IOP Conference Series: Materials Science and Engineering

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To cite this article: Amjad Hussien *et al* 2020 *IOP Conf. Ser.: Mater. Sci. Eng.* **671** 012089

IOP Conf. Series: Materials Science and Engineering **671** (2020) 012089 doi:10.1088/1757-899X/671/1/012089

Development of optimal location and design capacity of wastewater treatment plants for urban areas: a case study in Samawah city

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Abstract. Water, and related wastewater structures, are critical factors in the existence and the improvement of civilizations. Wastewater gathering and management has a considerable effect on the climate and economy at both regional and global level, and, accordingly, it is appropriate to advance actions that guarantee effective management for wastewater, particularly in urban areas. This research thus examined the environmental and economic aspects of proposed locations for wastewater treatment plants. Samawah city, located in the southern part of Iraq, was selected as a case study for the research methodology, and for research purposes, the studied city was divided into three main zones (1, 2, and 3) of sixteen areas. The Google Earth tool was used to calculate the lowest elevations in the studied zones in order to assess the suggested positions of treatment plants. Additionally, the WinQSB program was utilised to select the most appropriate positions for treatment plants based on data obtained from local government departments. These data include population, water consumption, and required lengths and subsequent cost of pipes. This research thus developed a new strategy for assigning the locations of wastewater treatment plants.

1. Introduction

In general, treatment of wastewater is essential to meet regional and national water standards and policy aims. The focal aim of wastewater treatment plants is to discharge effluent to the surrounding environment with the fewest harmful effects possible, preventing environment pollution by releasing only treated wastewater [1,2]. Wastewater management thus becomes an extremely important issue for city councils, particularly in rural areas where no drainage and sewage systems are provided.

Treatment plants have been established since the 19th century [3,4,5]. However, public health is still negatively affected by untreated sewage and artificial flows polluting rivers, marshes, and land [6,7,8]. As a result, sewage treatment plants have become even more necessary, and enforcement laws have been introduced to reduce pollution and to increase health and environmental safety levels. Environmental analysis and research in recent years has taken both social and physical impacts into consideration during the allocation of treatment plants. More specifically, researchers have studied the environmental effects of treatment plants' location on both humans and other organisms. Other side effects such as odours, noise, and architectural design factors have also been considered in such studies [9,10,11]. Therefore, selection of the best location for wastewater treatment plants must consider many essential aspects with respect to the environment and public health.

The focus of this research is determine the shortest pathway of wastewater to the treatment plant, generating minimum cost. This is achieved by addressing the following targets: (i) selecting the appropriate place for a wastewater treatment plant; (ii) selecting the most effective cost plan among many potential alternatives; and (iii) balancing the required design capacity of the wastewater treatment plant and the governed area. Details of the data collection method and research analysis tools are described in the following sections. Linear goal programming using the WinQSB tool was applied in order to determine the most appropriate location for the treatment plant [12], as this provides mathematical models that are useful in making decisions with regard to engineering research management. This research can thus be applied and used as a helpful guide for engineers planning and designing wastewater treatment plants within urban areas.

2. Methodology

2.1 Study Area Description

Samawah is the contemporary capital of the Al-Muthanna Governorate. The city is located midway between Basra and Baghdad at Latitude 31.309 and Longitude 45.280, as shown in figure 1. The map of Iraq used in this study was downloaded from the Global Administrative Areas Database (GADM) [13], which is a spatial database of the world's administrative areas' locations for use in geographic information system (GIS). The GADM provides multiple attributes, including the name, address and elevation of each area.

To achieve the main aims of this research, Samawah was chosen for the application of the research methodology. The city was divided into three main zones (1, 2 and 3) featuring 16 areas, as shown in figure 2. Each zone included one treatment plant to serve multiple areas that was located in the minimum elevation of the zone. The zones were identified with aid of the Google Earth tool (GET), a program that renders a three dimensional (3D) representation of the earth from satellite imagery [14].

2.2 Data Collection

The main focus of this research is the disposal of wastewater, with this being moved from the populated areas to the treatment plant before extrusion into the Euphrates River.

The population figures considered in this study were based on data from the Samawah Census Office. The treatment plants (1, 2, and 3) were then located at the minimum elevations in each of the three city zones, as illustrated in figure 2. The elevations of the 16 areas were obtained using the Google Earth tool [14].

Sewage was deemed to stream from higher to lower levels in each area by means of pipelines with 1,200 mm diameters. The cost of one metre of pipeline is thus assessed as US$175 at local market prices. The average daily water consumption per capita was estimated to be 0.25 m^3. Hence, the average daily sewage flow was obtained by multiplying the number of people living in the area by the daily water consumption per capita [9]. Finally, the maximum design capacity of each treatment plant (45,360 m^3 per day) was assigned by the Al-Muthanna Sewage Department.

Table 1 summarises the studied areas with their corresponding elevations, major pipe details including length and cost, population, and water consumption per day.

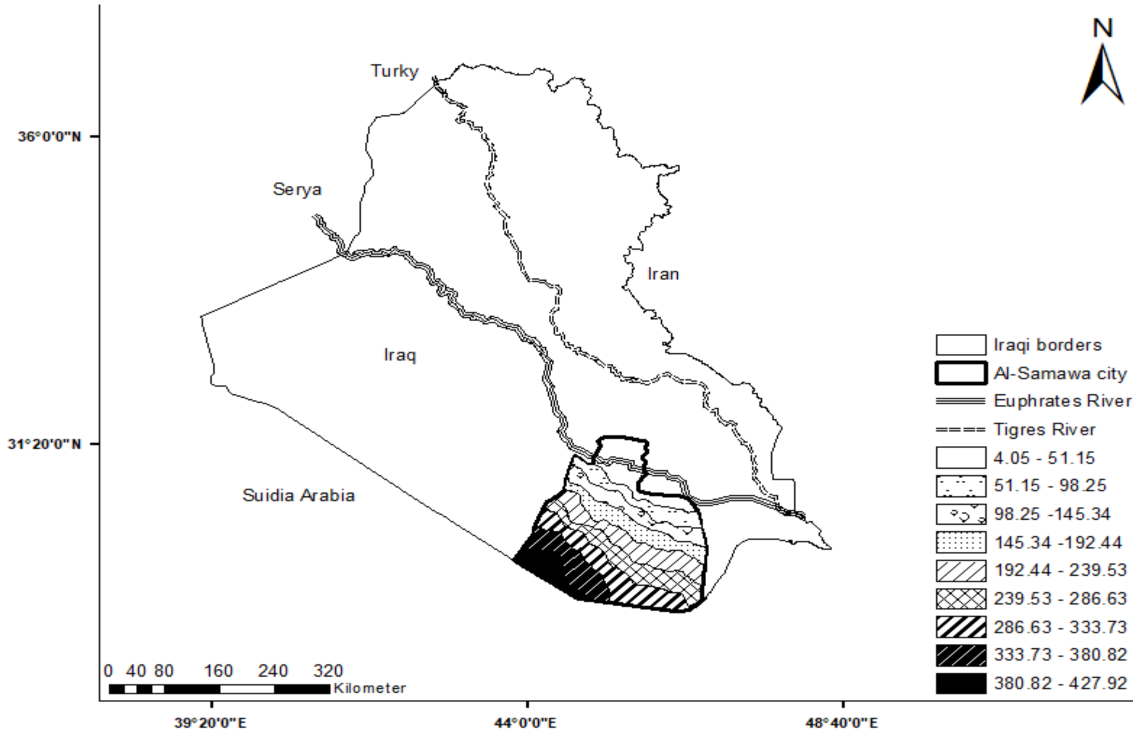

Figure 1. Location of the studied city with the corresponding elevations.

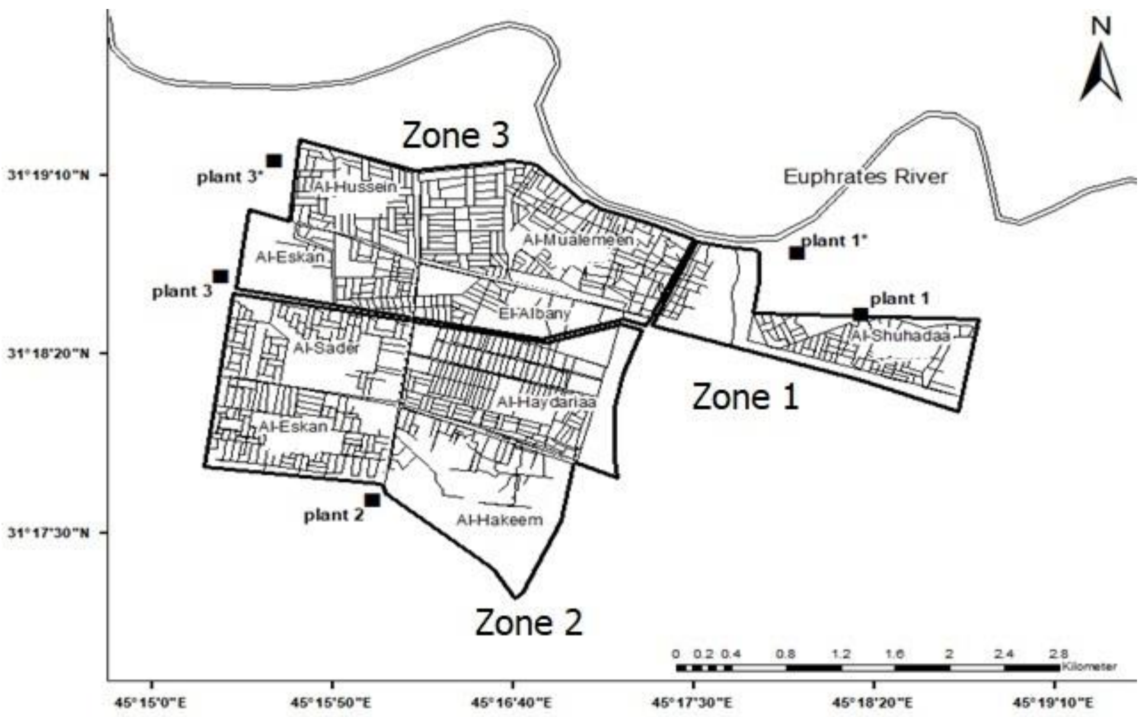

Figure 2. The selected zones (1, 2 and 3) with the corresponding areas and the wastewater treatment plant locations in Samawah.

Table 1. Samawah areas involved in this study. including elevation above the sea level, lengths and cost of pipes, population, and water consumption.

City zone	Area	Elevation (m)	Length of major pipe[a] (m)	Cost of major pipe[a] (USD)	Number of population[b]	Water consumption[a] (m^3 per day)
1	Al-Shuhada	17.38	192	33,600	15,308	3,827
1	Al-Sharqi	17.68	947	165,725	11,800	2,950
2	270 Dar	11.28	150	26,250	17,140	4,285
2	Al-Hakeem	12.20	1,231	215,425	26,468	6,617
2	Al-Askari	13.11	802	140,350	19,350	4,837
2	Al-Jumhori	13.72	50	8,750	23,752	5,938
2	Al-Sader	14.02	1,604	280,700	13,974	3,493
2	Al-Haydaria	14.33	100	17,500	23,350	5,882
2	Al-Shurtah	14.63	75	13,125	23,794	5,948
3	Al-Hussain	12.50	1,550	271,250	29,350	7,337
3	Al-Eskan	12.80	200	35,000	23,794	5,993
3	Al-Bani	13.11	1,400	245,000	17,530	4,382
3	Al-Jaddan	13.41	100	175,000	17,538	4,384
3	Al-Mualmeen	14.02	700	122,500	23,752	5,938
3	Al-Dhubat	14.94	150	26,250	21,384	5,346
3	Al-Gharbi	15.55	1,500	262,500	19,796	4,949
Total			10,751	2,038,925	328,080	82,106

[a] Data were obtained from Al-Muthanna Sewage Department.

[b] Data were obtained from Samawah Census Office.

2.3 The Goal Programming Model

The WinQSB Program utilises Linear Goal Programming (GP) and Integer Linear Goal Programming (IGP) [15,16,17,18,19]. GP and IGP models involve one or more linear goals (objective functions) and a limited number of linear constraints. Decision variables may also be bounded with specific values. All decision variables are considered to be continuous numbers [20,21,22]. The general equation of minimisation linear goal programming thus follows the formula [23]

$$Minimise\ Z\ =\ \left(\sum_{j=1}^{n} C_{ij}\, X_j +\ \cdots\ + C_{in}\, X_n\right) +\ n_i - p_i\ = b_i \qquad (Goal\ level\ i) \tag{1}$$

where Z is the objective function, X_j is a variable, C_{ij} is the coefficient of variable, b_i is the level of priority of goal i, n_i is the degree of the minimum achievement of goal, and p_i is the degree of the maximum achievement of goal. Since p_i and n_i cannot be added together, one or both of them must equal zero [24]. Hence, $p_i \times n_i = 0$.

To solve this problem here, the variable (X_{ijk}) was defined as the quantity of sewage from area i sent to treatment plant j in city zone k.

