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Journal of Hazardous Materials 155 (2008) 76–82

Journal of Hazardous Materials

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The use of constructed wetland for dye-rich textile wastewater treatment

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Received 30 July 2007; received in revised form 12 November 2007; accepted 12 November 2007
Available online 23 November 2007

Abstract

The objective of the present paper was to examine the treatment efficiency of constructed wetlands (CW) for the dye-rich textile wastewater with special focus on colour reduction. Preliminary, a series of dynamic experiments was performed in the CW model packed with gravel, sand, and zeolitic tuff on three synthetically prepared wastewaters using chemically differ dyestuffs, auxiliaries and chemicals, in order to investigate the potential of low-cost materials as media for textile dye-bath wastewater treatment. The obtained results evidence that applied CW model reduces colour by up to 70%, and COD and TOC by up to 45%. Based on these results, the pilot CW with vertical (VF) and horizontal flow (HF) was constructed near textile factory mainly for cotton and cotton/PES processing with intention to treat real textile wastewater in situ. It was designed for 1 m^3/day, covering 80 m^2, packed with sand and gravel, and planted with *Phragmites australis*. The average treatment efficiency of the CW for the selected pollution parameters were: COD 84%, BOD_5 66%, TOC 89%, N_{total} 52%, $N_{organic}$ 87%, NH_4-N −331%, sulphate 88%, anion surfactant 80%, total suspended solids (TSS) 93%, and colour 90%, respectively. The results unequivocally proved that the CW could offer an optimal solution to meet the environmental legislation as well as requirements for effective and inexpensive textile wastewater treatment.

Keywords: Coloured textile wastewater; Constructed wetland; *Phragmites australis*

1. Introduction

Coloured textile effluents represent severe environmental problems as they contain mixture of chemicals, auxiliaries and dyestuffs of different classes and chemical constitutions with elevated organic parameters such as chemical oxygen demand (COD), total organic carbon (TOC), adsorbable organic halogens (AOX), inorganic parameters such as metals, chloride, sulphate, sulphide and nitrogen. A literature review regarding dye-bath wastewater treatments reveals the consideration of different approaches to handling such effluents, which include biodegradation, adsorption, advanced oxidation and membrane filtration [1–5]. Choosing the most appropriate treatment method or combination depends on the nature and the amount of effluent from textile processing plant.

In the past few years, we became aware that systems imitating the self-cleaning ability of natural wetland ecosystems by establishing optimal physical, chemical and biological conditions for in situ wastewater treatment should be considered with greater importance [6]. Constructed wetland (CW) is an example of such system that is also simple to use, environmentally friendly, with low construction and operational costs, and efficient enough to treat diverse wastewaters, although the experience in treating textile wastewaters is limited. CW's designs differ regarding to the type of flow and applied bed material [7,8]. The removal efficiencies of natural systems could be exceedingly variable, and influenced by numerous parameters such as water/bed material temperature, air temperature, sedimentation, pollutant concentration, and vegetation. These parameters cause changes in uptake or release of chemical substances, and biochemical activities of microorganisms and plants.

Methods of decolourisation of dyes, reported by a number of researchers [1–5], are often not feasible for treating dye-rich wastewater because of technology intensiveness, reliable power demand, and complexity of components, unproven long-term effectiveness and high investment and maintenance costs [9]. Promising results of CW recorded in last few years for industrial wastewater and lacking of literature on textile wastewater treatment with CW conducted us to research the viability of treating dye-rich wastewater with CW. The main objective

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doi:10.1016/j.jhazmat.2007.11.068

of our experiment was to establish a model of CW and an integrated CW system, belonging to the generation of VHSSF (vertical–horizontal-sub-surface flow) and to evaluate its applicability in the treatment of coloured textile wastewater. The experiment was based on the selection of different bed material and mass loadings to reveal which option is more effective.

