reed 160 ± 79 Cattail 58 ± 24
Overground biomass (dry weight g/plant)	Reed 31.14 ± 12.84 Cattail 38.00 ± 8.92	Reed 26.90 ± 2.21 Cattail 37.38 ± 3.9
Inflow (average, mg/L)		
COD	21.00	15.58
TP _w	0.57	1.00
TDP	0.38	0.95
SRP	0.34	0.86
TN	16.56	11.91
NO ₃ ⁻	11.21	6.26
DO	14.06	26.62
pH	8.36	7.99
T (°C)	15.4	22.7

to phosphorus was determined by two-way ANOVA in the statistics program R (version 3.3.1). Post hoc comparisons were applied using the Tukey HSD test at the 0.05 level of significance.

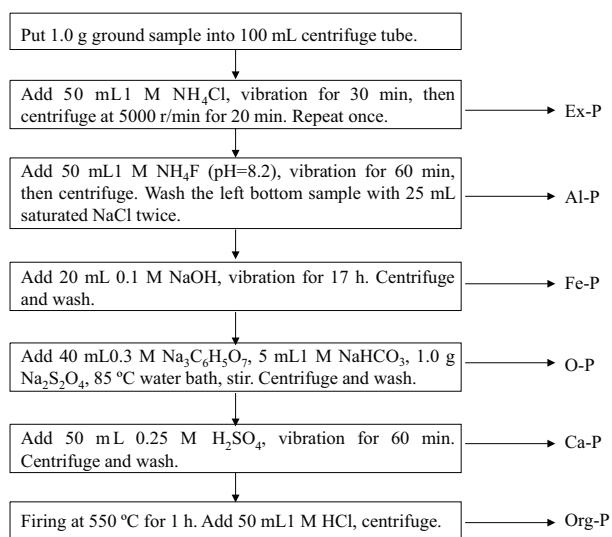


Fig. 2. Process of extracting phosphorus fractionations (PFs) according to published methods.

3. Results and discussion

3.1. Spatial distribution and removal of phosphorus in overlying water

Fig. 3 shows the distribution of P concentrations with water flow in autumn and spring. The concentrations of TP_w , SRP and TDP declined differently with flow in autumn and spring (Fig. 3(A–C)). In autumn, TP_w , SRP and TDP did not reduce in the first three transects. They began to decline from the middle of the flow (the fourth transect) and then gradually decreased in further downstream. In contrast, in spring, the declines in TP_w , SRP and TDP were remarkable in the first two transects and then slowed in the subsequent transects.

The removal efficiencies are summarized in Table 2. Like most wetlands, Qixinghu wetland can provide significant phosphorous removal from wastewater through a combination of physical, chemical and biological processes [37]. On average, the removal efficiency of TP_w was lower than that of SRP and TDP regardless of the season or the impact of macrophytes. Considering seasonal impacts, the average removal efficiencies of TP_w ($p < 0.05$ in OWAs) and TDP (not statistically significant) in autumn were lower than those in spring for the overlying water of OWAs, RCAs and CCAs. The removal efficiency of SRP in the overlying water of OWAs and RCAs was lower in spring than in autumn ($p < 0.05$); in overlying water of CCAs, it was higher in spring than in autumn ($p < 0.05$).