The final equation was formulated according to the goals prepared by the Al-Muthanna Sewage Office, using the collected data shown in table 1 with reference to the suggested locations of treatment plants. The collected data were input into the program to achieve the goals as follows:

2.3.1 Goal One

This had 16 constraints, each of which represented the quantity of sewage (m^3 per day) from each area to be disposed of to treatment plants, as shown in the following equations set from (*a*) to (*p*):

$$\left.\begin{cases} X_{111} + n_1 - p_1 = 3{,}827 & (a) \\ X_{211} + n_2 - p_2 = 2{,}950 & (b) \\ X_{122} + n_3 - p_3 = 4{,}285 & (c) \\ X_{222} + n_4 - p_4 = 6{,}617 & (d) \\ X_{322} + n_5 - p_5 = 4{,}837 & (e) \\ X_{422} + n_6 - p_6 = 5{,}938 & (f) \\ X_{522} + n_7 - p_7 = 3{,}493 & (g) \\ X_{622} + n_8 - p_8 = 5{,}882 & (h) \\ X_{722} + n_9 - p_9 = 5{,}948 & (i) \\ X_{133} + n_{10} - p_{10} = 7{,}337 & (j) \\ X_{233} + n_{11} - p_{11} = 5{,}993 & (k) \\ X_{333} + n_{12} - p_{12} = 4{,}382 & (l) \\ X_{433} + n_{13} - p_{13} = 4{,}384 & (m) \\ X_{533} + n_{14} - p_{14} = 5{,}938 & (n) \\ X_{633} + n_{15} - p_{15} = 5{,}346 & (o) \\ X_{733} + n_{16} - p_{16} = 4{,}949 & (p) \end{cases}\right\} \quad \textit{Goal (1)}$$

2.3.2 Goal Two

This included three constraints on the proposed plants in order to achieve the maximum design capacity of 45,360 m^3 per day. Sewage collected from the studied areas and disposed of into the treatment plants at each zone was thus presented in the following set of equations from (*a*) to (*c*):

$$\left.\begin{cases} X_{111} + X_{211} + n_{17} - p_{17} = 45{,}360 & (a) \\ X_{122} + X_{222} + X_{322} + X_{422} + X_{522} + X_{622} + X_{722} + n_{18} - p_{18} = 45{,}360 & (b) \\ X_{133} + X_{233} + X_{333} + X_{433} + X_{533} + X_{633} + X_{733} + n_{19} - p_{19} = 45{,}360 & (c) \end{cases}\right\} \quad \textit{Goal (2)}$$

2.3.3 Goal Three

This included three constraints representing the shortest sewage disposal network from the served areas to the treatment plants at each zone, as illustrated in equations from (*a*) to (*c*):

$$\left.\begin{cases} 192\,X_{111} + 947\,X_{211} + n_{20} - p_{20} = 0 & (a) \\ 150\,X_{122} + 1{,}231\,X_{222} + 802\,X_{322} + 50\,X_{422} + \\ 1{,}604\,X_{522} + 100\,X_{622} + 75\,X_{722} + n_{21} - p_{21} = 0 & (b) \\ 1{,}550\,X_{133} + 200\,X_{233} + 1{,}400\,X_{333} + 100\,X_{433} + \\ 700\,X_{533} + 150\,X_{633} + 1{,}500\,X_{733} + n_{22} - p_{22} = 0 & (c) \end{cases}\right\} \quad \textit{Goal (3)}$$

2.3.4 Goal Four

This had a single constraint representing the cost of sewage disposal from the corresponding areas into the treatment plants, as shown in the following equation:

$$\left.\begin{cases} 33{,}600\,X_{111} + 165{,}725\,X_{211} + 26{,}250\,X_{122} + 215{,}425\,X_{222} + 140{,}350\,X_{322} + \\ 8{,}750\,X_{422} + 280{,}700\,X_{522} + 17{,}500\,X_{622} + 13{,}125\,X_{722} + 27{,}125\,X_{133} + \\ 35{,}000\,X_{233} + 245{,}000\,X_{333} + 17{,}500\,X_{433} + 122{,}500\,X_{533} + 26{,}250\,X_{633} + \\ 262{,}500\,X_{733} + n_{23} + p_{23} = 0 \end{cases}\right\} \quad \textit{Goal (4)}$$

3. Solutions, Results, and Discussion

The optimum solution program was applied in order to select the best locations for the sewage treatment plants taking into consideration the outlined environmental engineering requirements. The results of the statistical and engineering analysis were based on two main considerations, the first being to select the most appropriate location for the treatment plant, and the second being to select the most economical solution from among several potential alternatives.

It was found that the locations of the sewage treatment plants near populated areas required more than one solution to be generated to determine suitable positions for these plants. It was thus necessary to determine the optimum solution based on the outputs of the WinQSB program. A brief description of these solutions is presented in the next sections.

3.1 First Solution

In order to achieve the goals outlined, the input data of suggested plants 1, 2, and 3 (see figure 2) were considered. After running the program, results were obtained as presented in table 2.

Table 2. Results for the 1st solution for treatment plants 1, 2 and 3.

Variable X_{ijk}	Value	Variable n_i	Value	Variable p_i	Value
X_{111}	42,410	n_1	0	p_1	38,583
X_{211}	2,950	n_2	0	p_2	0
X_{122}	4,285	n_3	0	p_3	0
X_{222}	6,617	n_4	0	p_4	0
X_{322}	4,837	n_5	0	p_5	0
X_{422}	5,938	n_6	0	p_6	0
X_{522}	3,493	n_7	0	p_7	0
X_{622}	5,882	n_8	0	p_8	0
X_{722}	5,948	n_9	0	p_9	0
X_{133}	7,337	n_{10}	0	p_{10}	0
X_{233}	5,993	n_{11}	0	p_{11}	0
X_{333}	4,382	n_{12}	0	p_{12}	0
X_{433}	4,384	n_{13}	0	p_{13}	0
X_{533}	5,938	n_{14}	0	p_{14}	0
X_{633}	5,346	n_{15}	0	p_{15}	0
X_{733}	4,949	n_{16}	0	p_{16}	0
		n_{17}	0	p_{17}	0
		n_{18}	8,360	p_{18}	0
		n_{19}	7,031	p_{19}	0
		n_{20}	0	p_{20}	10,936,370
		n_{21}	0	p_{21}	19,601,524
		n_{22}	0	p_{22}	31,526,150
		n_{23}	0	p_{23}	10,861,207,552

Goal one: The obtained value is zero. This represents the total quantity of sewage taken from all areas and disposed into the treatment plants in each city zone.

Goal two: After running the program, the resulting value was 15,391 m^3 per day. In fact, this value obtained from the WinQSB program was related to the items of this goal ($n_{18} + n_{19} = 8{,}360 + 7{,}031 =$

15,391). The three treatment plants could thus possibly have larger design capacities later. The value of 15,391 m^3 per day represents the additional quantity of sewage flow that should be considered in the design capacity of the treatment plants in the future.

Goal three: Its value is 62,064,044 m.m^3 per day. This value is obtained from running the WinQSB program related to the items of this goal ($p_{20} + p_{21} + p_{22} = 10{,}936{,}370 + 19{,}601{,}524 + 31{,}526{,}150 = 62{,}064{,}044$). The value of 62,064,044 is obtained by multiplying of the total length of pipeline by the quantity of sewage flow, about 15,391 m^3 per day, in the future. Therefore, the length of the pipelines used to dispose of the sewage can be obtained by dividing this value by the quantity of sewage flow. The result is 4,032 m which represents the possible length of pipelines that should be considered in the design capacity of the three treatment plants in the future.

Goal four: The suggested value is 10,861,207,552 \$.m^3 per day. This was obtained from the WinQSB program in relation to the items of this goal ($p_{23} = 10{,}861{,}207{,}552$). The value of 10,861,207,552 is equal to the cost of sewage disposal of about 15,391 m^3 per day in the future. Therefore, the cost to dispose the sewage can be obtained by dividing the mentioned value by the quantity of sewage flow. The resulting figure, US$705,685 should thus be considered in the design capacity of those three treatment plants as well as sewage networks in the future.

3.2 Second Solution

In this solution, the input data of proposed plants 1*, 2, and 3 were considered. Treatment plant 1 was moved to a new location, 1*, while other plants, 2 and 3, were fixed as illustrated in figure 2. Subsequently, the cost and lengths of the pipelines connected to those plants varied. The new statements were input as items and the WinQSB program re-run to obtain results as shown in table 3.

Goal one: The obtained value was zero. This represents the total quantities of sewage taken from the areas and disposed of into the treatment plants of the city zones.

Goal two: After running the program, the resulting value was 15,391 m^3 per day. This value is related to the items of this goal ($n_{18} + n_{19} = 8{,}360 + 7{,}031 = 15{,}391$).

Goal three: This value was 74,070,708 m.m^3 per day. This value is obtained from the items of this goal ($p_{20} + p_{21} + p_{22} = 22{,}943{,}056 + 19{,}601{,}524 + 31{,}526{,}150 = 74{,}070{,}730$). The value of 74,070,730 results from multiplying of the total length of pipeline by the quantity of sewage flow, which is about 15,391 m^3 per day. Therefore, the length of the pipelines used to dispose of sewage can be obtained by dividing the value of ($p_{20} + p_{21} + p_{22}$) by the quantity of sewage flow. The result, 4,812 m, represents the possible length of pipeline that should be considered in the design capacity of the three treatment plants in the future.

Goal four: This value is 12,962,377,728 \$.m^3 per day, obtained from the items of this goal ($p_{23} = 12{,}962{,}377{,}728$). The value of 12,962,377,728 is equal to the cost of sewage disposal of about 15,391 m^3 per day in the future. Therefore, the cost of disposing of the sewage can be obtained by dividing the mentioned value by the quantity of sewage flow. The result is US$842,205, which should be considered as part of the design capacity of the three treatment plants and sewage network in the future.

Table 3. The results for the 2nd solution for treatment plants 1*, 2 and 3.

Variable X_{ijk}	Value	Variable n_i	Value	Variable p_i	Value
X_{111}	3,827	n_1	0	p_1	0
X_{211}	41,533	n_2	0	p_2	38,583
X_{122}	4,285	n_3	0	p_3	0
X_{222}	6,617	n_4	0	p_4	0
X_{322}	4,837	n_5	0	p_5	0
X_{422}	5,938	n_6	0	p_6	0
X_{522}	3,493	n_7	0	p_7	0
X_{622}	5,882	n_8	0	p_8	0
X_{722}	5,948	n_9	0	p_9	0
X_{133}	7,337	n_{10}	0	p_{10}	0
X_{233}	5,993	n_{11}	0	p_{11}	0
X_{333}	4,382	n_{12}	0	p_{12}	0
X_{433}	4,384	n_{13}	0	p_{13}	0
X_{533}	5,938	n_{14}	0	p_{14}	0
X_{633}	5,346	n_{15}	0	p_{15}	0
X_{733}	4,949	n_{16}	0	p_{16}	0
		n_{17}	0	p_{17}	0
		n_{18}	8,360	p_{18}	0
		n_{19}	7,031	p_{19}	0
		n_{20}	0	p_{20}	22,943,056
		n_{21}	0	p_{21}	19,601,524
		n_{22}	0	p_{22}	31,526,150
		n_{23}	0	p_{23}	12,962,377,728

3.3 Third Solution

In this solution, the input data for suggested plants 1, 2 and 3* were considered. Treatment plant 3 was moved to a new location, 3*, while other plants 1 and 2 were fixed, as shown in figure 2. Subsequently, the cost and lengths of the pipelines connected to those plants varied. Similarly, new statements were input as items and the WinQSB program re-run to obtain results as presented in table 4.

Goal one: the given value is zero. This represents the total quantities of sewage taken from the areas and disposed of to the treatment plants in the city zones.

Goal two: The obtained value was 15,391 m^3 per day. This value is related to the items of this goal ($n_{18} + n_{19}$ = 8360 + 7,031 = 15,391).

Goal three: The value was 65,844,328m.m^3 per day, obtained from ($p_{20} + p_{21} + p_{22}$ = 10,936,370 + 23,381,810 + 31,526,150= 65,844,330). The 65,844,330 figure is the result of multiplying the total length of pipeline by the quantity of sewage flow, which is about 15,391 m^3 per day. Therefore, the length of the pipelines used to dispose of the sewage can be obtained by dividing the value of ($p_{20} + p_{21} + p_{22}$) by the quantity of sewage flow. The result is 4,278 m, which represents the possible length of pipelines that should be included in the design capacity of the three treatment plants in the future.

Goal four: This value is 11,522,757,632 \$.m^3 per day, obtained from the items of this goal (p_{23} = 11,522,757,632). Fundamentally, the value of 11,522,757,632 is equal to the cost of sewage disposal of about 15,391 m^3 per day in the future. Therefore, the cost of disposing of the sewage can be obtained by dividing the value of p_{23} by the quantity of sewage flow. The result is US$748,668, which should be considered in the design capacity of treatment plants and sewage networks in the future.

Table 4. Results of the 3rd solution for treatment plants 1, 2 and 3*.

Variable X_{ijk}	Value	Variable n_i	Value	Variable p_i	Value
X_{111}	42,410	n_1	0	p_1	38,583
X_{211}	2,950	n_2	0	p_2	0
X_{122}	4,285	n_3	0	p_3	0
X_{222}	6,617	n_4	0	p_4	0
X_{322}	4,837	n_5	0	p_5	0
X_{422}	5,938	n_6	0	p_6	0
X_{522}	3,493	n_7	0	p_7	0
X_{622}	5,882	n_8	0	p_8	0
X_{722}	5,948	n_9	0	p_9	0
X_{133}	7,337	n_{10}	0	p_{10}	0
X_{233}	5,993	n_{11}	0	p_{11}	0
X_{333}	4,382	n_{12}	0	p_{12}	0
X_{433}	4,384	n_{13}	0	p_{13}	0
X_{533}	5,938	n_{14}	0	p_{14}	0
X_{633}	5,346	n_{15}	0	p_{15}	0
X_{733}	4,949	n_{16}	0	p_{16}	0
		n_{17}	0	p_{17}	0
		n_{18}	8,360	p_{18}	0
		n_{19}	7,031	p_{19}	0
		n_{20}	0	p_{20}	10,936,370
		n_{21}	0	p_{21}	23,381,810
		n_{22}	0	p_{22}	31,526,150
		n_{23}	0	p_{23}	11,522,757,632

3.4 Fourth Solution

In this solution, the input data for proposed plants 1*, 2 and 3* were considered. Treatment plants 1 and 3 were moved to alternative locations 1* and 3* (see figure 2) while plant 2 was fixed. Subsequently, the cost and lengths of the pipelines connected to those plants varied. The new statements were input as items and the WinQSB program re-run in order to obtain results as given in table 5.

Goal one: the obtained value is zero. This value represents the total quantities of sewage taken from the areas and disposed of to the treatment plants of the city zones.

Goal two: The resulting value is 15,391 m^3 per day, obtained in relation to the items of this goal ($n_{18} + n_{19}$ = 8,360 + 7,031 = 15,361).

Goal three: This value is 77,851,016 m.m^3/day, related to the items of this goal ($p_{20} + p_{21} + p_{22}$ = 22,943,056 + 23,381,810 + 31,526,150 = 77,851,016). The value of 77,851,016 results from multiplying the total length of pipeline by the quantity of sewage flow, which is about 15,391 m^3 per day in the future. Therefore, the length of the pipelines used to dispose of the sewage can be obtained by dividing the value above by the quantity of sewage flow. The result is 5,058 m, which represents the possible length of pipelines that should be included in the design capacity of the three treatment plants in the future.

Goal four: This value is 13,623,927,808 \$.m^{3}per day, found in relation to the items of this goal (p_{23} = 13,623,927,808). Accordingly, the value of 13,623,927,808 is equal to the cost of sewage disposal of about 15,391 m^3 per day. Therefore, the cost of disposing of the sewage can be obtained by dividing the value of p_{23} by the quantity of sewage flow. The result is US\$885,188, which should be considered in the calculation of design capacity of treatment plants and sewage network in the future.

Table 5. Results of the 4th solution for treatment plants 1*, 2 and 3*.

Variable X_{ijk}	Value	Variable n_i	Value	Variable p_i	Value
X_{111}	3,827	n_1	0	p_1	0
X_{211}	41,533	n_2	0	p_2	38,583
X_{122}	4,285	n_3	0	p_3	0
X_{222}	6,617	n_4	0	p_4	0
X_{322}	4,837	n_5	0	p_5	0
X_{422}	5,938	n_6	0	p_6	0
X_{522}	3,493	n_7	0	p_7	0
X_{622}	5,882	n_8	0	p_8	0
X_{722}	5,948	n_9	0	p_9	0
X_{133}	7,337	n_{10}	0	p_{10}	0
X_{233}	5,993	n_{11}	0	p_{11}	0
X_{333}	4,382	n_{12}	0	p_{12}	0
X_{433}	4,384	n_{13}	0	p_{13}	0
X_{533}	5,938	n_{14}	0	p_{14}	0
X_{633}	5,346	n_{15}	0	p_{15}	0
X_{733}	4,949	n_{16}	0	p_{16}	0
		n_{17}	0	p_{17}	0
		n_{18}	8,360	p_{18}	0
		n_{19}	7,031	p_{19}	0
		n_{20}	0	p_{20}	22,943,056
		n_{21}	0	p_{21}	23,381,810
		n_{22}	0	p_{22}	31,526,150
		n_{23}	0	p_{23}	13,623,927,808

4. Conclusions and Recommendations for Future Research

Based on the outputs of the WinQSB program, the best solution to assigning the locations of treatment plants is the fourth solution. This, however, suggests that the sewage treatment plants might need longer pipeline lengths (approximately 5,058 m) and increased costs (approximately US$885,188) in order to take expansion of the sewage network in the future into account.

