

Article

## Phosphorus Accumulation Pattern in a Subsurface Constructed Wetland Treating Residential Wastewater

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**Abstract:** A horizontal subsurface flow constructed wetland was investigated after eight years of residential wastewater discharge (150 person equivalents). Twenty core samples distributed over the entire wetland were taken from the soil matrix. The distribution pattern of phosphorus (P) accumulation in the substrate of the wetland was determined using kriging technique and P sorption was related to the content of aluminum (Al), calcium (Ca) and iron (Fe). The correlations found between Al, Ca and Fe content and P accumulation in the bed substrate were weak:  $R^2 = 0.09$ ,  $R^2 = 0.21$  and  $R^2 = 0.28$ , respectively. Great heterogeneity was observed in the distribution of Ca, P and organic matter in the superficial and deeper layers of the bed. Hydraulic problems associated with wastewater discharge and conductivity of the bed substrate were suggested to have negative effects on the wetland performance.

**Keywords:** kriging technique; organic matter; phosphorus distribution; sorption; wastewater

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## 1. Introduction

The use of horizontal subsurface flow constructed wetlands (HSFCWs) is a well-known method for alternative wastewater treatment in rural areas. They are recognized to be effective in the removal of organics and suspended solids from domestic wastewater [1], but they are not particularly effective as phosphorus (P) sinks [2,3]. P removal in HSFCWs occurs via adsorption, precipitation within the wetland substrate and microbial and plant uptake if the biomass is harvested [4,5]. P sorption capacity depends mainly on the type of substrate used in the bed construction [5-8]. HSFCWs are reportedly not effective at P removal because the filtration medium most commonly used (pea gravel, crushed stones) does not contain adequate concentrations of calcium (Ca), iron (Fe) or aluminum (Al) [3]. Sandy materials with high contents of Ca, Fe, Al or magnesium (Mg) theoretically provide the highest P removal [9], but they have much lower hydraulic permeability than either gravel or crushed rock. The maximum P adsorption capacity of a filter medium generally increases as the filter medium size decreases [10,11]. The P removal due to sorption decreases over time because of finite P sorption by the bed [4,12-14], which is referred to as the 'ageing phenomenon' in wetlands that receive wastewater [15]. Richardson [16] observed that in HSFCWs saturated with P, leaching of P from the substrate occurred. Adsorption and precipitation of P by the substrate are finite processes and once the material is saturated, it has to be rejuvenated, or more probably replaced [14].

In this paper, we present data from an investigation of a constructed wetland (CW) in Poland. A wide range of CWs in Poland treat wastewater, from small on-site systems to municipal plants serving several thousand residents. The P removal in small-scale wastewater treatment systems is a critical issue that has not yet been sufficiently resolved. In Poland, there is no discharge limit for P for wastewater treatment plants serving <10,000 person equivalents (PE) and therefore CWs, which usually treat wastewater in rural areas, are not designed for P removal. However, they are efficient in the removal of biochemical oxygen demand (BOD) and suspended solids (SS), which is required by law, making them a very popular solution for on-site treatment. Since the 1990s, over 30 site soil based CWs (Kikuth technology) with daily flow of more than 5 m<sup>3</sup> per day have been implemented in Poland. Extensive studies have been carried out on their treatment efficiency, but such studies have mainly focused on measuring inlet/outlet concentrations of selected parameters.

To our knowledge, this paper is the first to report results from an investigation of substrate-bound P in a HSFCW in Poland and to use the kriging technique to illustrate the horizontal distribution of this element. The aim was to examine P distribution and assess the level of wetland substrate P saturation in a CW that had been treating domestic wastewater for eight years.

## 2. Materials and Methods

### 2.1. Site Description

The HSFCW in Sadowa (a village close to Warsaw) consists of two parallel CW surface areas of 990 m<sup>2</sup> each, planted with common reed (*Phragmites australis* (Cav.) Steud.) and constructed with medium sand, with additions of calcium (Ca), scraped iron, bentonite, crushed bark and straw (Kikuth technology) [17]. Each CW is 0.6 m deep, underlined with gravel and isolated with weld polyethylene liner (2 mm) from the surrounding soil. The plant has been operating since December 1998 and treats