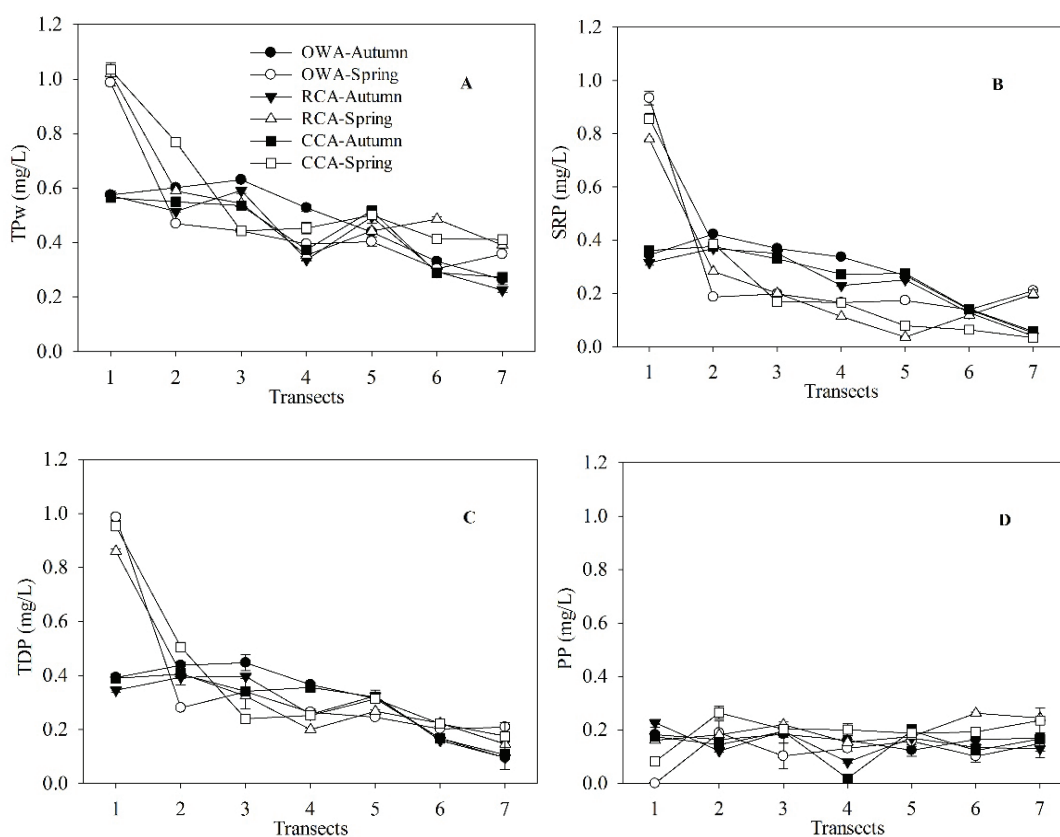


Fig. 3. Variation of total phosphorous in water (TP_w) (A), soluble reactive phosphorus (SRP) (B), total dissolved phosphorous (TDP) (C) and PP (D) with flow in the overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) in autumn (close symbols) and spring (open symbols).

Table 2
Removal efficiency of TP_w , SRP and TDP in the studied wetland

Type of overlying water	Season	Removal efficiency (%)			
		TP_w	SRP	TDP	PP
OWAs	Autumn	53.86 ± 4.40 AB	84.75 ± 0.41 AB	75.68 ± 0.74 A	3.79 ± 24.10 A
	Spring	63.75 ± 0.44 a*	77.41 ± 0.63 a*	78.89 ± 0.51 a	ND
RCAs	Autumn	60.65 ± 1.73 B	87.03 ± 0.42 B	72.23 ± 0.50 A	43.12 ± 3.12 A
	Spring	61.75 ± 0.58 a	74.52 ± 1.98 a*	83.02 ± 0.90 a	-51.67 ± 1.41 a
CCAs	Autumn	51.85 ± 3.63 A	83.84 ± 0.50 A	72.80 ± 13.77 A	2.16 ± 44.97 A
	Spring	60.17 ± 1.44 a	95.96 ± 0.09 b*	81.45 ± 5.56 a	-187.97 ± 18.46 a

Mean values ± Standard Error (SE). ND, not detected, TP_w , total phosphorous in water; SRP, soluble reactive phosphorus; TDP, total dissolved phosphorous; PP, particulate phosphorus. Significant differences were analyzed between different types of overlying water for TP, SRP and TDP, respectively. * and different letters, that is, A/B or a/b, represent significant differences at least significant difference < 5% (SPSS 17.0).

Considering the influence of macrophytes on P removal, it was found that the removal efficiencies of TP_w and SRP in overlying water of OWAs were not significantly different compared with RCAs or CCAs, in either autumn or spring. Thus, reed and cattail did not have a notable effect on P removal from the water in this study. In contrast, reed and cattail were reported to be of benefit for the removal of P from water, which lowered the risk of P losses to adjacent water courses during the growing season [26]. However, here, in autumn, the removal efficiency of TP_w and SRP in overlying water of RCAs was significantly higher than in overlying water of CCAs ($p < 0.05$). In spring, the removal efficiency of TP_w from overlying water of RCAs was close to that of CCAs, while the removal efficiency of SRP in the overlying water of RCAs was significantly lower than that of CCAs ($p < 0.05$).

Although both dissolved and particulate organic P may be biologically broken down to inorganic phosphorous (mineralization) and subsequently removed through biological and chemical processes [37], it seemed that PP was removed from the overlying water in autumn, but increased in macrophyte-covered areas in spring (Table 2). Most of the analyzed PP concentrations were from 0.1 to 0.2 mg/L (Fig. 3(D)). Therefore, dissolved P in the overlying water was more efficiently removed than PP and was better removed in spring than in autumn. This result agreed with previous research in a large-scale constructed treatment wetland receiving eutrophic lake water in the United States [38].

The ratio of nitrogen to phosphorus was largely affected by the season (two-way ANOVA: $F_{1,36} = 30.598$, $p < 0.001$), but not significantly influenced by type of sediment ($F_{2,36} = 0.429$, $p = 0.655$) or the interaction terms of seasons and the presence of reed or cattail ($F_{2,36} = 0.403$, $p = 0.671$). Redfield [39] reported that the similarity between the average nitrogen-to-phosphorus ratio in plankton was N:P = 16:1 (by atoms). This ratio ranged from 8.2:1 to 45.0:1 depending on the ecological conditions [40]. In the present study, the N:P ratio in water (by atoms) ranged from 78.6:1 to 81.8:1 in autumn and from 42.9:1 to 54.7:1 in spring (Fig. 4). It was close to the upper threshold of the reported range in spring and was higher than the reported range in autumn. Therefore, the present studied wetland system was likely P limited according to stoichiometry data. It is generally agreed upon that algal blooms occur in water bodies when the concentration of TP_w reaches 0.02 mg/L [41]. In the present study, the TP_w concentrations