2. Materials and methods

2.1. Experimental equipment and design

A series of dynamic trials was accomplished in two stages, firstly on a CW model and secondly, on a pilot scale CW. The CW model was designed as a preliminary experiment for colour reduction of selected dyestuffs. In the model gravel and sand was used as a bed material with an addition of zeolitic tuff to reduce the risk of inefficiency. Promising results of colour removal in the CW model was encouraging to test pilot CW for real textile water, containing similar dyestuffs. The pilot CW was constructed using bed material comparable with the CW model without zeolitic tuff. The purpose was to proof the efficiency for colour removal with common material (gravel, sand) to keep low construction cost.

2.1.1. CW model

It is a vertical-flow VFCW model, schematically presented in Fig. 1, made from polyethylene plates and Plexiglas, with dimensions of 0.8 m in length, 0.3 m in width, and 0.6 m in height that provides an empty volume of 144 l. The model was coated by PE-foil and filled with three different natural materials in layers from the bottom to up: washed gravel with particle size of 8 mm/12 mm, washed sand with particle size of 0 mm/4 mm, and washed zeolitic tuff with particle size of 9 mm/12 mm and chemical composition of 62.95% SiO_2, 15.92% Al_2O_3, 3.10% Fe_2O_3, 3.81% CaO, 1.31% MgO, 4.67% Na_2O, 4.67% K_2O and 0.03% SO_3, up to 29 cm. A constant 0.2–0.24 l/min vertical wastewater flow from the vessel downwards through the porous medium packed in CW model (across the entire width) was achieved by a perforated plastic pipe, located at the top of the model. The actual retention time was 1, 2, 3, 4, 5 and 24 h. The outflow of treated effluent was located at the bottom of the model.

2.1.2. Pilot scale CW

It was constructed as a VHSSF system (Fig. 2), consisting of three interconnected beds. The principal design criteria was based on a project started in 1991 in Austria [10], modified in 1997 [11] and upgraded according to the results obtained by means of laboratory scale CW experiments (optimization of retention time, flow and bed media with regard to the decolourisation efficiency of structurally different dyestuffs). The CW covered an area of 80 m^2 in total, which was planted with common reed (*Phragmites australis*) with 5 clumps/m^2, transferred from an old not-operating CW for sewage treatment. Two vertical flow (VF) beds covered 20 m^2 (5 m long × 4 m wide) each, with a depth of 0.6 m. The horizontal flow (HF) bed covered 40 m^2 (8 m × 5 m) with an average depth of 0.5 m. Bed material is composed as follows:

VF beds:	Coarse sand with particle size of 4 mm/8 mm fine sand with particle size of 0 mm/4 mm ratio; 1:1
HF bed:	Coarse sand with particle size of 8 mm/16 mm fine sand with particle size of 0 mm/4 mm ratio; 1:1 0.5 m wide inlet/outlet section with gravel with size of 16 mm/32 mm

The bottoms of both beds were fortified with a 1 mm thick LDPE foil. The real textile wastewater was transported with a 5 m^3 washed road tanker from the near textile factory and pumped into the retention reservoir at the pilot CW. The purified water was collected in a collective sump with outflow back to the wastewater treatment plant (WWTP).

2.1.3. Mode of operation

The surface hydraulic load of VF beds was intermittent, adjusted by the aperture of the valve. The VF bed was loaded

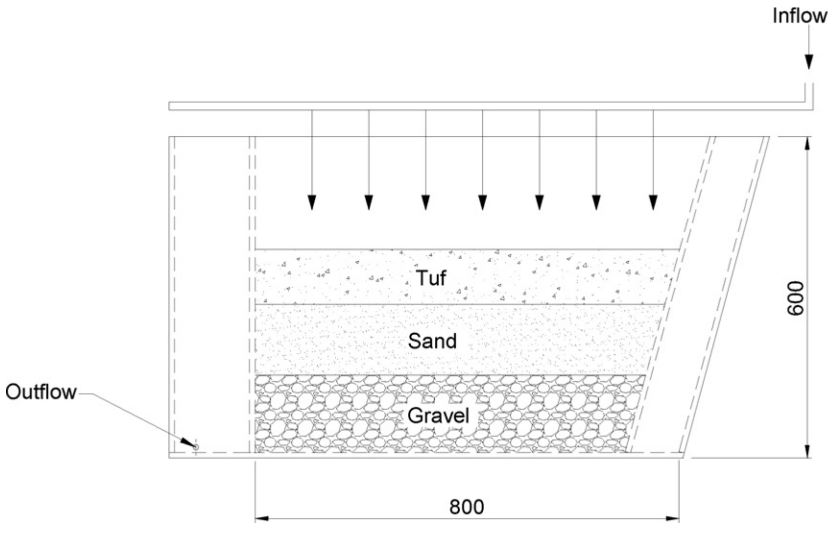

Fig. 1. Scheme of the CW model.

