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Phosphorus removal and effect of adsorbent type in a constructed wetland system

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ABSTRACT

This research project aimed to determine the technologically feasible and applicable wastewater treatment systems which will be constructed to solve environmental problems of small communities in Turkey. Pilot-scale treatment of a small community's wastewater was performed over a period of more than 2 y in order to show applicability of these systems. The present study involves removal of phosphorus in horizontal (HFCW) and vertical (VFCW) sub-surface flow constructed wetlands operated in series. The pilot-scale wetland was constructed downstream of anaerobic reactors at the campus of TUBITAK-MRC. Anaerobically pretreated wastewater was introduced into this hybrid two-stage sub-surface flow wetland system. Wastewater was first introduced into the HFCW and then VFCW before being discharged. VFCW achieved up to 60–90% phosphorus removal whereas HFCW could remove only less than 20%. The effect of type of filling material on adsorption of phosphorus was investigated both in adsorption studies and in 1 m^2 constructed wetlands filled with different materials. The results showed that iron slag was the most efficient material for phosphorus removal in constructed wetlands compared to gravel, marble stone and zeolite.

Keywords: Horizontal sub-surface flow; Vertical sub-surface flow; Hybrid constructed wetland; Phosphorus removal; Filling material; Phosphorus adsorption

1. Introduction

Constructed wetlands are considered as low-cost alternatives for the treatment of domestic wastewaters due to the advantages such as lowering the initial costs by using cheap materials, eliminating the need for sludge removal, and developing a pathogenically safe as well as aesthetic treatment by applying sub-surface flow [1,2]. Constructed wetlands generally remove about 80–99% of organic matter, 92–95% of bacteria, 30–80% of nitrogen and 20–70% of phosphorus from domestic wastewaters depending on the plant type used and flow regime [3]. A previous study in Turkey showed 93% COD, 90% nitrogen and 60% phosphorus removal by a recirculating constructed wetland [1]. Another application of constructed wetland for the treatment of anaerobically treated domestic wastewater of 500 people resulted in 84% COD, 92% TSS, 40% nitrogen and 54% phosphorus removal [4]. Application of anaerobic pretreatment protects constructed wetlands from clogging [5,6] and decreases the land requirement for the constructed wetlands by decreasing the organic matter loading.

Phosphorus concentration of 6–10 mg/l in domestic wastewaters may induce eutrophication in receiving water bodies. Therefore removal of phosphorus is

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an important issue in the treatment of domestic wastewaters. Constructed wetlands can remove phosphorus which is mostly in the form of orthophosphates. Removal of phosphorus occurs in constructed wetlands through several mechanisms, such as uptake by plants, sorption on filling material or soil, sedimentation and complexation. Sorption is probably the most important mechanisms [7,8]. Adsorption involves selective sorption to mineral interfaces as well as sedimentation reactions [8,9]. Phosphorus captured in a constructed wetland is finally removed through harvesting of plants and replacement of loaded wetland materials. Therefore the most important factor which affects the phosphorus removal capacity of a wetland is the capacity of wetland materials for phosphorus uptake [10].

Phosphorus accumulation may be as high as 23–25 $g/m^2/y$ at the lower layers of a constructed wetland. This value is 3–240 t greater than the values obtained for wetlands with mineral soil, and 22–600 t greater than the values for natural wetland systems [11,12]. These findings showed that most of the phosphorus uptake occurs via sorption on filling material. However, uptake by plants is another contributing way of phosphorus removal. Phosphorus uptake may be 1.4–2 t greater in a constructed wetland with plants compared to the ones without plants [13]. P is particularly stored at the roots of the plants rather than leaves [14].

In a constructed wetland, phosphorus adsorption capacity depends on two factors; surface area of the filling material and pH. Adsorption of phosphorus is the highest in alkaline wetlands containing high amounts of calcium. As an example, calcium undergoes a reaction with soluble phosphate at high pH and the formed calcium hydroxyapatite precipitates [12,15]. In the selection of media for constructed wetlands, chemical and physical characteristics of the material should be evaluated [16,17]. These materials should involve on their surfaces Al, Fe or oxide groups which can easily undergo reactions, or Ca which can precipitate PO_4 [18]. Materials with fine particles are expected to have a high P adsorption capacity, since they usually have large surface areas [12]. In the selection of filling material, hydraulic conductivity is also important [19]. If the particles are too small, hydraulic conductivity decreases.