Despite the fact that sewage projects and treatment plants have with high costs, they have multiple positive effects on the national economy and environmental safety. Purification techniques may reduce the risk of several diseases. However, these projects do have other implications for the environment, and more attention should be paid to these. In addition, wastewater treatment plants require good maintenance, management, and good engineering staff in order to achieve the best performance, adding to their ongoing costs.

5. Acknowledgements

The authors acknowledge the support of their respective institutions. Deep gratitude and sincere thanks are offered to Al-Muthanna Sewage Department and Samawa Census Office for providing the required data. This research did not receive any specific grants from funding agencies in the public, commercial, or not-for-profit sectors.

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Ecological Engineering 101 (2017) 28–38

Contents lists available at ScienceDirect

Ecological Engineering

journal homepage: www.elsevier.com/locate/ecoleng

Full research paper

Dye wastewater treatment by vertical-flow constructed wetlands

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A R T I C L E I N F O

Article history:
Received 9 June 2016
Received in revised form 6 January 2017
Accepted 10 January 2017

Keywords:
Acid Blue 113
Basic Red 46
Chemical oxygen demand
Phragmites australis
Reed bed filter
Textile wastewater

A B S T R A C T

Wetlands have long played an important role as natural purification systems. Textile industry processes are among the most environmentally unsustainable industrial processes, because they produce coloured effluents in large quantities polluting water resources. In this study, two different azo dyes (Acid Blue 113 (AB113) and Basic Red 46 (BR46)) have been fed as part of synthetic wastewater recipes to a laboratory-scale vertical-flow construction wetland set-up comprising wetlands with gravel media as controls and wetlands planted with *Phragmites australis* (Cav.) Trin. ex Steud. (Common Reed) for each dye. Two different concentrations (7 mg/l and 215 mg/l) were used for each dye at two different hydraulic retention times (48 h and 96 h). According to results for the low concentration of BR46, there is no significant ($p > 0.05$) difference between wetlands (unplanted and planted) in terms of dye removal. The use of plants concerning the short contact time scenario for ammonia-nitrogen (NH_4-N) and a low concentration of AB113 is linked to good removal. In case of low dye concentrations, the presence of plants for the long contact time scenario impacted significantly ($p < 0.05$) positive on the removal efficiencies of nutrients. For chemical oxygen demand (COD), the removal percentages were 50%, 59% and 67% for the control and for the wetlands with short and long retention times, respectively. All reductions were statistically significant ($p < 0.05$). For the high concentration of BR46, the removal percentages for this dye and COD were 94% and 82%, and 89% and 74% for the long and short retention times, respectively. For the low concentration of AB113, the percentage corresponding removals for the dye were 71%, 68% and 80%. The COD removals were 4%, 7% and 15% for the control, and the short and long retention times, respectively. Finally, for the high concentration of AB113, the percentage removals for the dye and COD were 71% and 73%, and 50% and 52% for the 48-h and 96-h retention times in this order.

1. Introduction

Azo dyes are important colouring agents in the textile, food and pharmaceutical industries (Tee et al., 2015; Yaseen and Scholz, 2016), and are linked to a relatively high toxicity, mutagenicity and carcinogenicity. Azo dyes and their corresponding breakdown products are difficult to treat in traditional wastewater treatment systems according to Erkurt (2010). Many technological solutions such as coagulation-flocculation and advanced oxidation process (Sivakumar et al., 2013), which have been applied to treat dyes, are not feasible in practice, because of too high costs and complex processes involved. Furthermore, some developing countries such as China and Bangladesh (Chen et al., 2007; Islam et al., 2011), which have a strong textile industry, but unreliable energy sources, may benefit from less-energy demanding methods of wastewater treatment.

Textile wastewater causes considerable environmental pollution. The main challenges are high concentrations of organic matter and particular persistent colorants (dyes) that have to be resistant to the effects of sweat, soap, water, light and oxidants (Olejnik and Wojciechowski, 2012). The azo dyes AB113 and BR46 are, therefore, widely used in the textile industry (Pervez et al., 2000; Olgun and Atar, 2009; Deniz and Karaman, 2011; Yaseen and Scholz, 2016). Typically, textile industry-processing effluents contain dyes in the range between 10 and 200 mg/l (Yassen and Scholz, 2016). Most textile dyes at a rather low concentration of even >1 mg/l can be detected by the human eye (Pandey et al., 2007).

Abbreviations: AB, acid blue; ANOVA, analysis of variance; AO, acid orange; AY, acid yellow; BR, basic red; CASRN, chemical abstracts survey registry number; COD, chemical oxygen demand; DO, dissolved oxygen; DY, disperse yellow; EC, electric conductivity; HF, horizontal-flow; N, nitrogen; NH_4-N, ammonia-nitrogen; n/a, not applicable; NO_2-N, nitrite-nitrogen; NO_3-N, nitrate-nitrogen; PO_4–P, orthophosphate-phosphorus; RB, reactive black; SD, standard deviation; SE, standard error; TDS, total dissolved solids; T-N, total nitrogen; TOC, total organic carbon; T-P, total phosphorus; TSS, total suspended solids; VF, vertical-flow; VY, vat yellow; Λ_{max}, maximum absorbance.

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http://dx.doi.org/10.1016/j.ecoleng.2017.01.016
0925-8574/

Vertical-flow constructed wetlands are engineered ecosystems designed to remove pollutants from wastewater (Kadlec and Wallace, 2009). These systems mimic the treatment that occurs in natural wetlands by relying on heterotrophic microorganisms, aquatic plants and a combination of naturally occurring processes (Scholz, 2015).

Researchers previously investigated the performance of ponds (Yaseen and Scholz, 2016) and, in particular, constructed wetlands to treat textile wastewater (Table 1). However, results rarely cover all seasons (Vymazal, 2014). Pervez et al. (2000) used aerated vertical-flow wetlands to remove two textile Azo dyes (RB171 and AB113) from synthetic wastewater for a period of 70 days. The percentage removal of 98% was high for both dyes. However, the results did not assess the removal of COD, phosphorus and nitrogen.

Davies et al. (2006) used vertical-flow wetlands for aerobic degradation of Acid Orange 7 (AO7) in a short-term study (Table 1). The results showed colour and COD removals of 99% and 93%, respectively. However, phosphorus and nitrogen removal were not recorded.

Yalcuk and Dogdu (2014) used vertical-flow constructed wetlands to treat Acid Yellow 2G E107 Dye-containing wastewater (Table 1). The constructed wetland consisted of three vertical wetland filters, which were filled with fine gravel, sand and zeolite. One wetland was kept unplanted as a control and the other two were planted with *Canna idica* L. and *Typha angustifolia* L., respectively. The period of operation was only three months. The results showed that the average colour removal percentages were 87% for the control and 98% for the other wetlands. For NH_4-N, the average percentages removals were 43%, 61% and 46% for the control, *C. idica* and *T. angustifolia*, respectively. The average percentages of ortho-phosphate-phosphorus (PO_4-P) for the control, *C. indica* and *T. angustifolia* were 84%, 87% and 88% respectively. Only 90 days was the time of operation.

The presence of *Phragmites australis* in vertical-flow constructed wetlands has a significant impact on the removal of organic matters, aromatic amines and NH_4-N (Ong et al., 2011). The growth cycle of *P. australis* traditionally completes between May and September in Britain (Haslam, 1972). Ferreira et al. (2014) detected normal growth for *Phragmites* with absence of toxic signs or depletion of leaf nitrogen content, when they used vertical-flow wetlands to treat an effluent comprising Diazo dye (DR81).

The aim of this study is to assess the efficiency of vertical-flow constructed wetlands in removing dye, COD, PO_4-P and other nutrients. The corresponding objectives are to assess (a) the role of *P. australis* on dye removal; (b) the influence of two groups of dyes (Acid and Direct) on the performance of constructed wetlands on dyes removal; and (c) the influence of operational parameters such as contact time, resting time and loading rate on dye removal.

2. Materials and methods

2.1. Wetland rig and operation

The study has been conducted between 1 May 2015 and 31 May 2016. The first month may be viewed as the start-up period. An experimental constructed wetland rig (Fig. 1) treating textile wastewater has been operated within a greenhouse located on top of the Newton Building (The University of Salford). The rig has been designed to assess the system performance by simulating processes occurring within full-scale constructed wetlands. The rig comprises twenty-two vertical-flow wetland filters, allowing wastewater to drain vertically, enhancing aerobic biodegradation of organic matter and nitrogen (Fuchs 2009). The experimental wetland filters were located randomly in the experimental rig to minimise random impacts of parameters such as sunlight direction and temperature differences on the wetland performances.

Fig. 1. Experimental vertical-flow constructed wetland rig located within a greenhouse at the beginning of the experiment (Picture taken by Mr. Amjad Hussein on 15 May 2015).

Operational parameters such as contact time, retention time and hydraulic loading rate on dye removal were assessed. Contact time is defined as the duration the wastewater is in contact with the wetland filter content. In comparison, resting time is the duration when the wetland filter is empty (i.e. no wastewater input). The dyes AB113 and BR46 were tested at two different concentrations (target concentrations of 7 mg/l and 215 mg/l) for different retention times (approximately 48 h and 94 h) to assess their influence on the performance of vertical-flow constructed wetlands.

Round and black plastic drainage pipes were used to construct the vertical-flow wetlands (Fig. 1). All twenty-two wetlands were designed according to the following dimensions: height of 100 cm and diameter of 10 cm. One wetland was filled with water, another one was filled to a depth of 90 cm with unwashed gravel and the other wetlands were filled to a depth of 90 cm with washed gravel (Table 2). Two different layers of gravel were used as filter media. Large gravel with a diameter of 10–20 mm was applied as the bottom layer to prevent clogging of the outlet. Pea gravel with a diameter of 5–10 mm was located at the top layer. The outlet valves were located at the centre of the bottom plate of each wetland. The internal diameter of the vinyl outlet tubing was 10 mm.

Selected wetlands were planted with *P. australis* (Table 2). The growth of *P. australis* was monitored. Dead above-ground plant parts were cut down to about13 cm height. The cuttings were recycled by placing them into their corresponding wetland filters.

Table 1
Previous studies (listed in order of date) on textile wastewater treatment by constructed wetlands.

Dye used	Type of wetland	Design characteristics	Plants used	Removal performance	Duration (days)	Country of operation	References
AB113, *RB171	VF	Gravel-sand	*P. australis*	98% colour	70	USA	Pervez et al. (2000)
AO7	VF	Gravel-sandy clay soil	*P. australis*	74% colour, 64% COD and 71% TOC	77	Portugal	Davies et al. (2005)
Various dyes in real wastewater	HF	Gravel-sand	*Typha* and cocoyam	77% colour, 72% COD and 59% sulphate	84	Tanzania	Mbuligwe (2005)
*RB5, DY211, VY46	VF-HV	Gravel-sand-tuff	*P. australis*	90% colour, 84% COD, 93% TSS, 52% T-N, 87% $N_{organic}$, −331% NH_4-N, 88% sulphate, 80% anion surfactant and 93% TSS	60	Slovenia	Bulc and Ojstršek (2008)
AO7	VF	Gravel-sludge	*P. australis*	94% colour, 95% COD and 86% NH_4-N	27	n/a	Ong et al. (2011)
AY 2G E107	VF	Gravel-sand-zeolite	*Canna* and *Typha*	95% colour, 64% COD, 94% PO_4-P and 77% NH_4-N	90	Turkey	Yalcuk and Dogdu (2014)

Note: AB, acid blue; *RB, reactive blue; AO, acid orange; VF, vertical flow; COD, chemical oxygen demand; TOC, total organic carbon; **RB, reactive black; DY, disperse yellow; VY, vat yellow; HV, horizontal flow; TSS, total suspended solid; T-N, total nitrogen; N, nitrogen; NH_4-N, ammonium nitrogen; PO_4-P, Ortho-phosphate-phosphorus; AY, acid yellow.

Table 2
Packing order of the experimental constructed wetland set-up. Samples were taken at the end of each contact time period, just before the start of the resting time period between 1 May 2015 and 31 May 2016.

Wetland number	Media	Plants	Dye			Resting time (h)	Contact time (h)
			Type	Mean (mg/l)	SD (mg/l)		
1	Only Water	No	None	n/a	n/a	2	94
2	Unwashed gravel	No	None	n/a	n/a	2	94
3	Washed gravel	No	None	n/a	n/a	2	94
4	Washed gravel	No	BR46	6.9	1.33	2	94
5	Washed gravel	No	AB113	6.6	1.82	2	94
6,7	Washed gravel	Yes	AB113	6.6	1.82	2	94
8,10	Washed gravel	Yes	BR46	6.9	1.33	2	94
11,12	Washed gravel	Yes	BR46	6.9	1.33	48	48
13,14	Washed gravel	Yes	AB113	6.6	1.82	48	48
15,16	Washed gravel	Yes	BR46	209.3	4.48	48	48
17,18	Washed gravel	Yes	AB113	221.5	31.86	48	48
19,20	Washed gravel	Yes	BR46	209.3	4.48	96	96
21,22	Washed gravel	Yes	AB113	221.5	31.86	96	96

Note: SD, standard deviation; n/a, not applicable; BR, basic red; AB, acid blue.

The dyes AB113 and BR46 were used at two inflow concentrations (low and high) to assess the performance of the vertical-flow wetland systems to treat dye at the presence of fertiliser only. No other contaminants were added to allow for a statistically sound assessment of the impact of dye. The fact that apart from some high dye concentrations all other concentrations in the inflow were relatively low may naturally result in low outflow concentrations as well, indicating good treatment performance, but not necessarily good removal performances.

Details for each dye can be found in Tables 2 and 3. The textile dye BR46 had a wavelength for the maximum absorbance (λ_{max}) of 530 nm (Khatae, 2009), and was obtained from Dystar (Am Prime Park, Raunheim, Germany). The dye AB113 had a λ_{max} of 566 nm (Shirzad-Siboni et al., 2014), and was obtained from Sigma Aldrich (The Old Brickyard, New Road Gillingham, Dorset, United Kingdom). Dye stock solutions were prepared by dissolving 10 g of each dye in 1000 ml of distilled water. The experimental solutions were obtained by diluting stock solution samples to the required concentrations.