domestic wastewater from 150 inhabitants. The wastewater is pre-treated in a three-chamber sedimentation tank and when distributed to the CW, BOD<sub>5</sub> and soluble reactive phosphorus (SRP) have a concentration of  $245 \pm 112 \text{ mg L}^{-1}$  and  $8 \pm 6.6 \text{ mg L}^{-1}$ , respectively. The CW is loaded with wastewater by pumps several times each day (intermittent discharge) with a total volume not exceeding  $24 \text{ m}^3 \text{ d}^{-1}$ , resulting in a theoretical hydraulic retention time of 8.6 days. A perforated pipe along the width of the bed distributes wastewater. Technical data on the treatment system are shown in Table 1 and selected physicochemical characteristics of the substrate in Table 2. Researchers from Warsaw University of Life Sciences (WULS) monitored the P removal efficiency of the treatment wetland from the year 2000 onwards. Samples of influent (wastewater pre-treated by sedimentation) and of effluent (after CW treatment) were taken at least six times per year (*i.e.*, every second month). Characteristic effluent concentrations of BOD<sub>5</sub> and SRP were recorded to be  $24 \pm 13.3 \text{ mg L}^{-1}$  and  $2 \pm 1.5 \text{ mg L}^{-1}$ . The monitoring results showed a clear decrease in P removal efficiency over the eight years of operation from 96% to 24%, while BOD<sub>5</sub> varied in treatment efficiency having an average of 89% [18]. Plant harvest has still not been undertaken in this CW.

**Table 1.** Technical data on the constructed wetland treatment system in Sadowa, Poland.

Parameter	Value
Person equivalents, (PE)	150
Daily flow of wastewater	
Max	$24 \text{ m}^3 \text{ d}^{-1}$
Average (winter/summer)	$16 \text{ m}^3 \text{ d}^{-1} / 20 \text{ m}^3 \text{ d}^{-1}$
<i>Pre-treatment:</i>	
3-chamber septic tank	Tank volume: $55 \text{ m}^3$
<i>Biological treatment:</i>	
HSF CW (2 parallel beds)	Total surface: $1980 \text{ m}^2$ ( $2 \times 990 \text{ m}^2$ ) Bed length: 33 m Bed width: 30 m Bed depth: 0.6 m
Hydraulic retention time (HRT)	8.6 d
Hydraulic loading rate (HLR)	$0.024 \text{ m}^3 \text{ m}^{-2} \text{ d}^{-1}$
Organic load (Dry matter)	$6.4 \text{ g m}^{-2} \text{ d}^{-1}$
Phosphorus load (SRP)	$0.15 \text{ g m}^{-2} \text{ d}^{-1}$

**Table 2.** Selected physicochemical characteristics of the substrate (medium sand) used in the constructed wetland (n = 3, mean values and SD).

Gravel (>2mm)	Sand (0.05–2mm) %	Organic matter	Al	Ca	Fe	Porosity %	pH in KCl
			% dry matter				
$1.72 \pm 0.72$	$97.81 \pm 3.58$	$0.67 \pm 5$	$0.062 \pm 0.01$	$0.301 \pm 0.01$	$0.144 \pm 0.015$	$37 \pm 5$	$7.11 \pm 2$

## 2.2. Analyses of Samples from the Wetland Soil Matrix

A total of 20 sampling points in one CW were selected with the help of a grid so that all parts of the wetland area were covered. At each point, the substrate was core-sampled to 30 cm depth and each core was divided into two sub-samples: one from 0–10 cm depth and one from 20–30 cm depth. The owner of the CW did not allow us to take samples from deeper levels because of the risk to perforate the bottom plastic liner. The samples were analyzed for P, Ca, Al and Fe content using atomic emission spectrometry with inductively coupled plasma (ICP-MS). The organic matter (OM) content of the substrate was estimated as loss of ignition after removal of living and dead rhizomes and roots, and dried to constant weight at 550 °C. STATGRAPHICS Plus 5.1 [19] was used for statistical analysis (non-linear regression, analysis of variance) and the P distribution was visualized by SURFER; kriging interpolation of values from the 20 observation points [20].