The most widely used filling materials in constructed wetlands is gravel because of its easy and economical availability. However, gravel is not effective in terms of phosphorus removal [8,20]. On the other hand, the presence of iron within the structure of gravels may increase their removal efficiencies for example from 20% [21] up to 90% [12]. Iron slag, an industrial by-product, may be an alternative filling material for increased P sorption since it contains high amounts of Ca, Fe and Al.

In this study pilot-scale studies were performed for the treatment of domestic wastewater of about 30 people in order to show applicability of constructed wetland systems following an anaerobic pretreatment. The experimental period covered more than 2 y (26 m). The present study involves removal of phosphorus in the treatment of domestic wastewater in horizontal and vertical sub-surface flow constructed wetlands operated in series. In order to investigate the effect of filling material, adsorption experiments and small-scale constructed wetland experiments were performed with gravel, marble stone, iron slag and zeolite.

2. Materials and methods

2.1. System configuration

The pilot-scale wetland was constructed downstream of anaerobic reactors at the campus of TUBITAK-MRC for the treatment of domestic wastewater of about 30 people. Wastewater flow to the system was about 3000 l/d. Pretreatment of domestic wastewaters of residential flats at the campus was performed in anaerobic reactors at ambient climate conditions. Anaerobically pretreated wastewater was collected in a basin before being introduced into the hybrid two-stage sub-surface flow wetland system. The hybrid system involved a horizontal sub-surface flow system (HFCW) and a vertical flow system (VFCW) operated in series (Fig. 1). Wastewater was first introduced into the horizontal sub-surface flow system (HFCW) by gravity. Effluent of HFCW was fed batch-wise to the vertical flow system (VFCW) with a submerged pump. Discharge was also pumped batch-wise. Discharge of the hybrid constructed wetland system was further treated for phosphorus removal in small-scale constructed wetlands filled with different materials.

HFCW aimed to perform removal of organic matter and support denitrification. VFCW aimed to obtain nitrification in the wastewater after achieving low levels of organic matter. HFCW was considered to be a buffer zone to protect VFCW from clogging and thereby increase nitrification performance in VFCW. It was aimed to enhance oxygen transfer and nitrification in VFCW by aeration pipes. Recirculation from VFCW to HFCW was performed in order to remove nitrate by means of denitrification in HFCW. The filling material used for VFCW was composed of marble stone, sand, lime stone and gravel and aimed to provide phosphorus removal besides ammonia removal. On the other hand, gravel was used as the filling material in HFCW.

The design and operational parameters involved flowrates of 2–3 m^3/d, base slopes of 0.001%, planting densities of 4 rhizome/m^2 and gravel as base material for both wetlands. Surface areas were 18 m^2 and 13.7 m^2, dimensions were 3×6×0.8 m and 3.7×3.7×0.8 m, hydraulic retention times ranged from 1.4 to 2.2 and from 0.5 to 1 d, hydraulic loading rates were 111–167 l/m^2 d and 146–219 l/m^2 d, and initial porosities were 28% and 33%, respectively for the horizontal and vertical flow systems. Hydraulic retention times were kept low and therefore hydraulic loading rates were relatively high. In VFCW, plants were well developed. However growth of plants was insufficient in HFCW. The hybrid system was operated for about 26 m. Samples were taken periodically from the inlet and outlet of both HFCW and VFCW for analysis.

2.2. Adsorption experiments

Adsorption experiments were performed in order to investigate the adsorption capacities of filling materials used in the system. Gravel, marble stone, iron slag and zeolite at 10 different weights were contacted with 500 ml wastewater in a shaker at room temperature of 21°C. Equilibrium time required for adsorbents to be loaded with phosphorus was determined to be 96 h. Wastewater used in the adsorption experiments was the effluent of the system described above, i.e., the effluent of the two-stage constructed wetland system which followed anaerobic treatment. The wastewater contained 7.81 mg/l TP and 3.6 mg/l PO_4-P. Total phosphorus (TP) and PO_4 analysis were performed with the samples taken at the end of the adsorption process after 96 h. Adsorption capacities (in terms of g P/g adsorbent) were determined from the differences between the initial and final concentrations. Adsorption experiments were conducted with two parallels.