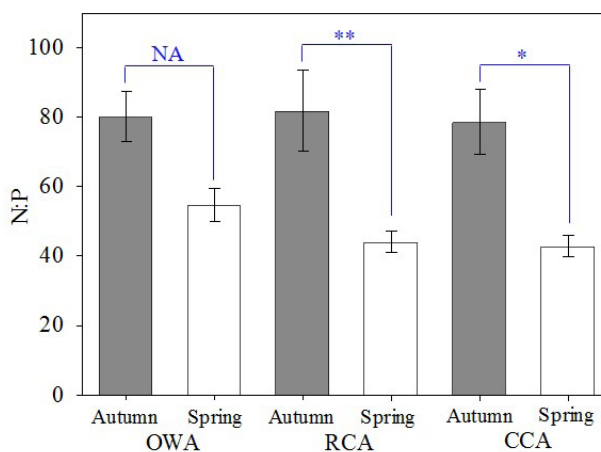


Fig. 4. Variation of N:P (by atoms) in the overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) in both autumn (gray bars) and spring (white bars). Error bars show the SE ($n = 7$). The analysis was carried out by two-way ANOVA. Post-hoc comparisons were applied using the Tukey HSD test at the 0.05 level of significance. NA, not significant; *, significant level at 0.05; **, significant level at 0.01.

were found to exceed this critical concentration even after decontamination of the wetland. However, in spite of high TP_w concentrations, no algal bloom was observed in the Qixinghu wetland.

3.2. Influence of macrophytes on P mobility

The mobility of P in sediments was determined by analyzing the variation of three categories of phosphorus in the present study, that is, Per-P, Semi-Per-P and Rea-P (Fig. 5). In OWAs, the percentage of Per-P in the TPs increased gradually with the water flow in both autumn and spring. Per-P represented almost half of the TPs in autumn and more than half in spring. Semi-Per-P also accounted for a large percentage of the TPs in autumn. In RCA, PFs distributions varied a lot in different transects. At transect R1, in autumn, Per-P was only about 10% of the TPs, while Semi-Per-P was about 90% of the TPs. Per-P increased to half or more of the TPs in the following transects. In both autumn and spring, the