## 2.3. Batch Sorption Experiment

Substrate samples collected from the HSFCW bed (with high and low P content) and unused substrate (medium sand) as reference material was used in a batch sorption experiment to assess substrate P sorption capacity. One gram samples were continually mixed during 24 hours with  $\text{KH}_2\text{PO}_4$  phosphate solution with the concentration of 5, 10, 20, 35 and 55 mg  $\text{PO}_4 \text{L}^{-1}$ . The solution was then filtered and the remaining phosphate concentration was determined by spectrophotometry (Spectrophotometer HACH DR/2400) and analyzed according to the method of Murphy and Riley [21]. P sorption was calculated from the difference between initial and remaining P concentration in solution and fitted by the Langmuir isotherm equation.

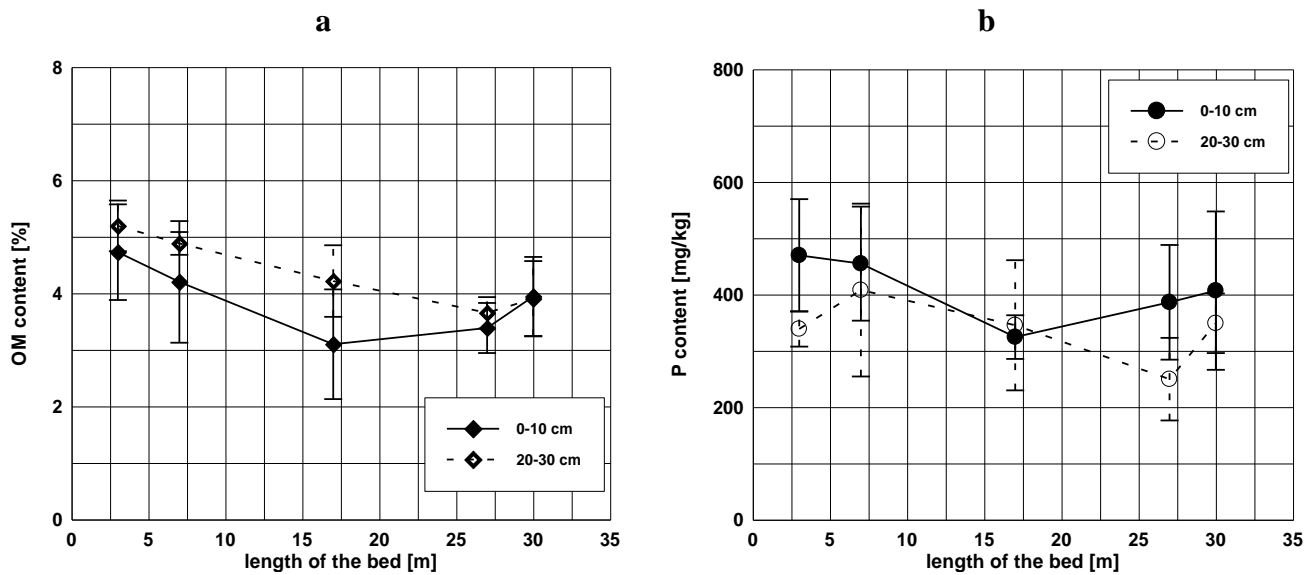
# 3. Results

## 3.1. Accumulation and Distribution of P, OM and Ca

The results showed variations in OM and P content in the substrate throughout the length and depth of the CW treatment bed. However, there was a trend of decreasing OM and P content from the inlet to the outlet area (Figure 1). The highest accumulation of OM and P generally occurred near the inlet zone of the wetland, but high contents were also observed at the outlet zone (Figure 1 and 3). The OM concentrations were higher in the lower layer (20–30 cm) than in the surface layer of the wetland (0–10 cm). Accumulation of Ca was evident in the inlet area. The highest concentrations of Ca were generally observed in the lower layer of the wetland.

In contrast to OM, P accumulation was higher in the upper layer of the treatment bed than in the lower layer (Figure 1). The mean concentration of P in the bed substrate dry matter after eight years of plant operation was 373 mg  $\text{kg}^{-1}$ . In the upper layer (0–10 cm), P concentration varied from 170 to 668 mg  $\text{kg}^{-1}$ , averaging 338 mg  $\text{kg}^{-1}$  and in the 20–30 cm depth layer from 233 to 626 mg  $\text{kg}^{-1}$ , averaging 373 mg  $\text{kg}^{-1}$  (Figure 1). The correlations found between Al, Fe and Ca content and P accumulation in the bed substrate were weak:  $R^2 = 0.09$ ,  $R^2 = 0.28$  and  $R^2 = 0.21$ , respectively.