2.3. Experiments with 1 m^2 constructed wetlands

The effect of the use of different filling materials were also investigated in small-scale (1 m^2) phosphorus removing horizontal flow constructed wetlands (P-HFCW) located at the end of the treatment system as described in Fig. 1. These small-scale wetlands consisted of 1×1 m polyester reservoirs with a depth of 30 cm. Four different materials, namely gravel, marble stone, iron slag and zeolite were investigated comparatively with and without plant in 8 wetlands in order to investigate the effect of plants on phosphorus removal. 4 reservoirs were filled only with each filling material and were not planted. 4 other reservoirs filled with each material were planted. The P-HFCW system was fed with the effluent of vertical-flow constructed wetland (VFCW) which contained 7.58 ± 2.80 mg/l TP and 5.30 ± 2.42 mg/l PO_4-P at a hydraulic loading rate of 100 l/m^2 · d. P-HFCWs (1 m^2 wetlands) were operated for three months.

Also, small-scale wetlands described above were used in series in order to investigate the total effect of two different types of filling materials. These two-stage adsorption systems were operated such that one of them contained 1 m^2 wetlands filled with zeolite and iron slag in series and the other contained 1 m^2 wetlands filled with zeolite and marble stone in series. These experiments lasted two months.

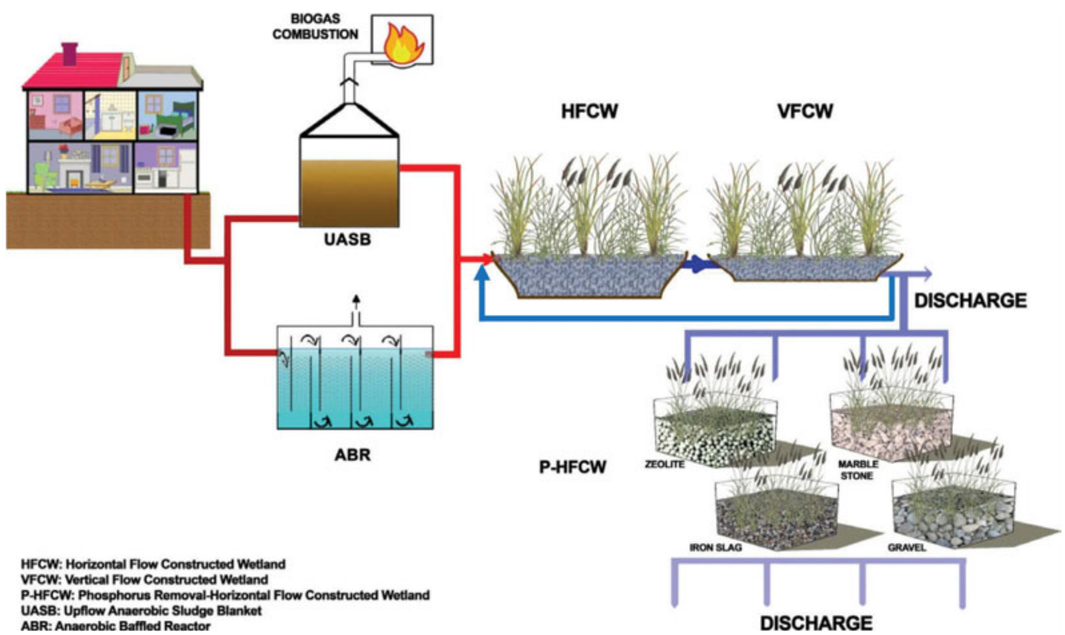

Fig. 1. Flow diagram of the hybrid constructed wetland system with anaerobic pretreatment.