The fertiliser TNC Complete, which is an aquatic plant supplement bought from TNC Limited (Spotland Bridge Mill, Mellor Street, Rochdale, United Kingdom), was used in the experimental work as a nutrient and trace element source for the plants. The corresponding ingredient composition was as follows: nitrogen (1.5%), phosphorus (0.2%), potassium (5%), magnesium (0.8%), iron (0.08%), manganese (0.018%), copper (0.002%), zinc (0.01%), boron (0.01%) and molybdenum (0.001%). Ethylene diamine tetra acetic acid (EDTA), which is used as a source for copper, iron, manganese and zinc, is also provided by TNC Complete. For ten litre of freshly prepared influent, one millilitre fertiliser was added.

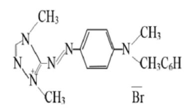
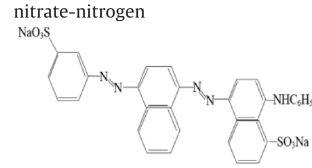
The wetland system has been designed to operate in batch flow mode to avoid expenses such as pumping and automatic control costs. Wastewater was poured directly into the wetland filter from the top. The wastewater stayed within the wetland for the duration of the contact time. The treated wastewater was released from the wetland through an outlet pipe located in the centre of the wetland bottom. The duration the liquid stayed within the wetland is the resting time (see also above).

2.2. Wetland filter set-up

Table 4 outlines the application of the simplified statistical wetland filter set-up design (Table 2) to assess the impact of individual key variables. Wetland 1 was deliberately left empty (without media and plants; Table 2). Only tap water was used as the influent. The mean tap water values for dissolved oxygen (DO), pH, electric conductivity (EC), redox potential, total suspended solids (TSS) and turbidity were 10.1 mg/l, 7.6, 85.1 μS, −32 mV, 1 mg/l and 1.3 NTU, respectively.

Wetland 2 was filled with unwashed gravel, while Wetland 3 comprised washed gravel and tap water. Wetlands 4, 8/10, and 11/12 were filled with the same media as Wetland 3, and planted with *P. australis*, except for Wetland 4, which was used as a control (Table 2). The rhizomes of the plants were washed free of sediment

Table 3
Details of dyes used in the experimental constructed wetlands.

Dye	BR46	AB113
Molecular weight	401.3	681.65
Molecular formula	$C_{18}H_{21}BrN_6$	$C_{32}H_{21}N_5Na_2O_6S_2$
Source	Dystar	Sigma Aldrich
CASRN	12221−69-1	3351−05-1
Purity of dye (%)	70–80	Approximately 50
Nutrient content for 200 mg of dye (mg/l)	50.0 ortho-phosphate-phosphorus, 7.5 ammonia-nitrogen and 0.93 nitrate-nitrogen	137.0 ortho-phosphate-phosphorus, 7.7 ammonia-nitrogen and 1.8 nitrate-nitrogen
Chemical structure		

Note: CASRN, chemical abstracts survey registry number; BR, basic red; AB, acidic blue.

Table 4
Application of the simplified statistical wetland filter set-up design (Table 2) to assess the impact of individual key variables.

Comparison of two wetland systems with each other		Impact to be assessed
First wetland with number	Second wetland with number	
1	2	Unwashed gravel
2	3	Washed gravel
3	4	BR46
3	5	AB113
4	5	Difference between BR46 and AB113
5	6,7	*Phragmites australis*
6,7	8,10	Difference between BR46 and AB113
8,10	11,12	Decrease in contact time(or increase in resting time)
11,12	13,14	Difference between BR46 and AB113
11,12	15,16	Increased BR46 concentration
13,14	17,18	Increased AB113 concentration
15,16	19,20	Increased contact and resting times
17,18	21,22	Increased contact and resting times
15,16	17,18	Difference between BR46 and AB113
19,20	21,22	Difference between BR46 and AB113

and planted in selected gravel-filled wetlands. The influent was tap water mixed with BR46 (6.9±1.33 mg/l).

The contact time for Wetlands 8 and 10 was 94 h, and the resting time was 2 h, while for Wetlands 11 and 12, the contact time was 48 h and the resting time was 48 h (Table 2). The purpose for different contact and resting times was to assess the performance of wetlands in removing dyes under different design conditions, and compare the results with those for Wetland 4 to assess the importance of the presence of plants on dye removal and other parameters. The same previously explained set-up was used for Wetlands 5, 6/7, and 13/14, but with the dye AB113 at a concentration of about 6.6±1.82 mg/l.

The Wetlands 15/16, and 19/20 contained the same media, plants and dye (BR46) as Wetlands 8/10, and 11/12, but the dye concentration was approximately 209 mg/l. The Wetlands 15 and 16 are replicates with 48-h contact time and 48-h resting time, while the Wetlands 19 and 20 are replicates with 96-h contact time and a resting time of 96 h. Wetlands 17, 18, 21 and 22 are similar to Wetlands 15, 16, 19 and 20, but the influent dye was AB113 at a concentration of about 216 mg/l (Table 2).

2.3. Experimental and statistical analysis

The water quality analysis was performed according to APHA (1995), unless stated otherwise, to assess the annual and seasonal treatment performance. Both influent and effluent were analysed. The effluent was obtained from the bottom of each wetland filter. The inflow wastewater was freshly prepared before it was poured into the wetland from the top.

The spectrophotometer DR 2800 Hach Lange (www.hach.com) was applied for the water quality analysis for variables including dyes, COD, NH_4-N, NO_3-N, PO_4-P and TSS. Turbidity was determined with a Turbicheck Turbidity Meter (Lovibond Water Testing, Tintometer Group, Division Street, Chicago, IL, USA). The pH and redox potential for all samples was measured using a VARIO pH meter (Wissenschaftlich-Technische Werkstätten, Weilheim, Germany). This meter was calibrated with standard buffer solutions of pH 4, 7 and 9 each two weeks or whenever required. The EC and total dissolved solids (TDS) were measured using a Mettler Toledo Education Line Conductivity Meter (Boston Road, Leicester, UK). The DO was measured using a Hach Lange HQ30d Flexi Meter (Pacific Way, Salford, UK). Water samples were taken regularly as shown in Table 2, and they were brought to the university-based laboratory for testing.

Water samples were always taken at the same time (10:00 to 11:00 am) to ensure that the various environmental boundary conditions, which are variable throughout the day, such as diurnal variations of pH and DO, would not impact on the results. Laboratory tests were performed to measure dyes, COD, PO_4-P, NO_3-N, NH_4-N, EC, DO, redox potential, pH, TSS, TDS and turbidity. The temperature at the site is recorded each week using a thermometer placed alongside the constructed wetlands.

In order to investigate statistically significant differences, the Shapiro-Wilk's test (Shapiro and Wilk, 1965; Razali and Wah, 2011) was used to assess the normality of data. A one-way analysis of variance (ANOVA) test using Statistical Package for the Social Science software was applied to analyse normal distributed data, while the Mann-Whitney test was used to analyse non-normal data

(Stoline, 1981; Kasuya, 2001). These ANNOVA and Mann-Whitney tests were used to compare means between different treatments such as the ones highlighted in Table 4. Significant findings have been outlined and discussed. Somehow surprising and/or important insignificant findings have occasionally been highlighted as well.

3. Results and discussion

3.1. Test of normality for plant and water quality variables

Tests of normality results for dimensions of *P. australis* and for effluent water quality characteristics regarding general physical and chemical variables are discussed in the supplementary material S1

3.2. Plant growth assessment

When temperatures started to decrease in winter (see Supplementary material S2), the plants in this experiment began to yellow. The above-ground parts died, were cut and returned to the wetland filters. The authors followed common practice to cut the above-ground plant parts down to between 10 and 15 cm height according to Stefanakis et al. (2014).

Based on the experimental results, the most critical observation is that for all considered concentrations and contact times, the plants that were subjected to the dye AB113 revealed more effective growth compared to those that were fed by the dye BR46. Taking into consideration the potential impact of retention time, it has been noted that plants associated with a long contact time showed much better growth than those plants linked to a short one (Table 5). These findings support those by Pagter et al. (2005), who investigated the effect of water stress tolerance of *P. australis* grown in the laboratory by examining effects of different levels of required water. The results showed that a water deficit reduces the leaf biomass per plant and the leaf area.

The ANOVA and Mann-Whitney tests were applied for normal and non-normal distributed data, respectively (see above). Regarding the growth of plants, there is no significant ($p > 0.05$) difference with respect to the length and diameter at the low concentration of the dye AB113, while for the high concentration, there is a significant ($p > 0.05$) difference for the diameter but no significant ($p > 0.05$) difference for the length. In case of plants grown in the presence of the dye BR46, there is a significant ($p < 0.05$) difference for both the length and diameter at the low concentration of dye, while no significance ($p > 0.05$) for both parameters was noted at the high concentration of dye. Readers may wish to consult Table S1 and the corresponding discussion in the supplementary material S1 for more details. However, no generic interpretation of the data can be made for both dyes, indicating further research needs by ecologists.

3.3. Dissolved oxygen and redox potential

Redox potentials of less than −100 mV indicate anaerobic environments, while values greater than 100 mV indicate aerobic environments (Suthersan, 2001). Redox potential and DO values for the effluents of both dyes were in the range between −32 and −6 mV and between 5.4 and 8.2 mg/l, respectively, for low dye concentrations, and between −32 and 4 mV and between 6.0 and 7.5 mg/l, respectively, for high dye concentrations (Tables 2 and 5). However, there is the possibility that the effluent samples got aerated between taking the samples and corresponding measurements. Nevertheless, these findings indicate that dye degradation may have taken place in both aerobic and anaerobic environments.

3.4. Electrical conductivity, total suspended solids and turbidity

Electrical conductivity can be used as an indirect measure of the charge (or the ion-carrying species) in the wetland outflow (Islam et al., 2011). For the low concentration of both dyes (BR46 and AB113), as shown in Table 6, an increase was noted for Wetlands 11/12 and Wetlands 13/14 (contact times of 48 h for each), respectively, when compared to Wetlands 8/10 and 4 as well as Wetlands 6/7 and 5 (contact times of 94 h for each), respectively. For the high concentration in case of dye AB113, the average value of EC in the effluent of Wetlands 21/22 (high resting and contact times) is the same as for the influent, while for Wetlands 17/18 (low resting and contact times), there is a decrease in the average value, if compared to the influent. In case of dye BR46, there is a statistically insignificant ($p > 0.05$) increase in the average value for the effluent of Wetlands 15/16 (low resting and contact times; 389 ± 99.73 μS/cm), and Wetlands 19/20 (385 ± 83.21 μS/cm), if compared to the influent (357 ± 112.3 μS/cm).

Yalcuk and Dogdu (2014) have reported similar findings. The measurement of TSS is important for the design of water treatment facilities (Dzurik, 2003). For the dye BR46 in case of low concentration, there is an increase in the effluent for Wetlands 4, 11 and 12 (Tables 2 and 6), while there is no increase in the average value for Wetlands 8 and 10 (Tables 2 and 6), if compared with the influent.

For the high concentration scenario, the removal in Wetlands 19 and 20 (high resting and contact times) is better than that in Wetlands 15 and 16 (low resting and contact times). For the dye AB113 in case of low concentration, increased values of TSS in the effluent were noted for the Wetlands 5, 6/7, and 13/14 (Tables 2 and 6). Only a slight increase was noted for Wetlands 6/7 (low resting and high contact times), while a higher increase was recorded for Wetlands 13/14 (high resting and low contact times). In case of the high concentration scenario, a statistically insignificant ($p > 0.05$) value was noted for the Wetlands 17/18 (low resting and contact times) and 21/22 (high resting and contact times). Increased values for Wetlands 21/22 were noted (392 ± 253.13 mg/l), if compared to the Wetlands 17/18 (371.7 ± 177.57 mg/l).

Turbidity is often used to interpret the degree of clarity of water. It is a variable often applied as an indicator of the amount of suspended sediments and/or larger micro-organisms in water. A high turbidity of surface water may also indicate elevated concentrations of TSS, reduced algal populations, and potential harm to fish and other aquatic fauna (Postolache et al., 2007).

For the dye BR46 ($p < 0.05$) in case of the low concentration scenario, removal was noted for the planted Wetlands 8/10 (3.0 ± 1.33 mg/l), while there was an increase linked to the unplanted Wetlands 4 (8.8 ± 7.12 mg/l; $p > 0.05$) and planted Wetlands 11/12 (3.6 ± 1.89 mg/l) (see also Tables 2 and 6), if compared to the influent (3.5 ± 2.89 mg/l). In case of high concentrations, good removal was noted for the Wetlands 15/16 (low resting and contact times; 6.6 ± 2.20 mg/l; $p > 0.05$) and the Wetlands 19/20 (high resting and contact times; 6.8 ± 3.90 mg/l; $p > 0.05$), when compared with the influent (16.6 ± 3.10 mg/l). For the dye AB113 in case of low concentration ($p < 0.05$), increased values in the effluent were noted for the unplanted Wetland 5 (39.2 ± 27.40 mg/l), the planted Wetlands 6/7 (19.1 ± 16.11 mg/l) and the planted Wetland 13/14 (46.8 ± 32.67 mg/l) (see also Tables 2 and 6), if compared to the influent (7.4 ± 5.67 mg/l). A greater increase was recorded for Wetlands 13/14 (high resting and low contact times), if compared to Wetlands 6/7 (low resting and high contact times). In case of the high concentration scenario, a removal was noted for Wetlands 17/18 (low resting and contact times; 208.3 ± 196.67 mg/l; $p < 0.05$), while there was an increase in the average value for Wetlands 21/22 (high resting and contact times; 224 ± 202.69 mg/l; $p > 0.05$), if compared to the influent (213.4 ± 157.62 mg/l; $p < 0.05$).

Table 5
Dimensions of *Phragmites australis* (Cav.) Trin. ex Steud. (Common Reed) planted in the experimental wetlands (Tables 2 and 4).

Characteristics			Length (cm)			Diameter (mm)		
Dye	Wetland number	Number of steams	Mini-mum	Maxi-mum	Mean ±SD	Mini-mum	Maxi-mum	Mean ±SD
BR46	8 and 10	40	40	110	86 ± 25.0	2.5	3.8	3.6 ± 0.16
	11 and 12	28	17	44	30 ± 9.0	1.1	1.8	1.5 ± 0.30
	15 and 16	34	14	50	27 ± 13.0	1.0	1.2	1.1 ± 0.07
	19 and 20	23	12	34	23 ± 8.1	0.9	1.5	1.2 ± 0.25
AB113	6 and 7	45	110	141	131 ± 9.5	3.4	3.8	3.6 ± 0.16
	13 and 14	30	23	41	32 ± 4.5	2.1	2.4	2.2 ± 0.10
	17 and 18	36	28	46	35 ± 6.0	2.1	2.3	2.2 ± 0.06
	21 and 22	28	22	52	39 ± 10.7	1.8	2.5	2.1 ± 0.25

Note: BR, basic red; AB, acidic blue; SD, standard deviation.