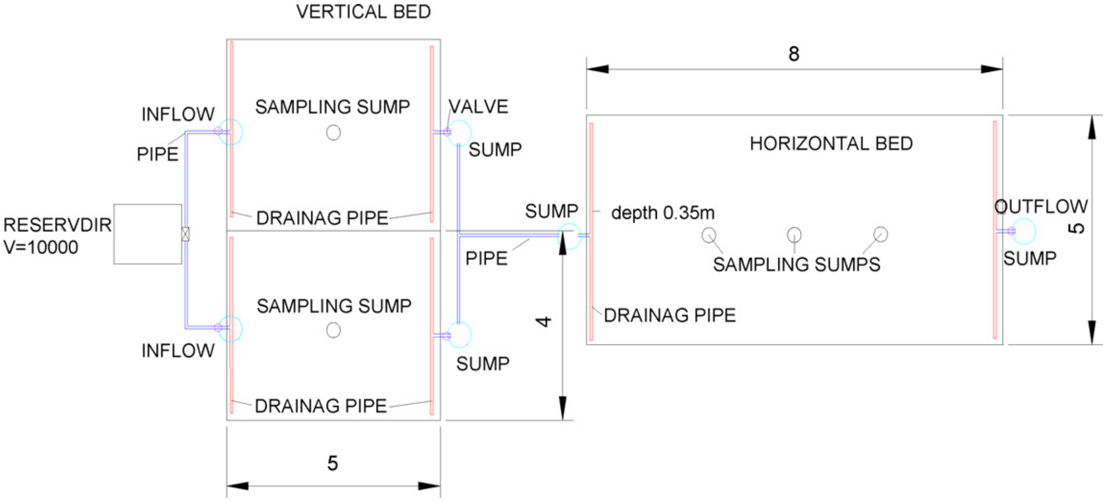

Fig. 2. Scheme of the pilot CW.

12 h with 0.07, 0.36 and 0.7 m/day alternately of wastewater with the flow rate of 1.44, 7.2, and 14.4 m^3/day. For the second 12 h an equal flow was led to the second VF bed. The outflow from the both beds was continually filling the HF bed. The same outflows were regulated with the valve in a collective sump at the outlet of the HF bed. The theoretical retention time at the flow 1.44 m^3/day was 8 days, at 7.2 m^3/day was 1.7 day and at 14.4 m^3/day was 0.8 day, and was calculated according to Kadlec and Knight [8]. The actual retention time was established using the salt tracer, based on the difference in the electric conductivity (EC), and it was 2, 0.7 and 0.4 day, respectively. Between each experiment the CW was rinsed two times with tab water and dried up for 1 day, in order to provide the comparable results.

2.2. Wastewater

2.2.1. CW model

Experiments were conducted on three synthetically prepared wastewaters differing in applied dyestuffs, auxiliaries, and chemicals (Table 1) in order to establish the colour reduction of structurally different dyestuffs and treatment efficiency of various dye-bath effluents during CW treatment. 150 l of synthetic wastewaters were prepared by dissolving dyes and other auxiliaries in a mixture of tap and distilled water in the ratio 2:3, with electrical conductivity of 452 μS/cm, thus simulating technologically water in a textile plant. The prepared wastewaters were maintained in a plastic tank for 24 h and, thereafter, flowed with constant velocity through the CW model.

2.2.2. Pilot scale CW

Treatments were carried out on real textile wastewater that were composed mainly reactive, and some vat and disperse dyestuffs, since it was set up near textile factory for cotton and cotton/PES processing.