2.4. Analyses

All analyses of total P and PO_4 were performed according to the Standard Methods for the Examination of Water and Wastewater [22]. These analyses were performed in the accredited laboratories of TUBITAK Marmara Research Center. The chemical composition of filling materials was determined through elemental analyses performed by Philips PW–2404 X-ray Fluorescence (XRF) Spectrometer. The method involves qualitative determination of the elements inside an inorganic or organic material, semi-quantitative determination of elements and compounds (analysis without the use of a standard), and quantitative elemental analysis of materials based on wavelengths of elements with the use of standard reference samples.

3. Results and discussion

3.1. Phosphorus removal in two-stage hybrid constructed wetland system

Phosphate removal was usually below 20% in HFCW as seen in Fig. 2a. Sometimes the effluent P concentrations even exceeded the influent values. HFCW contained gravel as filling material. Therefore, removal via adsorption was not high. Another reason for low P removal in HFCW could be the insufficient growth of plants in this wetland.

On the other hand, in the VFCW, PO_4-P removal efficiency was between 60–90% during the first three months of operation as seen in Fig. 2b. Following this highly efficient period, P removal decreased during a succeeding 3–4 m period. This showed that the

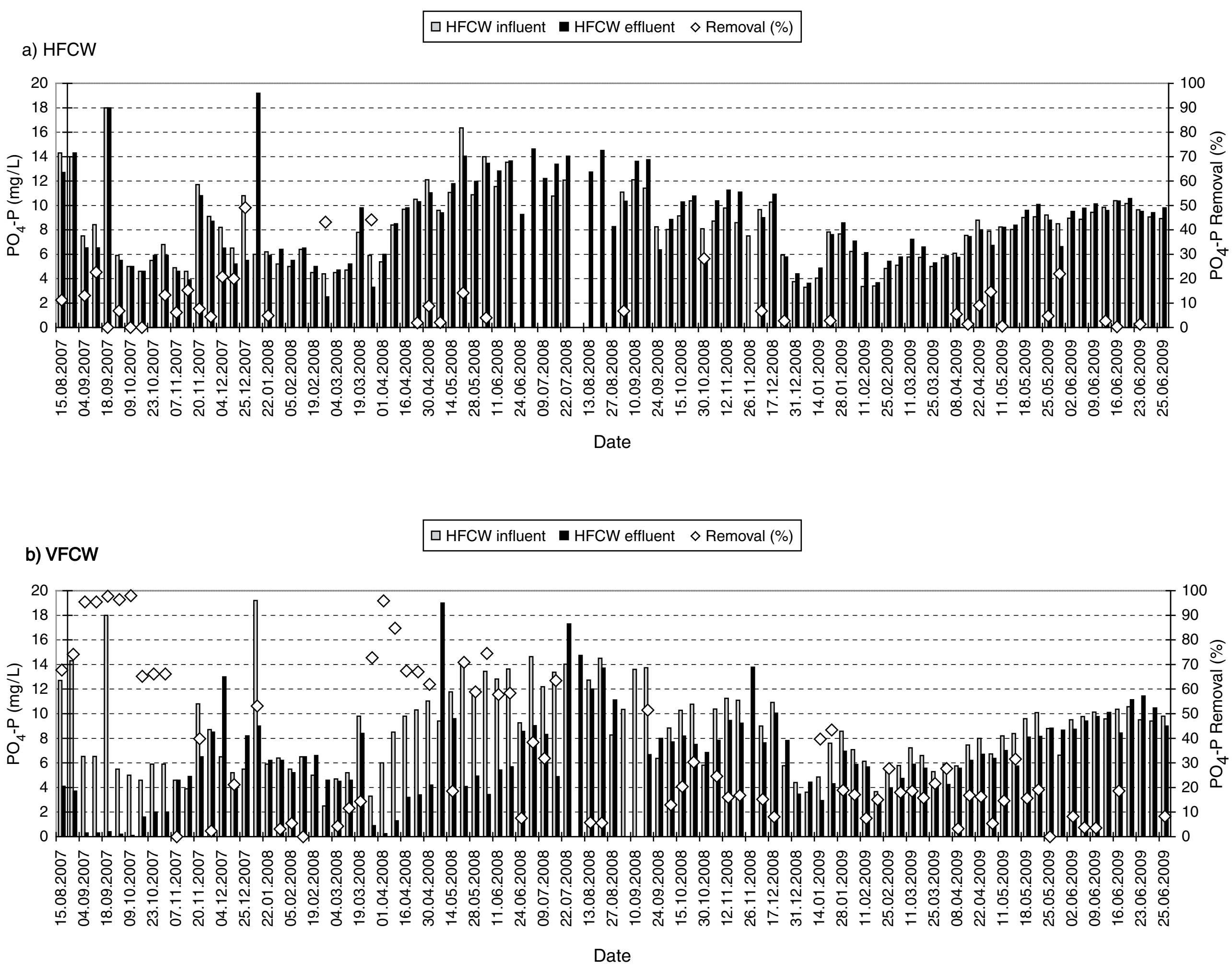

Fig. 2. Influent and effluent PO_4-P concentrations and removal efficiencies in a) HFCW and b) VFCW.

filling material and the plants were saturated with P. However, later P removal efficiency recovered and increased up to 60–90% for a new 3 m period between April and June 2008 (Fig. 2b). This was attributed to application of high hydraulic loading rates which probably opened new sites for sorption and sedimentation of phosphorus via complexation. In the later periods of operation which lasted for about a year, P removal efficiency decreased to below 40% since the capacity of the wetland deteriorated within time.

Higher P removal efficiencies in VFCW compared to HFCW can be attributed to the presence of lime stone within marble stone which can enhance phosphorus precipitation and adsorption. Another reason was better growth of plants in VFCW.

3.2. Adsorption studies

Adsorption capacities calculated for four filling materials; gravel, marble stone, iron slag and zeolite are shown in Table 1. P adsorption capacities of the filling materials were very low except iron slag. In addition, the materials except iron slag did not fit the isotherm equations namely, Freundlich, Langmuir and BET.