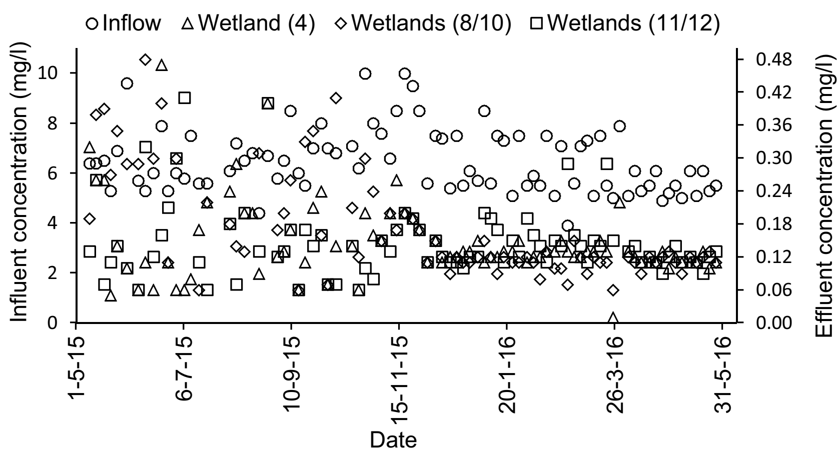


Fig. 2. Inflow and outlow concentrations of Basic Red 46 for Wetlands 4, 8/10 and 11/12 (see also Tables 2 and 4).

Many researchers confirmed that vertical-flow wetlands have a relatively poor ability to remove TSS and turbidity (Lin et al., 2005; Bulc and Ojstršek, 2008). Regarding this study, for the low concentration of BR46, a long contact time (94 h) was better than a short (48 h) one in removal of TSS and turbidity. While for high concentrations, the short contact time was better than the long contact time. In case of low and high concentrations for AB113, the four-day contact time was not enough to reduce TSS and turbidity.

3.5. pH value

For the low concentration of the dye BR46, the average pH inflow value was 7.2. A slight increase in the effluent pH values of 0.16 and 0.03 were noted for the unplanted Wetland 4 and the planted Wetlands 11/12 (see also Tables 2 and 6), respectively, while there was a decrease of 0.31 for Wetlands 8/10 (replicates), as shown in Table 6. For the high concentration, the average influent value was 6.49. Increases of 0.19 and 0.57 were noted for Wetlands 15/16 (low contact and resting times) and Wetlands 19/20 (high contact and resting times), respectively. In case of the dye AB113 for the low concentration scenario, the average influent pH value was 7.32. A slight pH increase of 0.11 was noted for the unplanted Wetland 5, while the pH value decreased for the planted Wetlands 6/7 and the planted Wetlands 13/14 (see also Tables 2 and 6) by 0.44 and 0.15, respectively. For the high concentration, the average influent pH value was 8.39. The pH value decreased for the Wetlands 17/18 (low resting and contact times) and the Wetlands 21/22 (high resting and contact times) by 1.10 and 1.06, respectively, as shown in Table 6.

These results indicate the ability of macrophytes such as *P. australis* to modify pH conditions in the rhizosphere, confirming results by Brix et al. (2002). However, these researches used a different wetland plant (*Typha angustifolia* L.).

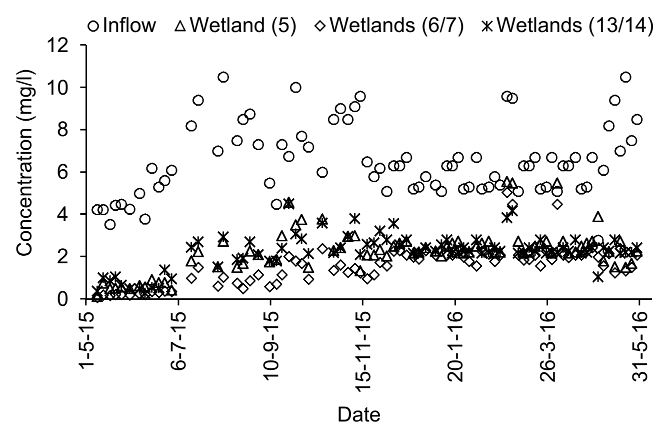


Fig. 3. Inflow and outlow concentrations of Acid Blue 113 for Wetlands 5, 6/7 and 13/14 (see also Tables 2 and 4).

3.6. Dye removal and chemical oxygen demand removal

The percentage removal of dyes is shown in Table 7. The degradation of azo dyes takes place both in aerobic and anaerobic conditions via various processes involving enzymes and/or chemical reduction (Pandey et al., 2007). BR46 is theoretically easier to degrade than AB113 due to a lower molecular weight (Table 3).

There is no significant ($p > 0.05$) difference in dye removal for the low concentration of BR46 between the unplanted control Wetland 4, the planted Wetlands 8/10 and the planted Wetlands 11/12 (Tables 2 and 6, and Fig. 2). It follows that the presence of plants does not affect dye removal. In comparison, for AB113, the dye removal in the planted Wetlands 6 and 7 was significantly ($p < 0.05$) better

Table 6
Inflow and outflow water quality characteristics for general physical and chemical variables related to different wetlands (Tables 2 and 4).

Characteristics				pH			Redox potential (mV)			Dissolved oxygen (mg/l)		
Dye	Type of flow	Wetland number	No. of samples	Mini-mum	Maxi-mum	Mean ±SD	Mini-mum	Maxi-mum	Mean ±SD	Mini-mum	Maxi-mum	Mean ±SD
BR46	In	n/a	70	6.80	7.54	7.2 ± 0.147	−39	5	−25 ± 7.7	9.0	10.9	9.7 ± 0.46
	Out	4	70	7.11	7.75	7.36 ± 0.137	−43	−14	−29 ± 6.35	5.5	10.0	8.2 ± 1.12
	Out	8 and 10	70	6.67	7.21	6.89 ± 0.148	−22	8	−5 ± 6.79	3.7	8.1	5.4 ± 0.92
	Out	11 and 12	70	6.85	8.18	7.23 ± 0.212	−40	−7	−22 ± 9.25	2.9	9.5	7.5 ± 1.54
	In	n/a	70	6.04	6.83	6.49 ± 0.179	−6	28	12 ± 8.06	8.7	10.7	9.6 ± 0.41
	Out	15 and 16	70	6.35	7.11	6.68 ± 0.217	−17	17	4 ± 7.53	3.8	9.2	6.3 ± 1.34
	Out	19 and 20	35	6.65	7.37	7.06 ± 0.163	−24	4	−13 ± 7.58	3.2	8.7	6.0 ± 1.29
AB113	In	n/a	70	7.14	7.65	7.32 ± 0.122	−43	−21	−30 ± 5.4	8.9	11.2	9.6 ± 0.55
	Out	5	70	7.20	7.77	7.43 ± 0.165	−51	−19	−32 ± 6.5	5.1	10.1	8.3 ± 1.38
	Out	6 and 7	70	6.62	7.4	6.88 ± 0.184	−33	17	−6 ± 8.2	3.7	6.9	5.4 ± 0.77
	Out	13 and 14	70	6.78	7.57	7.17 ± 0.199	−40	5	−23 ± 8.2	3.5	10.4	8.1 ± 1.45
	In	n/a	70	7.25	9.04	8.39 ± 0.432	−129	−46	−88 ± 18.2	8.2	10.7	9.5 ± 0.49
	Out	17 and 18	70	7.04	7.6	7.29 ± 0.118	−46	−10	−28 ± 9.6	5.4	9.9	7.5 ± 1.19
	Out	21 and 22	35	7.10	7.99	7.33 ± 0.202	−74	−12	−32 ± 14	3.9	10.0	7.1 ± 1.76
Characteristics				Total suspended solids (mg/l)			Turbidity (NTU)			Electric conductivity (μS/cm)		
BR46	In	n/a	70	0	5.1	2.0 ± 1.51	1.6	18.5	3.5 ± 2.89	90	270	159 ± 51.6
	Out	4	70	0	23.0	6.8 ± 5.81	1.9	32	8.8 ± 7.12	70	483	193 ± 79.1
	Out	8 and 10	70	0	6.2	2.0 ± 1.88	1.5	6	3.0 ± 1.33	103	749	257 ± 97.4
	Out	11 and 12	70	0	10.2	3.4 ± 3.33	1.9	12	3.6 ± 1.89	82	419	180 ± 62.4
	In	n/a	70	37.1	59.0	47.9 ± 5.11	8.9	24	16.6 ± 3.1	147	701	357 ± 112.3
	Out	15 and 16	70	1.1	21.3	7.6 ± 4.70	2.9	13	6.6 ± 2.2	253	647	389 ± 99.73
	Out	19 and 20	35	2.3	41.0	6.9 ± 8.41	2.9	20.1	6.8 ± 3.9	251	535	385 ± 83.21
AB113	In	n/a	70	5.5	25.0	14.1 ± 5.40	1.3	21.0	7.4 ± 5.67	70	350	220 ± 104.1
	Out	5	70	3.4	85.0	41.9 ± 23.07	5.8	134.0	39.2 ± 27.40	75	396	242 ± 87.9
	Out	6 and 7	70	1.4	64.0	18.8 ± 15.92	2.8	57.0	19.1 ± 16.11	114	448	294 ± 85.4
	Out	13 and 14	70	3.5	133.0	46.5 ± 33.60	2.3	128.0	46.8 ± 32.67	83	430	235 ± 99.6
	In	n/a	70	119.0	843.0	364.8 ± 195.47	21	660	213.4 ± 157.62	465	976	635 ± 123.2
	Out	17 and 18	70	121.0	706.0	371.7 ± 177.57	12	650	208.3 ± 196.67	456	854	624 ± 99.1
	Out	21 and 22	35	67.0	863.0	392 ± 253.13	9	675	224 ± 202.69	502	844	635 ± 93.3

Note: BR, basic red; AB, acidic blue; SD, standard deviation; n/a, not applicable.

Table 7
Dye and chemical oxygen demand (COD) removal for different wetlands (Tables 2 and 4).

Characteristics				Dye concentration (mg/l)			Dye loading rate (mg/l/d)				
Dye	Type of flow	Wetland number	No. of samples	Mini-mum	Maxi-mum	Mean ±SD	Removal (%)	Mini-mum	Maxi-mum	Mean ±SD	Removal (%)
BR46	In	n/a	70	4.4	10.0	6.9 ± 1.33	n/a	0.42	0.94	0.65 ± 0.12	n/a
	Out	4	70	0.1	0.9	0.3 ± 0.2	96	0.006	0.08	0.03 ± 0.02	95
	Out	8 and 10	70	0.1	0.7	0.2 ± 0.1	97	0.006	0.07	0.02 ± 0.01	97
	Out	11 and 12	70	0.1	0.5	0.2 ± 0.1	97	0.005	0.02	0.02 ± 0.01	97
	In	n/a	70	200.0	218.0	209.3 ± 4.48	n/a	19.4	21.1	20.3 ± 0.39	n/a
	Out	15 and 16	70	2.0	56	37.7 ± 19	82	0.19	5.4	3.7 ± 1.8	82
	Out	19 and 20	35	2.0	29.8	12.5 ± 9	94	0.12	1.74	0.7 ± 0.5	89
AB113	In	n/a	70	3.5	10.5	6.6 ± 1.82	n/a	0.27	0.82	0.51 ± 0.11	n/a
	Out	5	70	0.1	4.6	1.9 ± 1.0	71	0.007	0.33	0.14 ± 0.07	73
	Out	6 and 7	70	0.1	4.5	1.3 ± 1.1	80	0.006	0.32	0.09 ± 0.08	82
	Out	13 and 14	70	0.3	4.5	2.1 ± 1	68	0.216	0.32	0.15 ± 0.07	71
	In	n/a	70	155.0	292.0	221.5 ± 31.86	n/a	12.77	22.6	17.23 ± 2.5	n/a
	Out	17 and 18	70	17.1	191.5	63 ± 48	71	1.23	13.8	4.54 ± 3.45	74
	Out	21 and 22	35	26.91	187.5	59 ± 40	73	0.97	6.77	2.12 ± 1.44	75
Characteristics				COD concentration (mg/l)			COD loading rate (mg/l/d)				
BR46	In	n/a	20	11	32	22 ± 6	n/a	0.88	2.56	1.76 ± 0.48	n/a
	Out	4	20	2.8	19.5	11 ± 5.4	50	0.21	1.44	0.81 ± 0.4	54
	Out	8 and 10	20	0.3	13.8	7.2 ± 4.3	67	0.02	1.02	0.53 ± 0.32	70
	Out	11 and 12	20	0.7	16.3	9.1 ± 5.1	59	0.05	1.2	0.67 ± 0.38	62
	In	n/a	20	249	280	261 ± 11	n/a	19.9	22.4	20.88 ± 0.8	n/a
	Out	15 and 16	20	6	100	69 ± 34	74	0.44	7.4	5.1 ± 2.52	76
	Out	19 and 20	20	14	40	28 ± 9	89	0.52	1.48	1.04 ± 0.33	95
AB113	In	n/a	20	36.9	86.4	48.2 ± 12.2	n/a	2.95	6.91	3.84 ± 1.04	n/a
	Out	5	20	27.6	73	46 ± 14.7	5	2.04	5.4	3.4 ± 1.11	11
	Out	6 and 7	20	14	78	40.6 ± 19.4	16	1.04	5.77	3.03 ± 1.55	21
	Out	13 and 14	20	24	72	44.6 ± 12.9	7	1.78	5.32	3.18 ± 0.96	17
	In	n/a	20	543	610	584 ± 34	n/a	43.44	48.8	47.84 ± 1.3	n/a
	Out	17 and 18	20	117	454	271 ± 103	54	8.66	33.6	22.34 ± 7.7	53
	Out	21 and 22	20	126	362	262 ± 86	55	9.32	26.78	21.46 ± 6.3	55

Note: BR, basic red; AB, acidic blue; SD, standard deviation; n/a, not applicable.

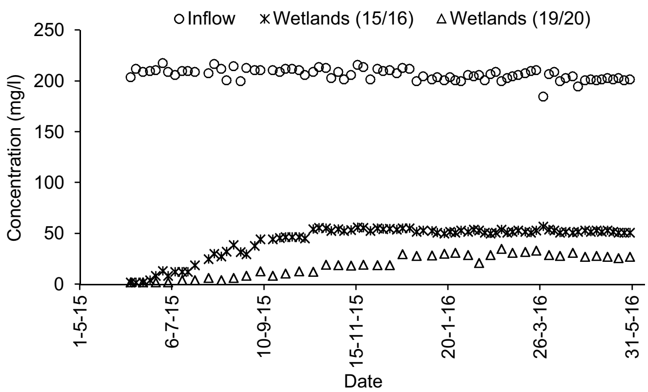


Fig. 4. Inflow and outlow concentrations of Basic Red 46 for Wetlands 15/16 and 19/20 (see also Tables 2 and 4).

than the one for the unplanted control Wetland 5 (Fig. 3) due to the presence of plants (Keskinkan and Lugal Göksu, 2007).