2.3. Sample collection and analytical methods

2.3.1. CW model

Six treated samples (250 ml) for each dye-bath wastewater were taken from the outflow after 1, 2, 3, 4, 5, and 24 h, and directly analysed. Chemical oxygen demand was measured according to the ISO 6060 standard using a Termoproc TBGE. Total organic carbon was measured using a DC-190 Analyzer (Dohrmann) and pH was determined according to the ISO 10523 using MA 235 (Mettler Toledo). The absorbance of the dye-bath wastewaters before and after treatment was recorded at a wavelength of maximum absorption for each dye, according to standard EN ISO 105-Z10 using a Carry 50 UV–VIS spectrophotometer (Varian) with a special measuring probe (10 mm optical length), and a spectral absorption coefficient (SAC) was calculated. Prior to absorbance measurement, the effluents were centrifuged 10 min at 3000 rpm to prevent turbidity. During the experiments, the hydrodynamic conditions in the pipe-flow were constantly controlled, and the air temperature was measured.

2.3.2. Pilot scale CW

Assessment of the pilot CW's performance was evaluated with six experiments by means of various physical–chemical

Table 1
Three synthetically prepared dye-bath wastewaters for CW model

Dye-bath A	Dye-bath B	Dye-bath C
0.03 g/l C.I. Reactive Black 5 0.3 g/l Alvirol AGK (Textilcolor) 0.3 g/l Cibaflow PAD (Ciba) 2 g/l NaCl 2 ml/l NaOH (32.5%)	0.03 g/l C.I. Disperse Yellow 211 2 g/l Irgapadol MP (Ciba) 0.3 g/l Alvirol AGK (Textilcolor) 0.3 g/l Cibaflow PAD (Ciba)	0.03 g/l C.I. Vat Yellow 46 2 g/l Irgapadol MP (Ciba) 0.3 g/l Alvirol AGK (Textilcolor) 0.3 g/l Cibaflow PAD (Ciba)

parameters. Sampling was carried out at the inflow and at the outflow of CWs' separate bed. In each cycle, the samples were collected every 15 min during 1 h trial experiment, according to three previously determined flows and supplementary measured retention times. The hydraulic profile of the CW was evaluated using volumetric method for measuring the effluent flow rates. Dissolved oxygen (DO), temperature (*T*), electrical conductivity, and pH were measured in situ on each sampling site, using the WTW MultiLine P4 portable universal pocket-sized meter with the pH combined electrode, SenTix 41 integrated temperature probe, CellOx 325 dissolved oxygen probe and TetraCon 325 standard conductivity cell. The analyses of COD, BOD_5, TOC, N_{total}, N_{organic}, NH_4-N, sulphate, anionic surfactant, colour and TSS were performed by an independent laboratory according to the standard methods [12]. For qualitative information on the colour changes, the absorbance was measured across the entire visible spectrum (400–700 nm) with Carry 50 spectrophotometer (Varian), according to the standard EN ISO 105-Z10, and a spectral absorption coefficient (SAC) was calculated at a wavelengths of 436, 525 and 620 nm.

3. Results and discussion

3.1. CW model

The experiment was run from September to November 2004 with daily temperature above 20 °C and nightly temperature below 10 °C. The treatment efficiency of the CW model was established by measuring selected pollution parameters such as pH, absorbance, COD and TOC, and gained results are presented in Fig. 3. It is evident from Fig. 3 that pH decreased only in dye-bath A, which was highly alkaline with initial pH 12.1–9.4, depending on treatment time. In dye-baths B and C, where disperse and vat dyes were presents, pH values increased from 7.6 to 9.0 in dye-bath B and from 7.0 to 8.1 in dye-bath C, because of the bed media alkalinity. In addition, the best decolouration was achieved when treating the dye-bath A, which included reactive dye, i.e. the percentage of colour reduction was between 53% (after 1 h) and 70% (after 24 h), and the minimum decolouration was reached for dye-bath B (only 7% after 1 h and up to 34% after 5 h). It is generally believed that the filtration and adsorption played a major part in colour reduction on account of the specific condition employed in the present study (short trial time of 24 h). If extending the trial duration upon approximately 20 days with retention time of 8–10 h, we assume that various population of microorganisms would grow on the substrata's surface that could notably accelerate the pollutants' elimination mechanisms during wastewater treatment by means of anaerobic/aerobic activity. It is obvious from Fig. 3 that the initial COD and TOC values measured in synthetically wastewaters are very high, i.e. COD from 350 up to 600 mg/l (mass rate from 101 up to 173 g/day) and TOC from 120 up to 305 mg/l (mass rate from 35 up to 88 g/day); especially for dye-baths B and C, which included disperse or vat dyes and highly polluted auxiliary Irgapadol MP (anionic migration inhibitor that prevent side-to-centre migration and back-to-face effect in continuous dyeing; and is chemically on acid base). The highest COD reduction, up to 85%, was reached for dye-bath A and the retention time of 24 h. The same results were obtained by measuring the TOC. In the light of the fact that selected dyestuffs have a much lower TOC and COD values in comparison to the auxiliaries, TOC and COD reduction after treatment were verified as a result of prefer-