The adsorption isotherm of iron slag for TP was found to fit the Langmuir and BET expressions. The linear form of the Langmuir equation is given in Eq. (1). From the slope and intercept b and q_{max} were calculated as 2.123 and 0.26 mg/g, respectively.

$$\frac{1}{q} = \frac{1}{q_{\max}} + \left(\frac{1}{b\, q_{\max}}\right)\left(\frac{1}{S_e}\right) \tag{1}$$

where S_e: the equilibrium adsorbate concentration (mg/l); q: adsorption capacity (mg adsorbate adsorbed per g adsorbent); q_{max}: solute adsorbed per unit weight of adsorbent in forming a complete monolayer on the surface, (mg/g); b: a constant related to the energy of adsorption.

Isotherm curve for iron slag at 21°C is shown in Fig. 3. Saturation times were also calculated for each filling material based on their average phosphate adsorption capacities and assuming a hypothetical pool filled with 10 tons (equivalent capacity of VFCW) of each material and fed with a loading rate of 24 g PO_4-P/d (8 mg PO_4-P /l at 3 m^3/d) and all PO_4 is removed through adsorption (Table 1). In accordance with its high adsorption capacity, iron slag was the material being saturated the latest. However, it should be noted that these values were obtained at laboratory conditions and may be different on-site considering other factors. All adsorption experiments were repeated at 4°C, but it did not provide any significant difference from the experiments performed at 21°C. Therefore temperature was not an important factor for phosphorus adsorption.

The higher adsorption capacity of iron slag can be mainly attributed to its high Fe content. Elemental analysis showed that iron slag contained Fe (in the form of Fe_2O_3) at a ratio of about 27% in its composition, whereas the ratio of Fe was less than 2% for the other three materials. The ratio of Ca (in the form of CaO) was about 21% in iron slag. Although marble stone and gravel contained about 50% Ca in their compositions, their adsorption capacities were much lower compared to iron slag. Aluminum (in the form of Al_2O_3) content was the highest in zeolite (about 7%), less in iron slag and gravel (about 3%) and very low in marble stone (0.13%). These results showed that in terms of P removal, the presence of iron was a more important factor compared to the presence of Ca and Al within the composition of filling material.