For high concentrations, the best percentage removals for BR46 were noted for Wetlands 19/20 (high resting and contact times), which were statistically significantly ($p < 0.05$) different, if compared to those of Wetlands 15/16 (low resting and contact times). Wetlands with a low loading rate performed better than those wetlands with a high loading rate (Fig. 4). For the dye AB113, there was no significant ($p > 0.05$) difference in the removal of dyes between Wetlands 17 and 18 (low resting and contact times), and Wetlands 21 and 22 (high resting and contact times) as shown in Table 7.

For COD removal concerning low concentrations of dyes (AB113 and BR46), wetlands with a long retention time had the best COD removal (regardless of planting regime), if compared to the control and other wetlands having short retention times. Cheng et al. (2011) operated two series of vertical-flow constructed wetlands (one planted and other one unplanted) under different C: N: P ratios. Their results obtained showed that the removal of COD depended on the presence and development of plants. The removal rate was higher in the planted wetlands (Cheng et al., 2011).

Concerning high concentrations of dye BR46, the best removal percentages for COD were noted for Wetlands 19/20 (high resting and contact times), which were statistically significantly ($p < 0.05$) different from Wetlands 15/16 (low resting and contact times). Wetlands with a low loading rate performed better than those wetlands with a high loading rate (Table 7). In case for the dye AB113, there was no significant ($p > 0.05$) difference in the removal of COD between Wetlands 17/18 (low resting and contact times) and Wetlands 21/22 (high resting and contact times) as shown in Table 7.

3.7. Nutrient removal

The removal of phosphorous is controlled by chemical and physical adsorption, sedimentation, plant uptake and precipitation in constructed wetland systems (Brix, 1997). For the low concentrations of BR46 and AB113, the removal percentages in Wetlands 8/10 (89%) as well as Wetlands 6/7 (30%) were significantly ($p < 0.05$) better than those of the control Wetlands 4 (78%) and 5 (27%) as well as Wetlands 11/12 (68%) and 13/14 (14%), which have short and long retention times in that order (Table 8). For the high concentrations of BR46, the removal percentages of Wetlands 19/20 (high resting and contact times; 81%) were significantly ($p < 0.05$) better than those for Wetlands 15/16 (low resting and contact times; 70%), while for AB113, the removal percentages of Wetlands 17/18 (low resting and contact times; 78%) ($p < 0.05$) were also significantly better than those for Wetlands 21/22 (high resting and contact times; 71%) as shown in Fig. 5.

Table 8
Inflow and outflow water quality characteristics for nutrients related to different wetlands (Tables 2 and 4).

Characteristics				Ammonia-nitrogen (mg/l)				Nitrate-nitrogen (mg/l)				Ortho-phosphate-phosphorus (mg/l)			
Dye	Type of flow	Wetland number	No. of samples	Mini-mum	Maxi-mum	Mean ± SD	Removal (%)	Mini-mum	Maxi-mum	Mean ± SD	Removal (%)	Mini-mum	Maxi-mum	Mean ± SD	Removal (%)
BR46	In	n/a	20	0.54	0.75	0.62 ± 0.065	n/a	17.9	18.0	18.0 ± 0.03	n/a	5.3	6.3	5.9 ± 0.33	n/a
	Out	4	20	0.45	0.68	0.55 ± 0.086	11	4.5	16.9	9.8 ± 3.98	45	0.9	1.7	1.3 ± 0.27	78
	Out	8 and 10	20	0.21	0.51	0.36 ± 0.111	42	2.5	13.9	7.5 ± 3.69	58	0.19	1.3	0.7 ± 0.4	89
	Out	11 and 12	20	0.49	0.90	0.68 ± 0.118	−10	3.5	16.0	8.9 ± 3.83	50	0.5	2.8	1.9 ± 0.83	68
	In	n/a	20	0.98	1.04	1.02 ± 0.016	n/a	19.3	20.0	19.7 ± 0.18	n/a	55.0	87.0	64.0 ± 8.50	n/a
	Out	15 and 16	20	0.25	0.47	0.35 ± 0.075	66	1.5	16.2	6.2 ± 5.43	69	6.5	47.0	19.3 ± 13.20	70
	Out	19 and 20	20	0.21	0.46	0.31 ± 0.078	70	0.6	12.1	4.9 ± 4.46	75	1.9	43.6	12.1 ± 14.60	81
AB113	In	n/a	20	0.63	0.70	0.66 ± 0.022	n/a	8.4	08.5	8.5 ± 0.03	n/a	6.9	8.3	7.6 ± 0.43	n/a
	Out	5	20	0.37	0.62	0.48 ± 0.079	27	10.5	16.3	12.1 ± 1.38	−42	4.8	6.4	5.6 ± 0.49	27
	Out	6 and 7	20	0.35	0.81	0.52 ± 0.143	21	3.9	6.5	4.9 ± 0.98	42	4.1	6.4	5.3 ± 0.58	30
	Out	13 and 14	20	0.35	0.56	0.47 ± 0.062	29	10.0	15.1	11.6 ± 1.45	−36	5.7	7.7	6.6 ± 0.51	14
	In	n/a	20	2.21	2.77	2.33 ± 0.159	n/a	19.4	20.0	19.8 ± 0.21	n/a	148.0	154.0	151.0 ± 2.10	n/a
	Out	17 and 18	20	0.73	1.54	1.00 ± 0.236	57	0.9	15.2	6.0 ± 5.8	70	15.0	50.4	33.1 ± 11.30	78
	Out	21 and 22	20	0.80	1.97	1.10 ± 0.315	53	0.8	15.2	5.3 ± 5.10	73	23.3	70.3	43.5 ± 15.30	71

Note: BR, basic red; AB, acid blue; SD, standard deviation; n/a, not applicable.

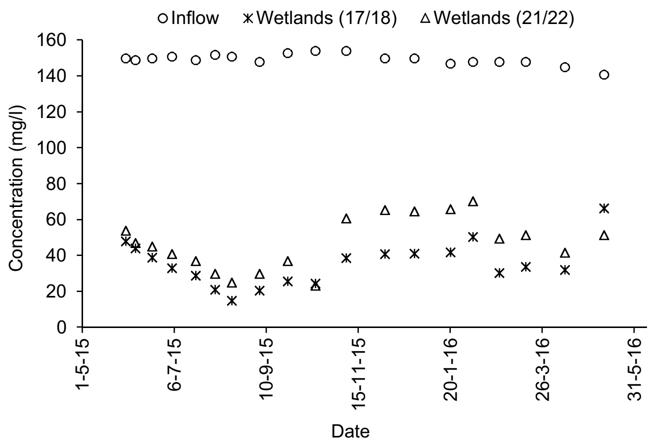


Fig. 5. Inflow and outlow concentrations of ortho-phosphate-phosphorus for Wetlands 17/18 and 21/22 (see also Tables 2 and 4).

As indicated in Table 8, the concentrations of NH_4-N for both dyes are very low due to their chemical structures (Yalcuk and Dogdu, 2014). For the high concentration of dyes, there are no significant ($p > 0.05$) differences between wetlands. For the low concentration of the dye BR46, the removals for the planted Wetlands 8 and 10 were significantly ($p < 0.05$) better than those for the corresponding removal of the unplanted Wetland 4, while there are increases in removal for planted Wetlands 11 and 12 (Table 8). For the low concentration of the dye AB113, the removal percentages with respect to planted Wetlands 13 and 14 were better than those for the unplanted Wetland 4 and the planted Wetlands 6 and 7 (Table 8), which was statistically insignificant ($p > 0.05$).

The removal of NO_3-N is a relatively time-consuming anaerobic process, and naturally takes place mostly in the sediment (Mustafa et al., 2009). For the high concentration of both dyes (BR46 and AB113), the removal percentages for wetlands with a long retention times are better than those for wetlands having short retention times, confirming findings by Van Loosdrecht and Clement (2005). Regarding the low concentration of BR46, the removal percentages for the planted Wetlands 8 and 10 were better than those for the unplanted Wetland 4 and the planted Wetlands 11 and 12 (Table 8). For the low concentrations of AB113, there were significantly ($p < 0.05$) better removal percentages for the Wetlands 6 and 7 (low resting and high contact times), but there were increases in the effluents regarding the NO_3-N removal of Wetlands 13 and 14 (high resting and low contact times) due to the presence of carbon under aerobic conditions (Hu et al., 2009). The removal percentages for the high concentrations of dyes were better than the ones for the low concentration of dyes due to a corresponding increase in carbon percentage.

4. Conclusions and recommendations for further research

The presence of *P. australis* did not affect dye removal ($p > 0.05$), while it had a minor but insignificant impact on COD removal ($p > 0.05$). The use of plants concerning the short contact time scenario for NH_4-N and a low concentration of AB113 is linked to a better removal compared to other cases apart from that for all nutrients; particularly PO_4-P and NO_3-N. In case of low dye concentrations, the presence of plants for the long contact time scenario impacted significantly ($p < 0.05$) positive on the removal efficiencies of PO_4-P, NH_4-N and NO_3-N.

The vertical-flow constructed wetlands showed significantly ($p < 0.05$) good dye removals for low and high concentrations of BR46, and low concentrations of AB113 during all seasons under greenhouse conditions. The COD removal was a function of the dye used. This was explained by the complexity of the chemical structure of the dye and seasonal variations in temperature.

For high concentrations of BR46, wetlands with a long hydraulic retention time were significantly ($p < 0.05$) better that those with a short hydraulic retention time for the removal of PO_4-P. For AB113, wetlands with a shorter hydraulic retention time removed PO_4-P significantly ($p < 0.05$) well. In case of the dye AB113, the four-day contact time was not enough to remove or reduce TSS and turbidity.

The authors recommend to assess (a) the effects of different plant species and an increase of the contact time on particle and dye removal processes; (b) the effect of pH (low and high) on dye removal; (c) the treatment of synthetic textile wastewater contaminated with different dyes, matching the typical characteristics of real effluent from a textile factory; (d) the impact of different dyes mixed with other contaminants and plant growth parameters such as stem diameter; and (e) wetland design implications based on the research findings in terms of investment of area and costs.

Competing interest

Authors have no competing interests.

Supplementary material

Note that on-line supplementary material is associated with this article. Supplementary material S1 comprises a discussion on tests of normality supported by two tables. Supplementary Material S2 shows the temporal variation of temperature throughout the experimental time period.

Acknowledgements

The lead author received a PhD Studentship from the Ministry of Higher Education and Scientific Research of the Iraqi Government via Al-Muthanna University. The authors acknowledge the technical support provided by Dina Ali Yaseen.

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecoleng.2017.01.016.

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IOP Conference Series: Materials Science and Engineering

The Significant Variables of Effluent Constructed Wetlands Treated Domestic Wastewater by Statistical Tests

To cite this article: A Hussein and K Al-Daffaie 2021 *IOP Conf. Ser.: Mater. Sci. Eng.* **1090** 012016

The Significant Variables of Effluent Constructed Wetlands Treated Domestic Wastewater by Statistical Tests

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Abstract. Domestic wastewater was treated by vertical flow constructed wetlands under different set-up designs and operation conditions. These conditions include different aggregate diameters, contact time, resting time, and chemical oxygen demand. The physical and chemical measurements of effluents were obtained. These measurements are Chemical oxygen demand; Biochemical oxygen demand; Ammonia-Nitrogen; Nitrate-Nitrogen; Ortho-Phosphate-Phosphorous; Suspended solids; Turbidity and ph. This research aims to find out which variables are significantly effective by using ANOVA with POST HOC tests. The results showed Chemical oxygen demand; Biochemical oxygen demand, Ammonia-Nitrogen, Nitrate-Nitrogen, Ortho-Phosphate-Phosphorous, and Suspended Solids have significantly effective($P<0.05$).

1. Introduction

Many researchers reported that constructed wetlands have efficiency in treating many types of wastewater, such as textile wastewater [1-3], domestic wastewater [4,5], urban runoff, animal wastewater, and mine drainage [6-8]. Furthermore, many researchers investigated the significant differences in each design and operation variables for using constructed wetlands to treat wastewater [4,5,9]; They used different statistical methods such as the Mann-Whitney U test ANOVA. ANOVA analysis has been employed to explore the differences of such parameters water quality specifically [10,11]. An ANOVA test is a way to determine if experimental data are significant. It helps the researchers to reject the null hypothesis or accept the alternate theory [10].

Al-isawi et al. [9] worked on treated domestic wastewater contaminated by hydrocarbon by vertical flow constructed wetlands located in Newton Building of The University of Salford, Greater Manchester, UK, through the period between 26 June 2012 and 10 June 2014 (six seasons). This wetlands system consists of different design set-up parameters and operational variables, as shown in Table 1. They used the Mann-Whitney test to determine the significant differences for such water quality parameters for all wetlands filter. The results showed no significant difference in COD removal for Wetlands 1-6 and a significant difference in SS for all Wetlands. At the same time, there is no significant difference for all wetlands in the case of nutrients [8]. This research aims to investigate the water seasonal outflow water quality parameters by using statistical approaches.

Table 1 Comparison of the experimental vertical flow wetland set-up [9].

Wetland filters	Aggregate diameter (mm)	Contact time (h)	Resting time (h)	Chemical oxygen demand (mg/l)
Filter 1 and 2	20	72	48	139.3
Filter 3 and 4	10	72	48	139.3
Filter 5 and 6	10	72	48	283.1
Filter 7	10	36	48	139.3
Filter 8	10	36	24	139.3

2. Methodology

2.1. Samples collection

The vertical flow constructed wetlands used to treat domestic wastewater located within a greenhouse on the Newton Building roof, University of Salford, Greater Manchester, UK, as shown in Table 2 [9].

Table 2 Comparison of seasonal inflow and outflow water quality parameters (26 June 2012–10 June 2014) measured in mg/L [9].