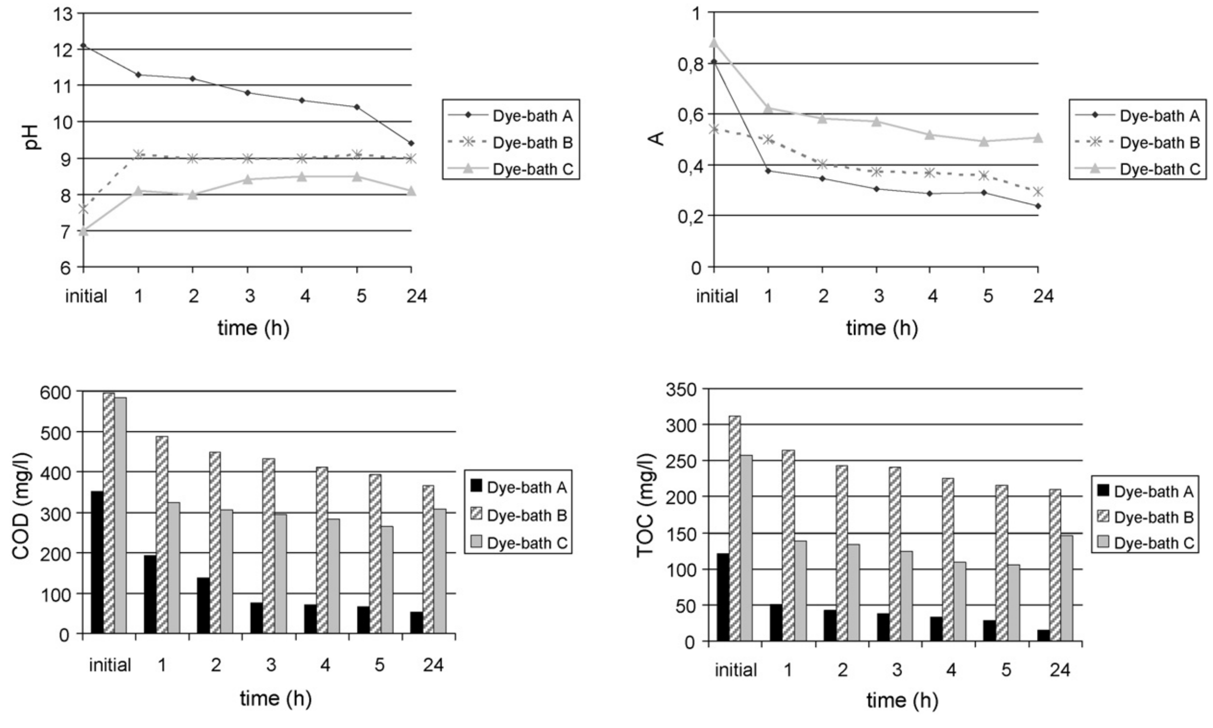

Fig. 3. pH, absorbance, COD and TOC before (initial) and after treatment on CW model.