The results of the study were in accordance with a previous study which showed that oven slag adsorbed much more than natural zeolite and soil from an operating wetland [23]. Natural zeolite was the least PO_4-P adsorbing material although it is widely used for NH_4-N removal. Another study also showed that PO_4-P adsorption capacity

Table 1
Adsorption capacities and saturation times required for 10 tons of different filling materials at a loading rate of 24 g PO_4-P/d

Filling material	q (gr PO_4-P/kg material)	Saturation time (days)
Gravel	0.017	6.89
Marble stone	0.025	10.56
Iron slag	0.110	45.99
Zeolite	0.020	8.39

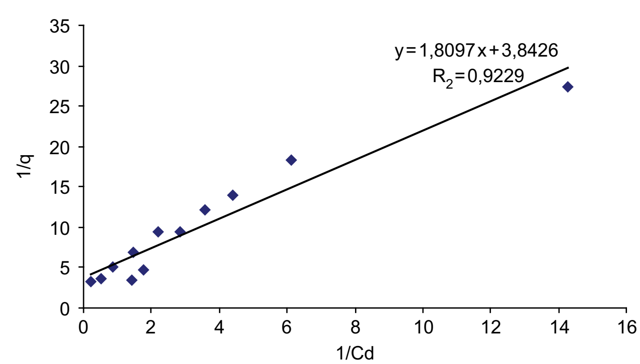

Fig. 3. Adsorption isotherm for PO_4-P obtained for iron slag.

of zeolite was less than other materials although it had the highest surface area [17]. More than 50% of P could be adsorbed by all materials at low P feed concentrations (0–200 mgP/l) [23]. However at high P feed concentrations (0–200 mgP/l), all soil types and zeolite could adsorb 30%, although oven slag and iron slag removed more than 50%. The finding was attributed to high amounts of Al, Fe and Ca minerals within the slag. Another study also showed that blast furnace slag is an appropriate filter media for adsorption of phosphorus [24].

The service time of a constructed wetland is dependent on the filling material as well as the area of the wetland per person. P removal efficiency starts to decrease after 1–2 y of operation in a constructed wetland filled with gravel [20]. A literature study showed that P removal efficiency which could be more than 90% during the first 4–5 y of operation exhibited drastic decreases afterwards [25]. The lifaspan of a typical constructed wetland is expected to be up to 20 y for an area of 10 m^2/person and a bed depth of 0.9 m [17].

3.3. Phosphorus removal in 1 m^2 constructed wetlands

As described in Section 2.3, small-scale horizontal-flow wetlands were operated either with or without plants. The results obtained in the absence of plants were compared with the ones obtained in wetlands where plant growth was achieved. The comparisons enabled investigation of both the effect of plants and filling material on phosphorus removal.

The efficiencies of the small-scale constructed wetlands were usually insufficient for the removal of phosphorus because of clogging problems. The performance of the wetlands decreased after a short period following the start of operation. Particularly in summer periods, algae formation at the surface occurred which led to clogging in the system and increased nutrients in the effluent. This effect generally resulted in higher phosphorus concentrations in the effluents of wetlands with plants. Iron slag was found to be the only material which could achieve satisfactory TP and PO_4 removals both in the absence and presence of plants. Fig. 4