	Parameters	Autumn 2012	Winter 2012/2013	Spring 2013	Autumn 2013	Winter 2013/2014	Spring 2014
Inflow	COD[1]	261.0	230.3	186.0	352.5	200.7	245.3
	BOD[2]	108.6	118.0	221.2	167.1	104.3	105.8
	NH4-N[3]	65.0	46.0	69.4	32.2	41.4	23.0
	NO3-N[4]	6.7	12.0	5.2	0.8	5.7	1.6
	PO4-P[5]	18.71	7.18	17.81	14.85	16.37	13.62
	SS[6]	125.7	158.5	379.9	166.6	147.5	122.6
Outflow Filter 1	COD	57.1	64.5	82.5	240.5	72.0	65.6
	BOD	42.7	23.0	54.2	28.3	18.3	26.3
	NH4-N	9.4	11.8	25.1	14.9	4.5	2.7
	NO3-N	1.2	4.0	0.7	0.4	0.5	0.42
	PO4-P	2.46	2.46	5.41	6.93	1.92	2.99
	SS	4.5	4.2	12.1	12.9	14.7	5.8
Outflow Filter 2	COD	57.1	64.5	82.5	86.4	24.9	19.8
	BOD	42.7	23.0	54.2	18.3	9.4	12.7
	NH4-N	9.4	11.8	25.1	12.0	3.9	3.2
	NO3-N	1.2	4.0	0.7	3.6	2.9	2.8
	PO4-P	2.46	2.46	5.41	4.1	3.08	2.44
	SS	4.5	4.2	12.1	6.1	9.6	5.5
Outflow Filter 3	COD	51.6	59.4	69.2	181.7	83.1	72.5
	BOD	35.6	19.4	54.4	33.6	22.3	17.9
	NH4-N	7.0	8.2	20.0	9.9	2.9	1.73
	NO3-N	2.0	5.1	3.0	0.37	0.42	0.42
	PO4-P	2.26	2.23	2.49	6.65	1.79	2.25
	SS	4.3	3.7	5.9	12.3	16.0	7.6
Outflow Filter 4	COD	51.6	59.4	69.2	81.4	81.6	13.59
	BOD	35.6	19.4	54.4	18.9	8.0	8.8
	NH4-N	7.0	8.2	20.0	8.9	3.5	1.89
	NO3-N	2.0	5.1	3.0	4.3	3.2	1.2
	PO4-P	2.26	2.23	2.49	3.76	3.36	2.13

SS	4.3	3.7	5.9	8.1	9.2	5.4

(continued)

Table 2 continued

	Parameters	Autumn 2012	Winter 2012/2013	Spring 2013	Autumn 2013	Winter 2013/2014	Spring 2014
Outflow Filter 5	COD	77.8	80.1	108.4	356.0	112.2	65.7
	BOD	49.0	26.5	76.4	37.9	19.5	14.6
	NH4-N	21.5	22.88	46.2	26.87	8.12	10.45
	NO3-N	5.9	7.1	6.9	0.7	1.0	1.4
	PO4-P	4.1	3.62	3.67	10.65	3.01	2.42
	SS	5.2	4.7	9.6	19.2	14.8	6.5
Outflow Filter 6	COD	77.8	80.1	108.4	107.6	29.5	19.2
	BOD	49.0	26.5	76.4	27.6	8.7	11.7
	NH4-N	21.5	22.88	46.2	23.0	11.7	3.2
	NO3-N	5.9	7.1	6.9	9.3	2.2	5.5
	PO4-P	4.1	3.62	3.67	8.12	3.07	2.75
	SS	5.2	4.7	9.6	10.0	7.8	7.1
Outflow Filter 7	COD	52.4	62.6	64.2	82.5	19.4	19.9
	BOD	40.4	19.4	28.1	18.1	10.1	12.7
	NH4-N	7.8	7.0	12.8	7.3	1.4	5.1
	NO3-N	4.3	6.3	15.8	2.5	4.2	3.1
	PO4-P	2.89	2.71	2.73	5.26	2.86	2.05
	SS	6.3	4.8	5.6	5.2	1.0	1.56
Outflow Filter 8	COD	60.0	62.7	75.0	174.4	18.85	40.0
	BOD	39.6	17.8	26.9	19.6	9.8	13.7
	NH4-N	9.4	8.6	23.1	6.5	1.2	1.6
	NO3-N	4.9	5.4	9.0	5.6	3.1	3.1
	PO4-P	3.08	2.72	2.83	4.74	3.32	2.47
	SS	5.0	4.8	7.9	4.5	1.1	3.9

[1]Chemical oxygen demand, [2]Biochemical oxygen demand, [3]Ammonia-Nitrogen, [4]Nitare-Nitrogen, [5]Ortho-Phosphate-Phosphorous, [6]Suspended solids

2.2. Data analysis

In this research, ANOVA analysis was used to test the differences in water seasonal outflow water quality parameters among general seasons. Also, the Post Hoc approach was utilized to investigate the differences of each parameter between every two seasons to get more in-depth insight. Both analyses were performed using the SPSS package [12-14].

3. Results and discussions

Table 3 summarizes the ANOVA tables of all parameters studied in this research (COD, BOD, NH4-N, NO3-N, PO4-P, and SS). This table reports only the P-Value of each table, where each parameter has its ANOVA Table. From Table 3, it is clear that there is a significant difference ($p<0.05$) in average levels of all parameters among seasons (Autumn 2012, Winter 2012/2013, Spring 2013, Autumn 2013, Winter 2013/2014, and Spring 2014), which are demonstrated in Table 2.

Table 3 P-Value for each parameter, related to Table 2

Parameters	P-Value
COD	0.000
BOD	0.000
NH4-N	0.000
NO3-N	0.044
PO4-P	0.000
S-S	0.005

After revealing significant differences among seasons for all parameters, it is better to use Post Hoc tests to get more detailed results. Tables 4 – 6 represent those results. Table 4 shows significant differences in COD levels ($P < 0.05$) between season 4 and 1,2,3,5 and 6, whereas the differences are not significant between other seasons. On the other hand, BOD levels have shown significant differences ($P < 0.05$) between season 1 and all different seasons. Similarly, season 2 has shown the same results against all other seasons.

Table 4 P-value (from Post Hoc test) for the COD and BOD

Season	COD						BOD					
	1	2	3	4	5	6	1	2	3	4	5	6
1		0.780	0.330	0.000	0.80	0.34		0.000	0.018	0.001	0.000	0.000
2			0.481	0.000	0.608	0.228			0.000	0.460	0.067	0.130
3				0.001	0.227	0.060				0.000	0.000	0.000
4					0.000	0.000					0.012	0.027
5						0.484						0.739
6												

Table 5 shows that, for the NH4-N and NO3-N variables, there are significant differences ($P < 0.05$) between several seasons, such as season 3 and all other seasons, etc., while there are no significant differences ($P > 0.05$) between different seasons. Furthermore, Table 6 has shown some significant differences ($P < 0.05$) such as season 4, in the case of PO4-P, and all other seasons. In the case of SS, the results showed some significant differences ($P < 0.05$) such as season 3 and seasons 1 and 2, while there are no significant differences ($P > 0.05$) between season 3 and seasons 4,5 and 6. At the same time, there are no significant differences ($p > 0.05$) of PO4-P and SS levels between seasons.

Table 5 P-Value (from Post Hoc test) for the NH4-N and NO3-N

season	NH4-N						NO3-N					
	1	2	3	4	5	6	1	2	3	4	5	6
1		0.773	0.000	0.572	0.059	0.034		0.139	0.100	0.955	0.377	0.397
2			0.000	0.782	0.031	0.017			0.865	0.125	0.021	0.023
3				0.000	0.000	0.000				0.090	0.014	0.015
4					0.016	0.008					0.408	0.429
5						0.799						0.970
6												

Table 6 P-Value for (from Post Hoc test) the PO4-P and SS

season	PO4-P						SS					
	1	2	3	4	5	6	1	2	3	4	5	6
1		0.906	0.203	0.000	0.966	0.512		0.745	0.038	0.007	0.015	0.769
2			0.165	0.000	0.939	0.591			0.018	0.003	0.006	0.536
3				0.000	0.189	0.057				0.488	0.691	0.072
4					0.000	0.000					0.767	0.015
5						0.540						0.030
6												

4. Conclusions

This paper has presented a methodology using the ANOVA and POST Hoc test to determine which physical and chemical measurements are significantly useful. The results showed Chemical oxygen demand; Biochemical oxygen demand, Ammonia-Nitrogen, Nitrate-Nitrogen, Ortho-Phosphate-Phosphorous, and Suspended Solids have significantly effective($P<0.05$).

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To cite this article: Ali Sadig Mohommed Bager and A Hussein 2021 *IOP Conf. Ser.: Mater. Sci. Eng.* **1090** 012052

Comparing Between the Imported and Local Bottled Drinking Water by LASSO Regression

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Abstract. Predict the significant variables of the quality measurement results of 10 and 5 kinds of imported and local bottled drinking water, respectively, tested in the Samawah city, Iraq, using regression analysis (LASSO). These variables were pH, turbidity, total dissolved solids, total hardness, calcium, magnesium, sodium, fluoride, nitrates, sulphates, chlorides, iron, manganese. Ph was selected as a y dependant, and others were chosen as x independents. The results showed that nitrates, sulphates, and magnesium were insignificant (Beta > 0.05) in imported and local bottled drinking water, while sodium was insignificant (Beta > 0.05) in local bottled drinking water only. From these results, LASSO regression gave better results.

Keywords: Environment; bottled drinking water packaging; local brands; nitrates; LASSO regression.

1. Introduction

The first time the water was packaged in a bottle to use as bottled drinking water was in 1621, UK [1]. Most people prefer bottled drinking water to tap water because of taste and water quality's regularity over time [2,3]. Another reason was tap water through flow from the source until the consumer may change due to the materials they are exposed from the surrounding environment. In contrast, it is very low in bottled drinking water because it is placed in sealed packaging [4].

In Iraq, the packaged drinking water industry has developed rapidly in the past 30 years and has a high production capacity. Bottled water has different sizes, most of which are used once, and items imported from other countries can be obtained on bottled water and are available on the local market.

Lasso (Least Absolute Shrinkage and Selection Operator) is a linear model estimation method proposed by Tibshirani [5]. It refers to a set of processes that use L1 penalty points to narrow parameter estimates and perform automatic variable selection: the L1 penalty at least squares regression. Like garrote, it shrinks some coefficients while setting the remaining coefficients to precisely zero. Tibshirani believes that lasso is better than ordinary least squares (OLS) regression for two reasons: First of all, the over-specified OLS model usually has a small deviation but a large variance, which is not conducive to its prediction accuracy. This effect can be improved by reducing or setting individual coefficients to zero and swapping some deviations to minimize model variance. Secondly, OLS models may sometimes have a large number of small coefficients, which add little value to the model and complicate the interpretation of effects [6]. The research aims to find the insignificant measurements from all chemical and physical measures tested on the imported and local bottled drinking water by Hussein and Mohammed [4]. LASSO regression will use to assess this aim.

2. Methodology

The methodology involved two sections in this research: sample collection and data analysis (LASSO regression).

2.1. Samples Collection

All the samples that will be analyzed by LASSO regression, as shown in Tables 1 and 2, had been tested according to APHA., (1995) [7,8,9].

Table 1. The quality measurement results of drinking water for the local items [4].

Symbol	Element						
	Mn[1]	Fe[2]	Cl[3]	No_3[4]	F[5]	Na[6]	So_4[7]
	mg/l						
L_1[a]	0.00	0.010	20	4.10	0.600	20	31
L_2	0.00	0.000	36	9	0.095	49.4	47
L_3	0.00	0.015	187	4.30	1.100	50	210
L_4	0.00	0.000	16	0.115	0.300	13	48
L_5	0.01	0.010	197	1.100	1.150	22.5	48
KSA[b] Standard	0.05	0.300	250	10	0.6-1	-	250
Symbol	Element						
	Mg[8]	$CaCo_3$[9]	Ca[10]	TDS[11]	TU[12]	pH	
	mg/l						
L_1	5.05	46	10	118	0.28	7.20	
L_2	3.60	184	68	123	0.22	7.50	
L_3	6.10	145	48	162	0.24	7.65	
L_4	2.35	20	4.1	19.8	0.10	7.30	
L_5	3.60	130	46	204	0.21	7.10	
KSA Standard	30.0	300	75	100-700	5.00	6.5-8.5	

[a]Local, [b] the Kingdom of Saudi Arabia, [1]Manganese, [2]Iron, [3]Chloride, [4]Nitrate, [5]Fluoride, [6]Sodium, [7]Sulphates, [8]Magnesium, [9]Calcium carbonate, [10]Calcium, [11]Total dissolved solids,[12]Turbidity.

2.2. Data Analysis (LASSO Regression)

LASSO regression is a linear regression using shrinkage. The shrinkage is where the data value shrinks towards the center point (for example, the average value). The LASSO program encourages the use of simple, sparse models (models with fewer parameters). This special regression type is very useful for models that show high multicollinearity or when you want to automate certain parts of the model selection (such as variable selection/parameter elimination) [10]. The acronym "LASSO" stands for the least absolute shrinkage and selection operator. LASSO regression aims to obtain the prediction subset that minimizes the prediction error of the quantitative response variable. LASSO achieves this by imposing constraints on the model parameters, which will reduce the regression coefficient of a specific variable to zero [11]. Formally define the method of linear regression model:

$$\underline{Y} = X\underline{\beta} + \underline{\varepsilon} \tag{1}$$

Where:
$\underline{Y}$: A vector represents the variable response observations of the class$(N \times 1)$;
X: The matrix represents the observations of the explanatory variables of the type $(N \times p)$.
$\underline{\beta}$: Vector parameters are estimated from the class$(p \times 1)$.
$\underline{\varepsilon}$: Random error vector of the class $(N \times 1)$ is considered by assuming $E(e) = 0,\ Var\ (e) = I_{\sigma^2}$ is the vector of random errors from a multivariate normal distribution $N(0, \sigma^2 In)$

The columns of X are denoted as $(X_1, X_2, \dots \dots, X_p)$ These represent the P independent variables. The LASSO estimate of β

$$\hat{\beta}^* = \min\{\sum_{i=1}^{n}(y_i - \sum_{j=1}^{p}\beta_j\, x_{ij}) + \lambda \sum_{j=1}^{p}|\beta_j|\} \quad (2)$$

Where:
$\sum_{i=1}^{n}(y_i - \sum_{j=1}^{p}\beta_j\, x_{ij})$ $\quad$ *Residual sum of squar.*
$\sum_{j=1}^{p}\lambda|\beta_j|$ $\quad$ LASSO penalty.

For estimating the parameter in the above equation, the R program (version 3.4.3) and LASSO package will use [12,13].

Table 2. The quality measurement results of drinking water for imported items [4].

Symbol	Element						
	Mn[1]	Fe[2]	Cl[3]	No_3[4]	F[5]	Na[6]	So_4[7]
	mg/l						
I_1[a]	0.0005	0.010	20	4.1	0.6	20	31
I_2	0.0000	0.000	22	5	0.9	22	25.6
I_3	0.0000	0.005	18	0.2	0.2	17	54
I_4	0.001	0.000	56	1.3	0.7	11	19
I_5	0.0000	0.015	21	0.6	0.7	19	33
I_6	0.0000	0.010	21	7.1	0.6	9	34
I_7	0.0000	0.010	15	4.51	0.8	12	51.5
I_8	0.0000	0.010	4.5	0.1	0.2	1	2.2
I_9	0.0000	0.000	43	6	0.67	19	5.95
I_{10}	0.0000	0.010	22.15	6.9	0.85	21	14
KSA[b] Standard	0.05	0.300	250	10	0.6-1	-	250

Symbol	Element					
	Mg[8]	$CaCo_3$[9]	Ca[10]	TDS[11]	TU[12]	pH
	mg/l					
I_1[a]	5.05	46	10	118	0.28	7.2
I_2[b]	7.7	45.5	5.5	105	0.23	7.2
I_3[c]	19.5	105	10	120	0.18	7.8
I_4[d]	10.55	101	23	111	0.1	7.65
I_5[e]	1.8	50	22.5	112	0.22	7.4
I_6[f]	6.45	121.5	38	159	0.26	7.7
I_7[g]	13.4	95	16.5	114	0.115	7.2
I_8[h]	5.55	50	10.7	50	0.23	7.65
I_9[i]	17.6	110	15	139	0.4	7.3
I_{10}[j]	8.5	75	16	120	0.16	7.7
KSA Standard	30	300	75	100-700	5	6.5-8.5

[a]Imported, [b] the Kingdom of Saudi Arabia, [1]Manganese, [2]Iron, [3]Chloride, [4]Nitrate, [5]Fluoride, [6]Sodium, [7]Sulphates, [8]Magnesium, [9]Calcium carbonate, [10]Calcium, [11]Total dissolved solids, [12]Turbidity.