Table 2
The range of pollution parameters monitored in real textile wastewater on the inflow and on the outflow of pilot CW

Parameter	COD (mg/l)	BOD (mg/l)	TOC (mg/l)	N_{tot} (mg/l)	N_{org} (mg/l)	NH_4-N (mg/l)	SO_4 (mg/l)	Anionic surfactant (mg/l)
Inflow range	276–1379	99–350	74–530	7–82	6–77	0.2–4.5	76–2200	1–10
Outflow range	122–487	43–95	15–50	10–19	1.5–8	7–16	75–251	0.5–3
Average values								
$Inflow_{total}$	771	198	261	29	26	2.4	1165	5.2
Outflow VF	235	110	73	21	10	11	284	2.7
Outflow HF	122	66	27	14	3.4	10	137	1.0
[a]*Limit values*	120	25	30	–	–	10	300	1
Average mass rate (g/day)								
$Inflow_{total}$	5596	1619	2074	297	265	26	9399	31
Outflow VF	2312	953	770	199	107	94	2976	187
Outflow HF	1030	514	262	118	32	86	58	6
Mass loads ($g/m^2/day$)								
$Inflow_{total}$	70	20	26	3.7	3.3	0.3	117	0.4
Inflow VF	140	40	52	7.4	6.6	0.6	235	0.7
Inflow HF	58	24	19	4.9	2.7	2.3	74	0.4

Parameter	SAC (436 nm)	SAC (525 nm)	SAC (620 nm)	TSS (mg/l)	DO (mg/l)	T (°C)	EC (μS/cm)	pH
Inflow range	5–34	9–100	4–16	27–408	1.2–1.7	30–38	2050–6430	8–9
Outflow range	0.1–4	0.8–4	0.4–1	7–11	0.2–2	12–25	1040–2260	7
Average values								
$Inflow_{total}$	18	31	7.9	129		33	3080	8.7
Outflow VF	4.5	3.6	1.6	25.2		19	1381	7.1
Outflow HF	2.2	2.0	0.8	9		18	1121	7.2
[a]*Limit values*	7	5	3	80	–	30	–	6.5–9

[a] Limit values of industrial wastewaters parameters for directly and indirectly discharge into water [18].

able auxiliary molecules' filtration and/or adsorption. Therefore, different time intervals had also a significant influence on treatment efficiency; the longer the time, the greater the effect of dye-bath wastewater treatment.

3.2. Pilot scale CW

The treatment efficiency of the pilot CW was examined by monitoring various pollution parameters in the initial and treated-wastewaters, and the obtained results are demonstrated in Table 2, and in Figs. 4–6. Table 2 summarizes the range and average of the inflow and the outflow values and the average mass rates and mass loads of the measured parameters. Fig. 4 demonstrates colour removal expressed as SAC values as the most relevant parameter in the experiment in VF and HF bed at different hydraulic loads. In Fig. 5, the percent of concentration reduction and percent of mass removal for monitored parameters are presented. Fig. 6 shows the efficiency as percent mass removal of VF and HF beds separately. The results showed that variable hydraulic load had a negligible impact on the treatment efficiency regarding the system conditions applied in the present research.

The outflow concentration of dissolved oxygen often dropped to 0.2 mg/l owing to the decomposition processes. The inflow temperatures were extremely high due to the exposed conditions of the retention basin. The electrical conductivity that depends upon the salt contents was decreased during wastewater treatment for 63% in average. The measurements in the sumps showed water stratification, proofed by the fact that the EC variation was measured only at the bottom, while the values within 15 cm under the water surface stayed almost unchanged. The pH monitoring of the inflow indicated that coloured textile wastewater was alkaline as expected regarding to the factory production, while the outflow values ranged around neutral pH. Evidently, the CW showed buffering capacity, lowering the pH of the inflow due to acids produced by microbial actions, as found-out also by Mbuligwe [9]. In addition, it is obviously that the CW had a significant performance advantage with respect to COD, which decreased by up to 84%, mostly owing to the filtration, sedimentation, and adsorption of various dyestuffs and auxiliaries. The COD reduction is similar to the results obtained by different authors, i.e. Baughman et al. [13] reported 20–34% efficiency for 50 mg/l COD inflow, while Winter and Kickuth [14] reached 65–76% efficiency for 1.400 mg/l COD inflow. Nevertheless, the ratio between BOD_5 and COD (0.26) in inflow indicated biologically hardly degraded nature of textile wastewater. Therefore, it was unrealistic to expect high BOD_5 reduction in treated effluents. The inflow and outflow values of TOC as well as the percent of treatment efficiency indicated that the adsorption of organic substances onto the pilot CW substrata was constant. The average removal efficiency for N_{total} was 52% and for $N_{organic}$ 87%, while it was negative for NH_4-N (−331%), presumably because of the oxygen lacking. It was also evident that during the months when the value of N_{total} in the inflow water was lower, its value increased slightly also at the outflow, mainly due to the NH_4-N. It was likely that NH_4-N was generated in