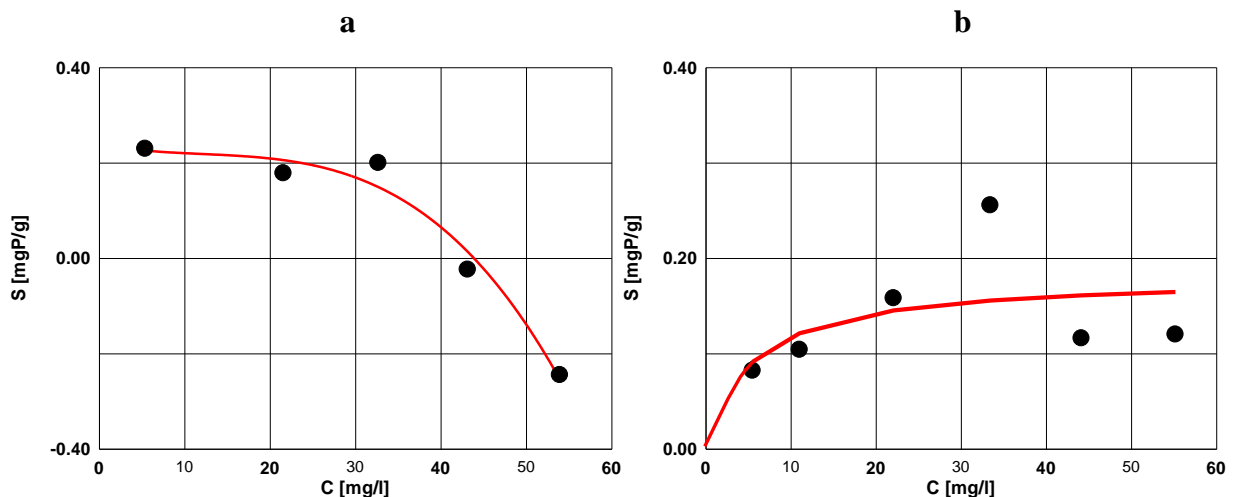
**Figure 1.** Changes in content of (a) organic matter (OM) and (b) phosphorus (P) in the substrate along the length of the constructed wetland treatment bed. Each point represents the mean value and standard deviation of five substrate analyses.



### 3.2. Phosphorus Sorption Capacity of the Substrate

The P sorption capacity of the substrate was estimated for two samples, containing high (549 mg P kg<sup>-1</sup>) and low (300 mg P kg<sup>-1</sup>) amounts of P (Figure 2). The latter sample was also representative of P maximum sorption capacity estimated in the laboratory for sand medium substrate prior to CW establishment in the field (320 mg kg<sup>-1</sup>). The sample representing highly P saturated substrate was not able to bind any more P and was liable to P leaching, *i.e.*, desorption, as shown when the quantity of P adsorbed was plotted as a function of the equilibrium concentration (Figure 2a). In the sample representing lower P content than the apparent sorption capacity of the sand, some sorption capacity was still present (Figure 2b). According to the Langmuir isotherm equation, the calculated S<sub>max</sub> was 0.3 g P g<sup>-1</sup> for the particular bed substrate studied.

**Figure 2.** Phosphorus sorption capacity of substrate samples representing (a) high P content (549 mg kg<sup>-1</sup>) and (b) low P content (300 mg kg<sup>-1</sup>). Note different units on y-axis.