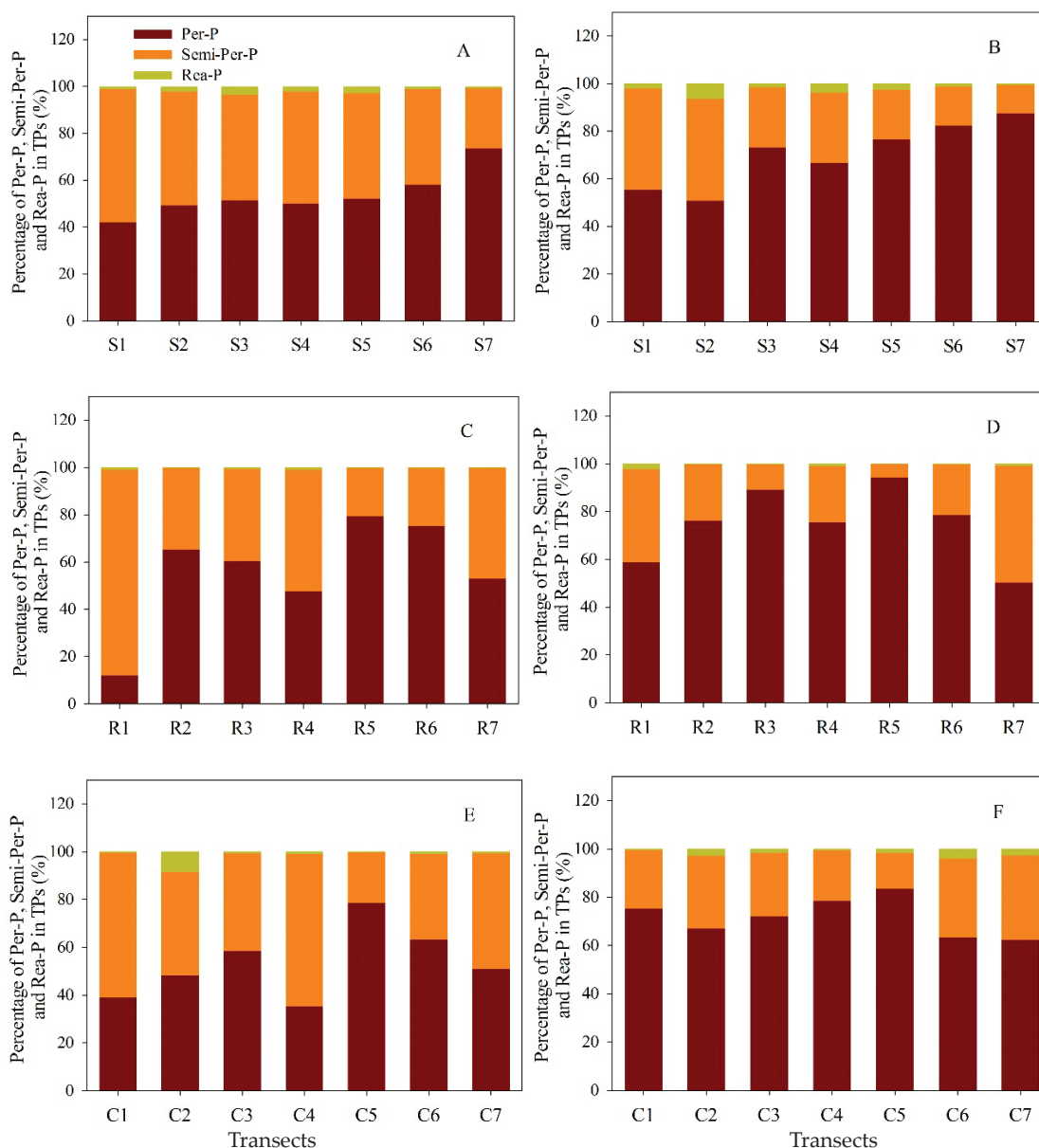


Fig. 5. Percentage of Per-P (dark red bars), Semi-Per-P (orange bars) and Rea-P (yellowish-green bars) in TP in three types of sediments, that is, open water area sediments (A and B), reed community area sediments (C and D) and cattail community area sediments (E and F) in both autumn (A, C and E) and spring (B, D and F) following the water flow direction. S1–S7, sample transects in open water areas (OWAs); R1–R7, sample transects in reed community areas (RCAs); C1–C7, sample transects in cattail community areas (CCAs).

highest Per-P in RCAs was found at transect R5, while in OWAs it was found at transect S7. Thus, more Per-P was locked in the middle of the flow in RCAs and it accumulated at the end of the flow in OWAs. Considering CCAs, the variations of Per-P and Semi-Per-P in autumn were similar to the results for RCAs. However, in spring, the percentages of Per-P in TPs were more stable in CCAs than that in OWAs and RCAs.

Soluble exchangeable phosphorus was the most immediately available phosphorus within sediments [42], and Ex-P is the best parameter for the assessment of the bio-availability of phosphorus [43]. The most available form of

phosphorus in this study was the Rea-P (i.e., Ex-P) fraction. The percentage of Rea-P in TPs ranged from 0.06% to 8.45% (Fig. 5), which was much lower than Per-P and Semi-Per-P. Relatively high Ex-P concentrations were observed in OWAs, meaning that loosely exchangeable phosphorus was readily available to algae, consequently promoting blooms in these areas. In other words, in the macrophyte-covered areas, the presence of macrophytes resulted in a low Ex-P content and reduced the risk of P release, especially in spring. These results supported the seasonal management strategy of constructed wetlands for the purpose of high efficiency [38].

3.3. Contributions of PFs in surface sediments to P in overlying water

3.3.1. Relationships between PFs and P in overlying water in different seasons

In the macrophyte-free area, TP_w , TDP and SRP were related to Fe-P in autumn (Table 3). This result was in line with previous discussion about P release. As Fe-P is rather active [4], its higher content indicated a higher risk of P release. Therefore, the sediments in autumn seemed rather mobile and have more potential for release than sediments in spring.

In macrophyte-covered areas (RCAs and CCAs), O-P was significantly related to TP_w , SRP and TDP in spring but not in autumn (Table 3). However, there were some differences between relationships in RCA and CCAs, indicating the different effects of reed and cattail. The relative coefficients between O-P and PFs in RCAs were higher than in CCAs. This might be due to the different structures of these two plant species. Reeds benefit from an extensive root and rhizome system and have a high plasticity with respect to variable nutrient conditions [44–46]. The relatively low P demand of reed compared with other fast-growing species allows this species to have higher production rates at lower P stocks (i.e., lower P concentrations) [26]. SRP was positively related to Ex-P in RCAs while PP was negatively related to Al-P and Fe-P in CCAs (details in Table 4). It is suggested that higher exchangeable P in the sediment might cause the increase of SRP in the overlying water of RCAs, which does not benefit from the P retention. However, further experimentation is required to explain the relationship between PP and Al-P/Fe-P.

Comparing the macrophyte-free area and macrophyte-covered area, it was found that O-P in sediments was related to P in overlying water in the macrophyte-covered area in spring, but not in autumn, or in the macrophyte-free area. Meanwhile, only Fe-P was related to P in overlying water in the macrophyte-free area in autumn. The results indicate that the presence of reed and cattail resulted in the variation of P forms transformation between water and sediments. Furthermore, this impact was greater in spring when the plants entered the growing season.

3.3.2. Relationships between PFs and P in overlying water regardless of season

Regardless of the season, O-P was found to be significantly related to P in overlying water in both macrophyte-free and macrophyte-covered areas (Table 5). The forms of P related to O-P were TP_w and TDP, TP_w and SRP and TDP; and SRP and TDP in overlying water of OWAs, RCAs and CCAs, respectively. However, the relationships between O-P and P in overlying water were negative in macrophyte-free areas, and positive in macrophyte-covered areas (Table 6).

Aside from O-P, the only relationship between Ex-P and SRP was found in the reed-covered areas (Table 5). As previously discussed, Ex-P is exchangeable and easily released into overlying water once environmental conditions permit. SRP has often been used to study the influence of environmental factors on the interactions at the sediment–water interface and the further impact on P release [17,18]. In lakes, the relative contribution of roots to phosphorus uptake (P) depends on the relative SRP concentrations of the sediment and the overlying water [47]. It can be estimated from the concentrations of dissolved reactive phosphorus in the sediment pore water (s) and overlying water (w) by $P = 99.81/1 + 2.66 (s/w)^{-0.83}$ [47]. In the present study, from the second to sixth transect, the SRP concentrations in spring were lower than those in autumn. This was probably due to the uptake of soluble phosphate by sorption into plant biofilms. At the onset, there may be some sorption of negatively charged phosphate particles to the bottom soil liner particles. Then, the insoluble organic phosphate was transformed into a soluble inorganic form by microorganisms. Once the phosphate was available to the plants, its uptake occurred during the growing season. But during the senescence stage of the plants and in the winter, the uptake decreased until plant death and decomposition followed [37].