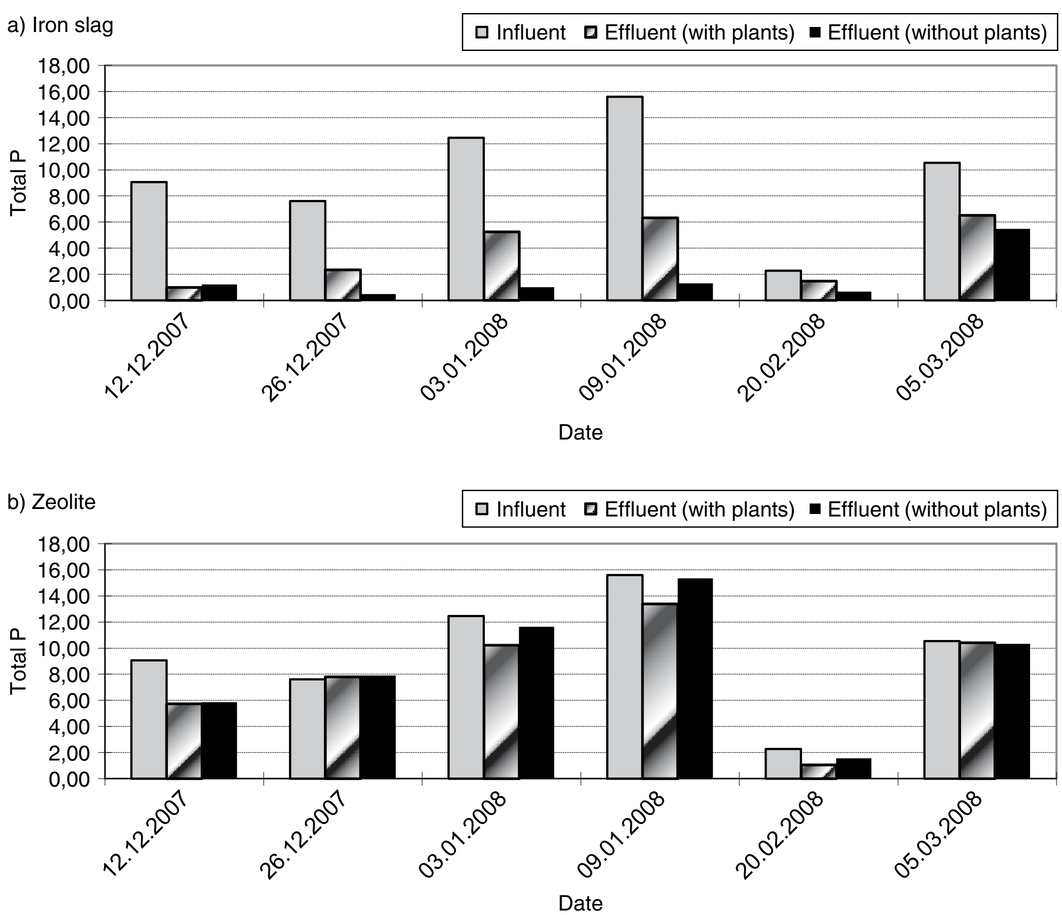

Fig. 4. Influent and effluent total P concentrations in 1 m^2 wetlands (P-HFCW) filled with a) iron slag and b) zeolite.

shows the removal of total P comparatively in small-scale wetlands filled with iron slag and zeolite. Effluent TP could be decreased to below 6 and 2 mg/l in the absence and presence of plants, respectively during the first two months of operation after which the effluent quality deteriorated. The effluent TP was usually above 6 mg/l for the wetlands filled with other three materials both in the absence and presence of plants. Fig. 5 shows that effluent PO_4-P concentrations were usually below 1 mg/l in wetlands filled with iron slag. These values were also much less compared with the wetlands filled with other three materials which usually had more than 3 mg/l PO_4-P in their effluents as can be seen in Fig. 5 in the case of zeolite. These findings are in agreement with the results of a previous study also showing that horizontal-flow constructed wetlands filled with gravel and crushed rock were not effective in phosphorus removal [26].

The problems encountered in 1 m² P removing wetlands necessitated the utilization of two-stage adsorption systems in series. Two systems were formed by connecting two 1 m² wetlands in series; first with zeolite-iron slag, second with zeolite-marble stone in series. Zeolite was selected as the first stage in both systems, since it is well-known for its NH_4-N adsorption. The effluent of VFCW was fed to the zeolite bed for further nitrogen removal. On the other hand, iron slag and marble stone were used as the second-stage basically for phosphate adsorption. Clogging in the system and algal blooms on the surface were also occasionally observed in the two-stage systems. During some periods, effluent P concentrations were very close to the influent values or even higher than that. The reason was P released from algal blooms and the saturation of the filling materials with P. Application of two-stage system with zeolite-iron slag did not seem to provide additional P removal.