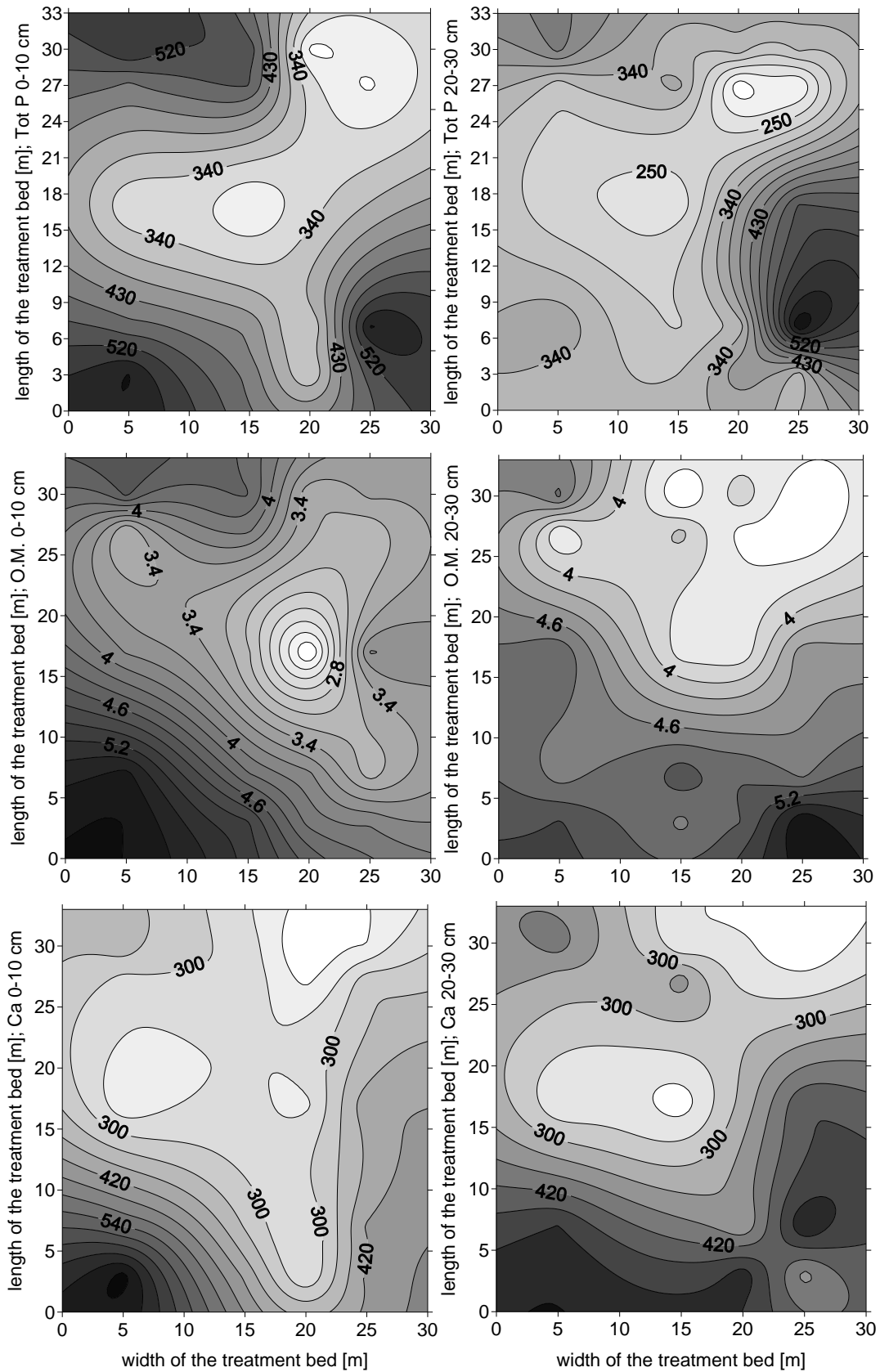
#### 4. Discussion

The lifetime of a bed substrate used for P removal in CWs is an important parameter for practical applications [8,22-24]. The P removal capacity of the wetland investigated here, a type that is commonly used in Poland, rapidly decreased after eight years of operation. We expected the Al, Fe and Ca in the wetland substrate to have a significant effect on P removal, but this proved not to be the case. We suggest that clogging and preferential flow had a role in the distribution patterns observed for Ca, OM and P. However, our investigation was carried out after eight years of operation when P removal efficiency was very low. The common reed was never harvested and extensive and irregular accumulation of decaying plant material could affect the P distribution. The pattern observed in the distribution of P (Figure 3) could also be explained from differences in plant uptake of P. Uneven plant growth was observed during the years of operation but no data was collected to study such a relationship. The substrate in the surface layer of the wetland proved to have higher content of P than the lower layer, indicating that the latter was not 'active' in P removal. An important factor influencing OM accumulation in CWs is the water level in the bed material. If the water is just above the surface, then OM accumulates in a thin layer close to the surface [25]. If the water level is at least 30 cm under the wetland surface, OM accumulates like in natural soil. Runge and Riecken [26] tested the relationships between drainage, P and organic carbon (OC) pattern in soil. They found that OC content decreases with the depth of un-drained soil and its top layer content is higher than that of drained soils.