Through further analysis, it was found that some relationships disappeared if seasonal influence was not considered. For example, Fe-P in sediments was no longer related to P in the overlying water of OWAs, and Al-P in sediments was no longer related to PP in the overlying water of CCA. However, some new relationships appeared, including the negative relationship between O-P in sediments and P in the overlying water

Table 3

Pearson correlations among PFs in sediments and P in overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) regard to season

	OWAs		RCAs		CCAs	
	Autumn	Spring	Autumn	Spring	Autumn	Spring
TP_w	Fe-P*			O-P**		O-P**
SRP	Fe-P*			Ex-P*, O-P**		O-P**
TDP	Fe-P*			O-P**		O-P**
PP					Al-P**, Fe-P**	

*, $p < 0.05$; **, $p < 0.01$. $n = 7$. TP_w , Total phosphorous in water; SRP, soluble reactive phosphorous; TDP, total dissolved phosphorous; PP, particulate phosphorous; Fe-P, iron-bound phosphorous; Ex-P, exchangeable phosphorous; O-P, occluded phosphorous; Al-P, aluminum-bound phosphorous.

Table 4

Correlations among all measured PFs in sediments and P in overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) with regard to season

		Ex-P	Al-P	Fe-P	O-P	Ca-P	Org-P	TPs
OWA								
Autumn	TP _w	0.648	0.773	0.839*	-0.582	0.181	0.617	0.500
	SRP	0.578	0.674	0.841*	-0.508	0.124	0.630	0.461
	TDP	0.637	0.721	0.808*	-0.501	0.134	0.594	0.463
	PP	0.233	0.256	0.404	-0.689	0.368	0.302	0.369
Spring	TP _w	-0.053	0.061	0.037	-0.636	-0.660	-0.054	-0.340
	SRP	-0.222	-0.106	-0.124	-0.529	-0.716	-0.163	-0.478
	TDP	-0.177	-0.055	-0.086	-0.599	-0.697	-0.046	-0.432
	PP	0.488	0.360	0.404	0.427	0.713	0.019	0.625
RCA								
Autumn	TP _w	0.236	0.127	0.091	-0.621	-0.382	0.270	-0.034
	SRP	0.267	0.194	0.267	-0.679	-0.248	0.180	0.046
	TDP	0.191	0.120	0.178	-0.706	-0.259	0.126	-0.036
	PP	0.240	0.086	-0.183	-0.160	-0.514	0.493	-0.019
Spring	TP _w	0.683	-0.569	-0.278	0.909**	-0.265	0.541	-0.200
	SRP	0.809*	-0.414	-0.226	0.954**	-0.462	0.728	-0.368
	TDP	0.720	-0.515	-0.220	0.904**	-0.202	0.510	-0.131
	PP	-0.477	-0.082	-0.217	-0.327	-0.248	-0.038	-0.314
CCA								
Autumn	TP _w	0.307	-0.237	-0.159	0.217	0.046	0.442	-0.085
	SRP	0.429	0.065	0.136	0.237	0.041	0.356	0.164
	TDP	0.421	0.222	0.296	0.291	0.074	0.200	0.301
	PP	-0.152	-0.900**	-0.876**	-0.095	-0.044	0.537	-0.736
Spring	TP _w	-0.554	-0.218	-0.003	0.776*	-0.108	-0.152	-0.158
	SRP	-0.660	-0.242	0.053	0.810*	-0.020	0.063	-0.049
	TDP	-0.635	-0.330	-0.083	0.794*	-0.061	-0.087	-0.125
	PP	0.748	0.681	0.385	-0.591	-0.157	-0.213	-0.061

*, $p < 0.05$; **, $p < 0.01$. $n = 7$. Ex-P, exchangeable phosphorus; Al-P, aluminum-bound phosphorus; Fe-P, iron-bound phosphorus; O-P, occluded phosphorus; Ca-P, calcium-bound phosphorus; Org-P, organic phosphorus; TPs, total phosphorus in sediments; TP_w, total phosphorus in water; SRP, soluble reactive phosphorus; TDP, total dissolved phosphorus; PP, particulate phosphorus.

Table 5

Pearson correlations among PFs in sediments and P in overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) regardless of season

	OWAs	RCAs	CCAs
TP _w	O-P*	O-P**	
SRP		Ex-P**, O-P**	O-P*
TDP	O-P*	O-P**	O-P*
PP			

*, $p < 0.05$; **, $p < 0.01$. $n = 14$. TP_w, total phosphorus in water; SRP, soluble reactive phosphorus; TDP, total dissolved phosphorus; PP, particulate phosphorus; Ex-P, exchangeable phosphorus; O-P, occluded phosphorus.

of OWA. The stable O-P in sediments in macrophyte-free areas originated from P in the overlying water; thus, the removal of P in macrophyte-free areas was carried out by deposition effects. In the macrophyte-covered areas, the removal of P was mainly a function of macrophytes and possibly microorganisms. In this case, the removal of P did not cause an increase of O-P, and might reduce O-P. The detailed processes of P exchange between sediment and water need further research.