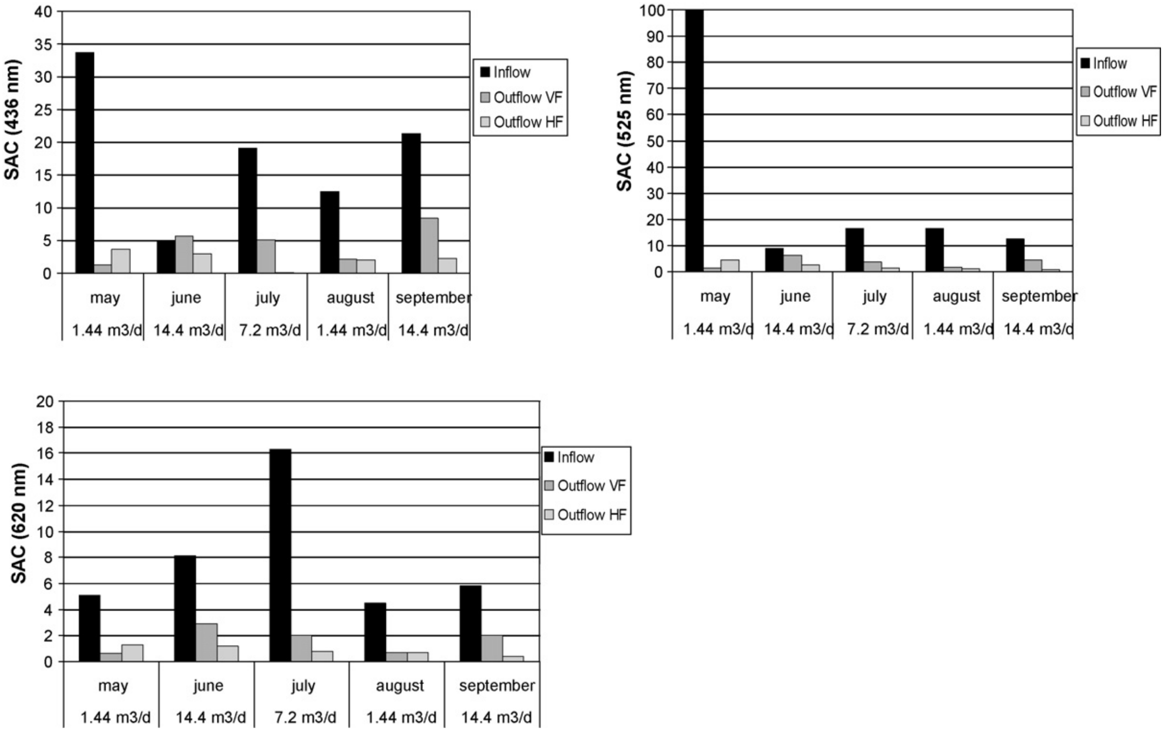

Fig. 4. The colour removal in VF and HF beds at different hydraulic loads.