The maximum P sorption capacity was only  $0.3 \text{ g kg}^{-1}$  for the wetland substrate studied here. According to Xu *et al.* [27], different sands show a varying P sorption capacity from  $0.13$  to  $0.29 \text{ g kg}^{-1}$ . It is well known that the removal of P in all types of CWs is low unless special substrates with high sorption capacity are used. Our study showed that desorption can occur when the wetland substrate is saturated. In combination with anaerobic conditions that regularly occur in CWs, P is released from loosely bound sites. Exhausted wetland substrates can, therefore, become sources instead of sinks for P.

In sand substrates, P is bound to the medium mainly as a consequence of adsorption and precipitation reactions with Ca, Al and Fe [5-8,12,23,28]. At pH levels above 6, the reactions are a combination of physical adsorption to iron and aluminum oxides and precipitation as sparingly soluble calcium phosphate [24,29]. Precipitation as an insoluble Ca-P can occur at pH values greater than 7 [3]. As the capacity of the medium to remove P may be dependent on Ca, Al and Fe content, some researchers have attempted to improve P removal in CW by injection of Ca [11,30,31] or by enriching the bed substrate with Fe (iron chips or iron-rich sand from water treatment plants) as in systems based on the Kickuth technology. However, the results from these studies have shown that using a medium with high iron and aluminum content is effective only during the first few months of operation [32,33]. Most of the CWs studied showed a decline in P removal efficiency after 1–2 years [34,35] or 4–5 years [36]. The theoretical lifetime of one particular CW filled with sand, with a sorption capacity of  $130 \text{ mg P kg}^{-1}$ , was estimated to be around nine months [27]. A number of studies have indicated that addition of reactive Ca (CaO or Ca(OH)<sub>2</sub>) is more efficient in improving the P adsorption capacity than Al and Fe addition [11,37,38].

**Figure 3.** Horizontal distribution of P ( $\text{mg kg}^{-1}$ ) (top panels), organic matter (OM) (%) (middle panels) and Ca ( $\text{mg kg}^{-1}$ ) (bottom panels) in the upper layer (0–10 cm) (left) and lower layer (20–30 cm) (right) of the horizontal subsurface flow constructed wetland.



In agreement with findings reported by Tanner *et al.* [39] and Tang *et al.* [40], the OM accumulation was highest in the inlet zone of the Sadowa wetland, but in some cases the OM content also increased in the outlet zone of the HSFCW (Figure 3). Contrary to our results, Tanner and Sukias [41] observed higher OM accumulation in the top layer (<10 cm) of planted and unplanted constructed wetlands. Drizo *et al.* [42] found a significant inverse relationship between P concentration in wastewater flowing through a bed planted with macrophytes and distance from the inlet.

One of the possible mechanisms of P immobilization in constructed wetlands is incorporation of P into accumulated OM [27]. However, OM accumulation can potentially contribute to the clogging of pore space, thus decreasing the wetland purification potential. The relationship between P sorption and organic matter content can vary, with some studies reporting a negative correlation [27,43] and others a positive correlation [44]. In the Sadowa wetland, the correlation between OM content and P accumulation in the bed substrate was weak. Wetland vegetation is important for BOD removal, and instead of the fine sand used in the Sadowa HSFCW, a porous reactive medium such as Polonite<sup>®</sup> should have been used [45]. Such material could improve the hydraulic conductivity and increase the lifetime of the CW for P removal.

## 5. Conclusions

The horizontal distribution in P accumulation varied widely in the Polish wetland studied and sorption experiments showed that only minimal P removal capacity remained after eight years of operation of the plant. The uneven patterns of P, Ca and OM concentrations in the substrate of the wetland reflect the complicated processes that take place. The P content in the wetland substrate was generally higher than the value received from the Langmuir isotherm modeling, indicating that the site soil used here was exhausted despite efforts to amend with materials rich in calcium and iron. Based on this research and previous studies it seems that CWs work as reactors for primarily BOD removal and that P can only efficiently be removed and recovered via reactive filter units preceding or following the wetland treatment.

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