It is well known that the presence of macrophytes causes variations of sediment environment and results in different sediment ecosystem structures, such as changes in microorganisms, benthic algae and protists, which play an important role in the interactions at the sediment–water interface [5]. In detail, the presence of macrophytes in aquatic systems can impact phosphorus release from the sediment into the lake

Table 6

Correlations among all measured PFs in sediments and P in overlying water of open water areas (OWAs), reed community areas (RCAs) and cattail community areas (CCAs) regardless of season

		Ex-P	Al-P	Fe-P	O-P	Ca-P	Org-P	TPs
OWA	TP _w	0.218	0.271	0.261	-0.591*	-0.063	0.154	0.081
	SRP	0.168	0.189	0.210	-0.489	-0.119	-0.043	0.001
	TDP	0.105	0.149	0.154	-0.536*	-0.209	-0.006	-0.077
	PP	0.342	0.266	0.264	0.435	0.335	0.200	0.358
RCA	TP _w	0.626	-0.136	-0.152	0.746**	-0.003	0.367	0.081
	SRP	0.686**	-0.066	-0.078	0.794**	-0.323	0.463	-0.219
	TDP	0.659	-0.142	-0.105	0.789**	-0.180	0.311	0.056
	PP	-0.418	-0.381	-0.397	-0.183	-0.096	-0.259	-0.226
CCA	TP _w	0.026	-0.183	-0.170	0.521	0.208	0.088	0.122
	SRP	-0.102	-0.027	0.063	0.594*	-0.022	0.103	-0.008
	TDP	-0.024	-0.016	0.018	0.580*	0.151	0.040	0.142
	PP	0.334	0.017	0.196	-0.316	-0.256	-0.055	-0.188

*, $p < 0.05$; **, $p < 0.01$. $n = 14$. Ex-P, exchangeable phosphorus; Al-P, aluminum-bound phosphorus; Fe-P, iron-bound phosphorus; O-P, occluded phosphorus; Ca-P, calcium-bound phosphorus; Org-P, organic phosphorus; TPs, total phosphorus in sediments; TP_w, total phosphorus in water; SRP, solubility reactive phosphorus; TDP, total dissolved phosphorus; PP, particulate phosphorus.

water at low levels, by creating oxygen in the sediment, establishing an oxidized barrier to P diffusion, raising sediment Eh, lowering pH and filterable Fe and P percentages and finally enhancing sediment P retention [16–18,48]. Moreover, the macrophyte species producing specific biogenic P compounds, the biomass and the community structure can also lead to disparity of P distribution [49]. To reveal the mechanisms of reed and cattail in removing P from sediment and water, P content in the biomass of reed and cattail as well as the influence of microorganisms should be considered in future research.

4. Conclusions

In the present study, the influence of seasons and macrophytes on the removal of P from eutrophic water and the mobility PFs in surface sediments were investigated in a P-limited constructed wetland. It was found that TP_w, especially dissolved P was more efficiently removed in spring than in autumn. Reed and cattail performed differently in SRP removal in the overlying water. The removal efficiency of SRP in CCAs was significantly higher than that in RCAs and OWAs in spring. The presence of macrophytes reduced the risk of P release, as indicated by the low percentage of Rea-P in TPs, especially in spring. Among all the PFs in sediments, O-P in sediments was most related to P in the overlying water, regardless of season. However, considering the seasonal differences, Fe-P was related to P in the overlying water in the macrophyte-free areas in autumn, but not in the macrophyte-covered areas. Thus, the presence of macrophytes induced variation of sediment–water P transformation which further influenced the bioavailability of PFs and reduced the potential of P release into water.

Acknowledgments

This work was funded by the National Special Water Programs (No. 2009ZX07210–009, No. 2015ZX07203–011, No. 2015ZX07204–007, 2017ZX07101–003), and the China Scholarship

Council (No. 201406730018). The authors would like to thank Mr. Yuanwu Xiong and Ms. Xin Zhong for checking the data and Jennifer Swann and James Allen for polishing the language.

References

- [1] J. Wang, X. Jiang, B. Zheng, C. Chen, X. Kang, C. Zhang, Z. Song, K. Wang, W. Wang, S. Wang, Effect of algal bloom on phosphorus exchange at the sediment–water interface in Meiliang Bay of Taihu Lake, China, *Environ. Earth Sci.*, 75 (2016) 1–9.
- [2] V. Ruban, J.F. López-Sánchez, P. Pardo, G. Rauret, H. Muntau, P. Quevauviller, Harmonized protocol and certified reference material for the determination of extractable contents of phosphorus in freshwater sediments – a synthesis of recent works, *Fresenius J. Anal. Chem.*, 370 (2001) 224–228.
- [3] L. Wang, T. Liang, Distribution characteristics of phosphorus in the sediments and overlying water of Poyang Lake, *PLoS One*, 10 (2015) e0125859.
- [4] C. Chen, W. Deng, X. Xu, J. He, S. Wang, L. Jiao, Y. Zhang, Phosphorus adsorption and release characteristics of surface sediments in Dianchi Lake, China, *Environ. Earth Sci.*, 74 (2015) 3689–3700.
- [5] X. Jin, X. Jiang, Y. Yao, L. Li, F. Wu, Effects of organisms on the release of phosphorus at the interface between sediment and water, *Water Environ. Res.*, 79 (2007) 2253–2259.
- [6] Y. Wu, Y. Wen, J. Zhou, Y. Wu, Phosphorus release from lake sediments: effects of pH, temperature and dissolved oxygen, *KSCE J. Civil Eng.*, 18 (2014) 323–329.
- [7] F. Zan, S. Huo, B. Xi, Q. Li, H. Liao, J. Zhang, Phosphorus distribution in the sediments of a shallow eutrophic lake, Lake Chaohu, China, *Environ. Earth Sci.*, 62 (2011) 1643–1653.
- [8] M.W. Beutel, Inhibition of ammonia release from anoxic profundal sediments in lakes using hypolimnetic oxygenation, *Ecol. Eng.*, 28 (2006) 271–279.
- [9] M. Dittrich, O. Gabriel, C. Rutzen, R. Koschel, Lake restoration by hypolimnetic Ca(OH)₂ treatment: impact on phosphorus sedimentation and release from sediment, *Sci. Total Environ.*, 409 (2011) 1504–1515.
- [10] J. Lin, Y. Zhan, Z. Zhu, Evaluation of sediment capping with active barrier systems (ABS) using calcite/zeolite mixtures to simultaneously manage phosphorus and ammonium release, *Sci. Total Environ.*, 409 (2011) 638–646.