beds and consequently rinsed from them. The average efficiency for sulphates at the outflow was 88%, although the influent concentration was very variable, most likely inducing variations in the effluent concentrations as well. Sulphate is removed via reduction to sulphide by sulphate reducing bacteria that can also cause the chemical decolourisation of dyestuffs [9]. The reduction of sulphate under anaerobic conditions produced hydrogen sulphide (H_2S) gas, which was characterised by the "rotten egg" smell, present most of the time. As the concentration of sulphates at the inflow was high, it probably accelerated the growth of sulphate reducing bacteria, which could play an important role in the mineralization of present organic substances. At the outflow, the reduction of anionic surfactants was on the average 80%. Del Buba et al. [15] stated higher concentrations of anionic surfactants for textile wastewater, and they noted that its removal in CW depended to a large extent on the quantity of suspended solids. Roughly the same picture was obtained also through our experiments. The CW proved potentially useful and beneficial method for wastewater decolouration. The average colour reduction in the final outflow at wavelengths of 436 nm was 88%, at 525 nm 93% and at 620 nm 89%. Decolouration efficiency was high measured across the entire visible spectra which indicated good retention of dye in the system. We should be aware that biotransformation results in a decaying of the dye molecule' chromophore and, consequently modifies the absorption characteristics of dyestuff [16]. For this reason, absorbance decreased while the visible colouring remained. Mbuligwe [9] and Davies et al. [17] reported 69–75% colour reduction, which is comparable with our results.

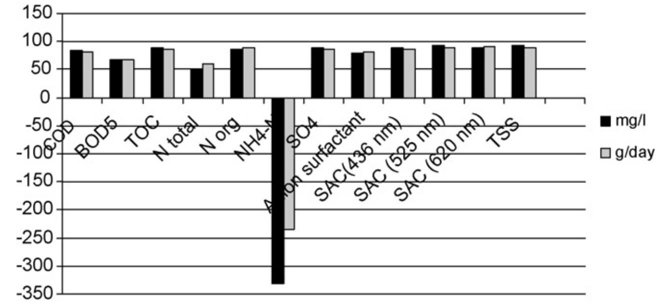

Fig. 5. The percent of concentration and mass removal for monitored parameters.

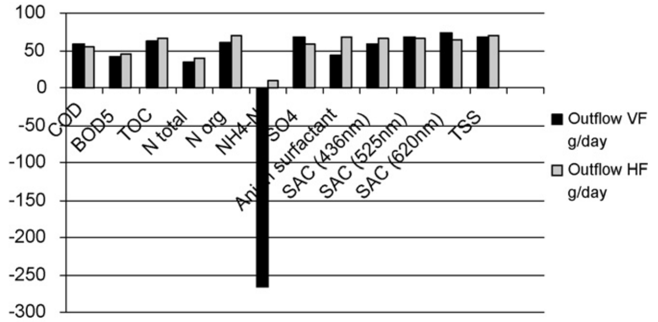

Fig. 6. The percent of mass removal in HF and VF bed.

4. Conclusions

The CW model and the pilot CW demonstrated its well capability to treat dye-rich textile wastewater. The obtained results indicate that the CW used in this study had significant effects on water quality parameters, specifically on colour reduction, i.e.

up to 70% in the CW model and up to 90% in the pilot CW. The model also appreciably lowered the COD and TOC values of all three dye-baths and decreased the pH in "reactive" dyebath, depending on the retention time. The pilot CW showed, beside superior decolourisation, high removal efficiency of COD (84%), TOC (89%), sulphate (88%), and anionic surfactants (80%). As expected, the reduction of sulphate produced the H_2S gas. The complete results of textile wastewater treatment using the CW system indicated that pollution parameters' reduction depending mostly on composition of textile wastewater, the chemical constitution of applied organic substances, and the design of the system, while the retention time had a minor effect. This study also suggests that hybrid system as a combination of VF and HF beds of the CW may have relative treatment efficiency advantages. The efficiencies expressed as a percent mass removal were similar in VF and HF bed, although mass loading rates on VF beds were at least two times higher than on HF bed. The efficiency of colour reduction was high by means of measuring an absorbance across entire visible spectra, which indicated good retention of dye in the system. The CW did not reach the limited values, allowed by Slovenian [18] and EU environmental regulations [19], only for BOD_5, and partially for anionic surfactants. The results have also proved that upgrading of the pilot CW with zeolitic tuff was not necessary. As such, CW is a very promising method and cost-effective option for coloured textile wastewater. Nevertheless, the limited duration of this study may have influenced the reported results, but on the other hand it could be a base for further experiments focused on long-term performance of CW.

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