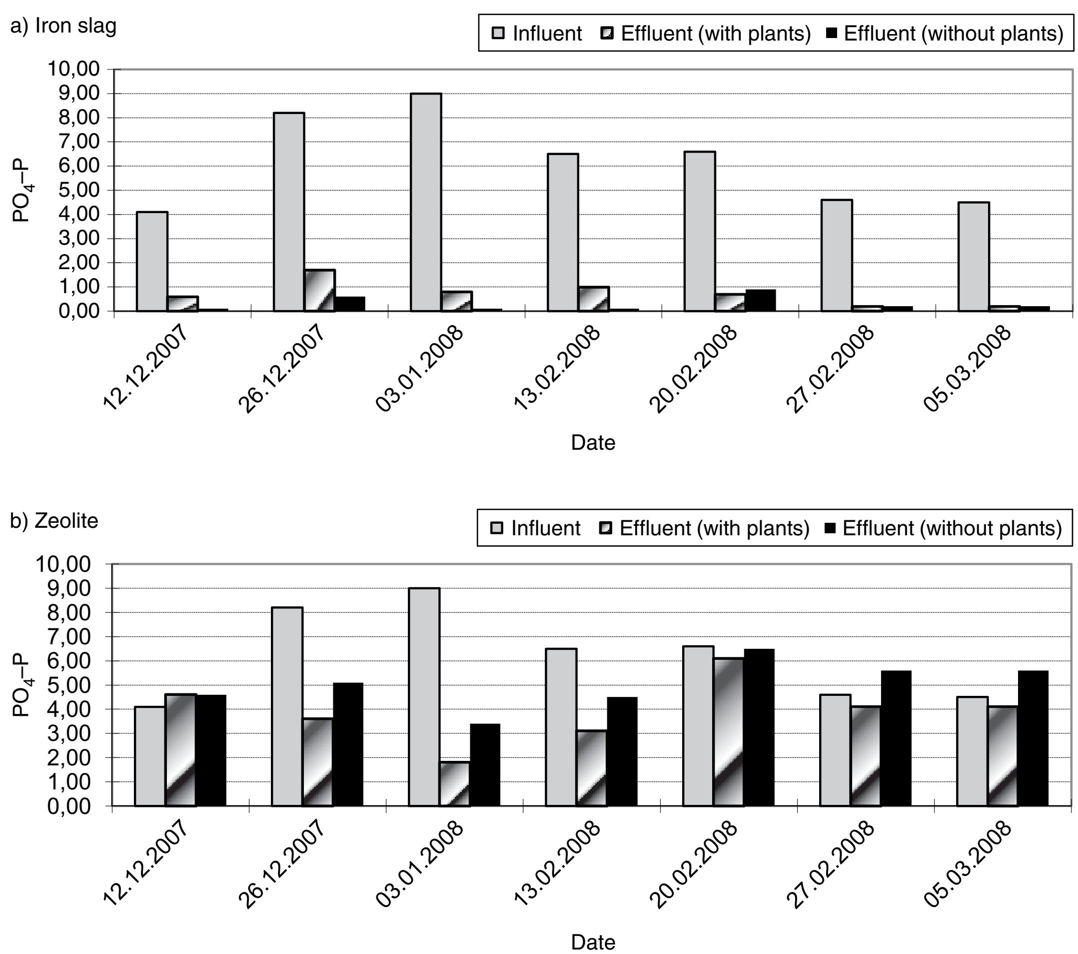

Fig. 5. Influent and effluent PO_4-P concentrations in 1 m² wetlands (P-HFCW) filled with a) iron slag and b) zeolite.

In the two stage system with zeolite-marble stone, TP removal seemed to increase during the first month of operation. However effluent TP concentrations drastically increased later on up to more than 10 mg/l.

4. Conclusions

Pilot-scale treatment of a small community's wastewater was performed over a period of more than 2 y and showed the applicability of constructed wetland systems for the removal of phosphorus in horizontal (HFCW) and vertical (VFCW) sub-surface flow constructed wetlands operated in series. VFCW achieved up to 60–90% phosphorus removal whereas HFCW could remove only less than 20%. Higher P removal efficiencies in VFCW compared to HFCW is attributed to use of marble stone in VFCW instead of gravel and insufficient growth of plants in HFCW. The investigation of the effect of type of filling material on adsorption of phosphorus both in adsorption studies and in 1 m^2 constructed wetlands showed that iron slag was the most efficient material for phosphorus removal in constructed wetlands compared to gravel, marble stone and zeolite. The results of this pilot-scale research study will be used for determination of technologically feasible and applicable wastewater treatment systems which will be constructed to solve environmental problems caused by small communities in Turkey.

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