- [11] E. Rydin, Potentially mobile phosphorus in Lake Erken sediment, *Water Res.*, 34 (2000) 2037–2042.
- [12] X. Jin, S. Wang, Y. Pang, F.C. Wu, Phosphorus fractions and the effect of pH on the phosphorus release of the sediments from different trophic areas in Taihu Lake, China, *Environ. Pollut.*, 139 (2006) 288–295.
- [13] R. Zhang, F. Wu, C. Liu, P. Fu, W. Li, L. Wang, H. Liao, J. Guo, Characteristics of organic phosphorus fractions in different trophic sediments of lakes from the middle and lower reaches of Yangtze River region and Southwestern Plateau, China, *Environ. Pollut.*, 152 (2008) 366–372.
- [14] D. Kapiti, A. Bekteshi, Phosphorus bioavailability in sediments of a sludge-disposal shkodra lake, *J. Environ. Prot. Ecol.*, 15 (2014) 48–52.
- [15] A. Kaiserli, D. Voutsas, C. Samara, Phosphorus fractionation in lake sediments – Lakes Volvi and Koronia, N. Greece, *Chemosphere*, 46 (2002) 1147–1155.
- [16] W. Qu, R.J. Morrison, R.J. West, Inorganic nutrient and oxygen fluxes across the sediment–water interface in the inshore macrophyte areas of a shallow estuary (Lake Illawarra, Australia), *Hydrobiologia*, 492 (2003) 119–127.
- [17] S. Schneider, A. Melzer, Sediment and water nutrient characteristics in patches of submerged macrophytes in running waters, *Hydrobiologia*, 527 (2004) 195–207.
- [18] A. Topçu, S. Pulatsü, Phosphorus fractions and cycling in the sediment of a shallow eutrophic pond, *J. Agric. Sci.*, 20 (2014) 63–70.
- [19] D.K. Pelton, S.N. Levine, M. Braner, Measurements of phosphorus uptake by macrophytes and epiphytes from the LaPlatte River (VT) using ^{32}P in stream microcosms, *Freshwater Biol.*, 39 (1998) 285–299.
- [20] D. Jamet, C. Amblard, J. Devaux, Seasonal changes in alkaline phosphatase activity of bacteria and microalgae in Lake Pavin (Massif Central, France), *Hydrobiologia*, 347 (1997) 175–195.
- [21] M. Schulz, H.-P. Kozerski, T. Pluntke, K. Rinke, The influence of macrophytes on sedimentation and nutrient retention in the lower River Spree (Germany), *Water Res.*, 37 (2003) 569–578.
- [22] E. Grisey, X. Laffray, O. Contoz, E. Cavalli, J. Mudry, L. Aleya, The bioaccumulation performance of reeds and cattails in a constructed treatment wetland for removal of heavy metals in landfill leachate treatment (Etueffont, France), *Water Air Soil Pollut.*, 223 (2012) 1723–1741.
- [23] Y. Dong, Application of Integrated Constructed Wetlands for Contaminant Treatment and Diffusion, The University of Edinburgh, Edinburgh, 2013.
- [24] Z. Ben Salem, X. Laffray, A. Ashoour, H. Ayadi, L. Aleya, Metal accumulation and distribution in the organs of Reeds and Cattails in a constructed treatment wetland (Etueffont, France), *Ecol. Eng.*, 64 (2014) 1–17.
- [25] Y. Vergeles, N. Butenko, A. Ishchenko, F. Stolberg, M. Hogland, W. Hogland, Formation and properties of sediments in constructed wetlands for treatment of domestic wastewater, *Urban Water J.*, 13 (2016) 293–301.
- [26] D. Zak, J. Gelbrecht, S. Zerbe, T. Shatwell, M. Barth, A. Cabezas, P. Steffenhagen, How helophytes influence the phosphorus cycle in degraded inundated peat soils – implications for fen restoration, *Ecol. Eng.*, 66 (2014) 82–90.
- [27] G.A. Di Luca, M.A. Maine, M.M. Mufarregge, H.R. Hadad, C.A. Bonetto, Influence of *Typha domingensis* in the removal of high P concentrations from water, *Chemosphere*, 138 (2015) 405–411.
- [28] M.E. Baldizon, R. Dolmus, J. Quintana, Y. Navarro, M. Donze, Comparison of conventional and macrophyte-based systems for the treatment of domestic wastewater, *Water Sci. Technol.*, 45 (2002) 111–116.
- [29] C.S.C. Calheiros, A.O.S.S. Rangel, P.M.L. Castro, Constructed wetland systems vegetated with different plants applied to the treatment of tannery wastewater, *Water Res.*, 41 (2007) 1790–1798.
- [30] L. Marchand, M. Mench, D.L. Jacob, M.L. Otte, Metal and metalloid removal in constructed wetlands, with emphasis on the importance of plants and standardized measurements: a review, *Environ. Pollut.*, 158 (2010) 3447–3461.
- [31] R. Menon, M.M. Holland, Phosphorus retention in constructed wetlands vegetated with *Juncus effusus*, *Carex lurida*, and *Dichanthelium acuminatum* var. *acuminatum*, *Water Air Soil Pollut.*, 224 (2013) 1–11.
- [32] J.T. Kao, J.E. Titus, W.-X. Zhu, Differential nitrogen and phosphorus retention by five wetland plant species, *Wetlands*, 23 (2003) 979–987.
- [33] H. Zhang, Y. Tian, S. Cui, L. Zhang, X. Zhong, Y. Xiong, Influence of macrophytes on phosphorus fractionation in surface sediments in a constructed wetland: insight from sediment compositions, *Ecol. Eng.*, 97 (2016) 400–409.
- [34] APHA, Standard Methods for the Examination on of Water and Wastewater, 18th ed., American Public Health Association, Washington, D.C., USA, 2005.
- [35] G.M. Pierzynski, Methods of Phosphorus Analysis for Soils, Sediments, Residuals, and Waters, 1st ed., North Carolina State University, Raleigh, Manhattan, 2000.
- [36] S.D. Bao, Soil and agricultural chemistry analysis, China Agricultural Press, Beijing, 2000, pp. 355–356.
- [37] D. Mazumder, Scope of BOD, nitrogen and phosphorous removal through plant–soil interaction in the wetland, *Int. J. Environ. Chem. Ecol. Geol. Geophys. Eng.*, 7 (2013) 82–91.
- [38] E.J. Dunne, M.F. Coveney, V.R. Hoge, R. Conrow, R. Naleway, E.F. Lowe, L.E. Battoe, Y.P. Wang, Phosphorus removal performance of a large-scale constructed treatment wetland receiving eutrophic lake water, *Ecol. Eng.*, 79 (2015) 132–142.
- [39] A.C. Redfield, The biological control of chemical factors in the environment, *Am. Sci.*, 46 (1958) 230A, 205–221.
- [40] C.A. Klausmeier, E. Litchman, T. Daufresne, S.A. Levin, Optimal nitrogen-to-phosphorus stoichiometry of phytoplankton, *Nature*, 429 (2004) 171–174.
- [41] Y.E. Sallade, J.T. Sims, Phosphorus transformations in the sediments of Delaware’s agricultural drainageways: I. Phosphorus forms and sorption, *J. Environ. Qual.*, 26 (1997) 1571–1579.
- [42] D.C. Ribeiro, G. Martins, R. Nogueira, J.V. Cruz, A.G. Brito, Phosphorus fractionation in volcanic lake sediments (Azores – Portugal), *Chemosphere*, 70 (2008) 1256–1263.
- [43] X. Tang, M. Wu, X. Dai, P. Chai, Phosphorus storage dynamics and adsorption characteristics for sediment from a drinking water source reservoir and its relation with sediment compositions, *Ecol. Eng.*, 64 (2014) 276–284.
- [44] J.P. Ondok, Estimation of Seasonal Growth of Underground Biomass, D. Dykyjová, J. Kvet, Eds., *Pond Littoral Ecosystems*, (1978).
- [45] T. Asaeda, P. Hietz, N. Tanaka, S. Karunaratne, et al., Seasonal fluctuations in live and dead biomass of *Phragmites australis* as described by a growth and decomposition model: implications of duration of aerobic conditions for litter mineralization and sedimentation, *Aquat. Bot.*, 73 (2002) 223–239.
- [46] C.H. Sim, M.K. Yusoff, B. Shutes, S.C. Ho, M. Mansor, Nutrient removal in a pilot and full scale constructed wetland, Putrajaya city, Malaysia, *J. Environ. Manage.*, 88 (2008) 307–317.
- [47] R. Carignan, An empirical model to estimate the relative importance of roots in phosphorus uptake by aquatic macrophytes, *Can. J. Fish. Aquat. Sci.*, 39 (1982) 243–247.
- [48] M.L. Jaynes, S.R. Carpenter, Effects of vascular and nonvascular macrophytes on sediment redox and solute dynamics, *Ecology*, 67 (1986) 875–882.
- [49] J. Liu, H. Wang, H. Yang, Y. Ma, O. Cai, Detection of phosphorus species in sediments of artificial landscape lakes in China by fractionation and phosphorus-31 nuclear magnetic resonance spectroscopy, *Environ. Pollut.*, 157 (2009) 49–56.