3. Results and discussions

Many researchers have been using LASSO regression as a significance test [14, 15,16,17,18,19]. Both local and imported bottled drinking water, pH element was selected as y parameter, and others were selected as x parameters. The value (*Beta*) for imported and local elements, as shown in table (3). In the case of imported items, the results showed that the elements (Nitrate, Sodium, Sulphates, and Magnesium) are insignificant (*Beta* >0.05) while the elements (Iron, Chloride, Fluoride, Calcium carbonate, Calcium, Total dissolved solids, and Turbidity) are significant (*Beta* <0.05). For the local items, the results showed that the elements (Nitrate, Sulphates, and Magnesium) are insignificant (*Beta* >0.05) while the elements (Iron, Chloride, Fluoride, Sodium, Calcium carbonate, Calcium, Total dissolved solids, and Turbidity) are significant (*Beta* <0.05).

Table 3. The *Beta* elements value of bottled drinking water for imported and local items.

Elements	Named on Program	*Beta* (Imported items)	*Beta* (Local Items)
Mn[1]	VAR001	0.000	0.000
Fe[2]	VAR002	0.000	0.000
Cl[3]	VAR003	0.000	0.000
No_3[4]	VAR004	0.300	0.268
F[5]	VAR005	0.000	0.000
Na[6]	VAR006	0.100	0.000
So_4[7]	VAR007	0.201	1.684
Mg[8]	VAR008	0.416	0.062
$CaCo_3$[9]	VAR009	0.000	0.000
Ca[10]	VAR010	0.000	0.000
TDS[11]	VAR011	0.000	0.000
TU[12]	VAR012	0.000	0.000

[1]Manganese, [2]Iron, [3]Chloride, [4]Nitrate, [5]Fluoride, [6]Sodium, [7]Sulphates, [8]Magnesium, [9]Calcium carbonate, [10]Calcium, [11]Total dissolved solids, [12]Turbidity.

4. Conclusions

This paper has presented a methodology by analyzing LASSO regression for both imported and local bottled drinking water. From the results which have been obtained, we observed that LASSO regression gave better results. LASSO regression is to get the subset of predictors that minimizes prediction error for a quantitative response variable.

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The Quality of Drinking Water Bottled Domestic and Imported in Iraq

Article *in* Journal of Engineering and Applied Sciences · January 2020

DOI: 10.36478/jeasci.2019.10572.10578

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Journal of Engineering and Applied Sciences 14 (Special Issue 9): 10572-10578, 2019
ISSN: 1816-949X

The Quality of Drinking Water Bottled Domestic and Imported in Iraq

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Abstract: Assess the quality of 10 kinds of imported water and 5 domestic varieties of bottled drinking water during the first half of 2017 in the city of Samawah, Iraq and the results compared with the attributes of bottled drinking water issued by the Saudi Arabian Standards Organization standards for the lack of standard special Iraqi drinking bottled water. Evaluation criteria included the physical and chemical the following: pH, turbidity, total dissolved solids, total hardness, calcium, magnesium, sodium, fluoride, nitrates, sulfates, chlorides, iron, manganese. Results showing that the levels of water quality standards of domestic and imported brands were identical to different specifications with the exception of dissolved solids in the category of one domestic and one imported brand and fluoride in all varieties and types of domestic importers. Measurements also revealing explosives that have been selected for each item of different types of variation in the values of water standards explosives with rates ranging from 0-35% for domestic brand and 0-100% for imported brand and the values of most of the criteria listed on the packaging does not reflect the actual content of the water bombs.

Key words: Water bottled, environment, packaging, domestic, brands, nitrates

INTRODUCTION

The most important characteristic of bottled drinking water when compared it with tap water is the quality of bottled water, particularly in terms of taste and the regularity of water quality over time (Huerta-Saenz *et al*., 2012; Johnstone and Serret, 2012) and the tap water of the networks may change as a result of the materials they are exposed from the surrounding environment from reservoirs and pipes that pass in before they reach the consumer while the probability of contamination of bottled water is very low because it is placed in sealed packaging.

A 4 year study in the United States, reported by US Natural Resources Defense Council in which more than 1,000 packages covering 104 items of bottled drinking water were screened in some states, revealed that bottled water is not necessarily more pure or safe than tap water with about 33% of the bottled water at least one of them has different contaminants including some organic chemicals and bacteria at levels higher than permitted in the specifications of bottled drinking water and the results showed that about 25% of bottled water is actually a tap water that has been bottled after further treatment or without treatment, according to the survey, the main reason for the growing consumption of bottled drinking water in the US States is the marketing and promotional means used by some manufacturers to persuade the costumer of the purity and safety of bottled water, exploiting the anxiety and suspicions of people about the quality and safety of the water networks.

In Iraq, the drinking water industry, packed over the past 20 years has grown significantly and has a high production capacity and bottled water is available in different sizes, most of which are used for one time and items imported from different countries are available on the local market. This research aims to assess the water quality of five domestic produced bottled drinking water and ten imported bottled drinking. The assessment included the following comparison of results with international standards for bottled drinking water Comparing the quality of domestic varieties with the quality of imported items comparing the results with the water content on the packaging and assessment of the difference in water quality between packaging of each item.

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Abbreviations:

I	:	Imported
KSA	:	The Kingdom Saudi Arabia
NBOS	:	Number of Bottled Out of Standard
Mn	:	Manganese
Fe	:	Iron
Cl	:	Chloride
NO_3	:	Nitrate
F	:	Fluoride
Na	:	Sodium
So_4	:	Sulphates
Mg	:	Magnesium
$CaCO_3$	:	Calcium Carbonate
Ca	:	Calcium
TDS	:	Total Dissolved Solids
TU	:	Turbidity

MATERIALS AND METHODS

Samples collection method: The process of obtaining bottled drinking water packages from some of the city's shops randomly during the first half of 2018, taking into account the fact that the factory production date for packaging taken from the same packaging to avoid the possibility of a change in the characteristics of the water with the expiry of the packaging. The water quality analysis was performed according to APHA., (1995) Hussein (2017) and Hussein and Scholz (2017), unless stated otherwise. Two packages of each item were purchased and the physical and chemical characteristics were measured twice per item to confirm the accuracy of the measurement. The spectrophotometer DR 2800 Hach Lange (www.hach.com) was applied for the water quality analysis for variables NO_3-N, Fe, Cl, F, Na, SO_4, Mg, $CaCO_3$, Ca and Mn. Turbidity was determined with a Turbicheck Turbidity Meter (Lovibond Water Testing, Tintometer Group, Division Street, Chicago, IL, USA) (Al-Isawi *et al*., 2015 a, b; Al-Isawi *et al*., 2017). The pH for all samples was measured using a VARIO pH meter (Wissenschaftlich-Technische Werkstätten, Weilheim, Germany) (Al-Isawi *et al*., 2015a, b). This meter was calibrated with standard buffer solutions of pH 4, 7 and 9 each two weeks or whenever required. The Total Dissolved Solids (TDS) were measured using a Mettler Toledo Education Line Conductivity Meter (Boston Road, Leicester, UK). The packaging's included 10 imported items and 5 domestic items, Table 1, showing the names of the domestic and imported items studied as well as water quality, raw water sources and packing city.

In order to investigate statistically significant differences, the Shapiro-Wilk's test (Shapiro and Wilk, 1965; Razali and Wah, 2011) was used to assess the normality of data all data were non-normal distribution. A Mann-Whitney test was used to analyse non-normal data (Stoline, 1981; Kasuya, 2001). The Mann-Whitney tests were used to compare means between different treatments. Significant findings have been outlined and discussed. Somehow surprising and/or important insignificant findings have occasionally been highlighted as well.

Table 1: Locally produced and imported bottled water

Type of bottled water/ Name of bottled water	Source of water	Country of origin
Imported		
Muen	(n/a)[a]	KSA[b]
Alwadi	Fatima valley	KSA[b]
Zalal	(n/a)[a]	KSA[b]
Aquafina	(n/a)[a]	Kuwait
Abraj	(n/a)[a]	Kuwait
Faihaa	(n/a)[a]	KSA[b]
Raudhtain	Raudhtain field	Kuwait
Hania	(n/a)[a]	KSA[b]
Delta	(n/a)[a]	KSA[b]
Algdeer	(n/a)[a]	KSA[b]
Locally		
Sawa	(n/a)[a]	Iraq/Samawah
Alraya	(n/a)[a]	Iraq/Samawah
Razan	(n/a)[a]	Iraq/Karbala
Alkhazer	(n/a)[a]	Iraq/Karbala
Salsabeel	(n/a)[a]	Iraq/Karbala

[a]Not applicable; [b]The kingdom Saudi Aribia

RESULTS AND DISCUSSION

Domestic and imported water quality: Table 2 and 3 show the values of arithmetic average of the measurements of quality standards (intended as a statistical term) of two packaging water for each domestic and imported item, respectively indicating the values of the standards contained in the specifications of the bottled drinking water issued by the Saudi Arabian authority. The two tables also included the number of items that did not meet the required specifications. A code of one character and a number for each of the tested items was given to avoid mentioning the trade names, representing the character (L) of the domestic items and the character (I) of the imported items, Fig. 1 and 2 show the value of the arithmetic mean for elements reading of the domestic and imported items in case of high and low readings. A Mann-Whitney test (Nonparasmetric statistics) was used to determine whether the mean values of water standards for domestic varieties were statistically different from those in imported water.

pH: The pH values of the domestic varieties water varied from 7.1-7.8 (mean value was 7.34) and for the imported items from 7.2-7.8 (mean value was 7.48) and by comparing the results with Saudi Arabian Standard Specifications which determine the value of pH between 6.5 and 8.5, the researchers found that both the domestic and imported items conform to the specifications with a significant ($p>0.05$) differences between them.

Table 2: The quality measurements results of drinking water for the local items

	Element						
	Mn^1	Fe^2	Cl^3	$NO_3{}^4$	F^5	Na^6	$SO_4{}^7$
Symbols				mg L^{-1}			
$L_1{}^a$	0.00	0.010	20	4.10	0.600	20	31
L_2	0.00	0.000	36	9	0.095	49.4	47
L_3	0.00	0.015	187	4.30	1.100	50	210
L_4	0.00	0.000	16	0.115	0.300	13	48
L_5	0.01	0.010	197	1.100	1.150	22.5	48
KSA[b] standard	0.05	0.300	250	10	0.6-1	-	250
NBOS[c]	0.00	0.000	0	0.00	5.00		0

	Element					
	Mg^8	$CaCO_3{}^9$	Ca^{10}	TDS^{11}		
Symbols		mg L^{-1}			TU^{12}	pH^{13}
L_1	5.05	46	10	118	0.28	7.20
L_2	3.60	184	68	123	0.22	7.50
L_3	6.10	145	48	162	0.24	7.65
L_4	2.35	20	4.1	19.8	0.10	7.30
L_5	3.60	130	46	204	0.21	7.10
KSA standard	30.0	300	75	100-700	5.00	6.5-8.5
NBOS`	0.00	0	0	1.00	0.00	0.00

[a]Local; [b]The Kingdom Saudi Arabia; [c]Number of bottled out of standard; [1]Manganese; [2]Iron; [3]Chloride; [4]Nitrate; [5]Fluoride; [6]Sodium; [7]Sulphates; [8]Magnesium; [9]Calcium carbonate; [10]Calcium; [11]Total dissolved solids; [12]Turbidity

Table 3: The quality measurements results of drinking water for imported items

	Element						
	Mn^1	Fe^2	Cl^3	$NO_3{}^4$	F^5	Na^6	$SO_4{}^7$
Symbols	mg L^{-1}						
$I_1{}^a$	0.0005	0.010	20	4.1	0.6	20	31
I_2	0.0000	0.000	22	5	0.9	22	25.6
I_3	0.0	0.005	18	0.2	0.2	17	54
I_4	0.001	0.000	56	1.3	0.7	11	19
I_5	0.0	0.015	21	0.6	0.7	19	33
I_6	0.0	0.010	21	7.1	0.6	9	34
I_7	0.0	0.010	15	4.51	0.8	12	51.5
I_8	0.0	0.010	4.5	0.1	0.2	1	2.2
I_9	0.0	0.000	43	6	0.67	19	5.95
I_{10}	0.0	0.010	22.15	6.9	0.85	21	14
KSA[b] standard	0.05	0.300	250	10	0.6-1	-	250
NBOS[c]	0.00	0.000	0	0	2		0

	Element					
	Mg^8	$CaCO_3{}^9$	Ca^{10}	TDS^{11}	TU^{12}	pH^{13}
Symbols	mg L^{-1}					
$I_1{}^a$	5.05	46	10	118	0.28	7.2
$I_2{}^b$	7.7	45.5	5.5	105	0.23	7.2
$I_3{}^c$	19.5	105	10	120	0.18	7.8
$I_4{}^d$	10.55	101	23	111	0.1	7.65
$I_5{}^e$	1.8	50	22.5	112	0.22	7.4
$I_6{}^f$	6.45	121.5	38	159	0.26	7.7
$I_7{}^g$	13.4	95	16.5	114	0.115	7.2
$I_8{}^h$	5.55	50	10.7	50	0.23	7.65
$I_9{}^i$	17.6	110	15	139	0.4	7.3
$I_{10}{}^j$	8.5	75	16	120	0.16	7.7
KSA standard	30	300	75	100-700	5	6.5-8.5
NBOS	0	0	0	1	0	0

[a]Imported; [b]The Kingdom Saudi Arabia; [c]Number of bottled out of standard; [1]Manganese; [2]Iron; [3]Chloride; [4]Nitrate; [5]Fluoride; [6]Sodium; [7]Sulphates; [8]Magnesium; [9]Calcium carbonate; [10]Calcium; [11]Total dissolved solids; [12]Turbidity

Turbidity (TU): The range of turbidity for the domestic items was (0.1-0.24; 0.195 as a mean value) and for the imported items was (0.1-0.4; 0.217 as a mean value). Both of them were characterized by low levels of turbidity

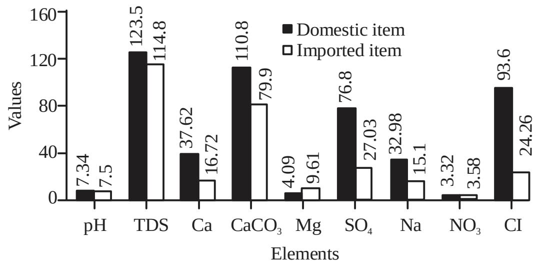

Fig. 1: Values of the arithmetic mean for elements reading of the domestic and imported items for high reading

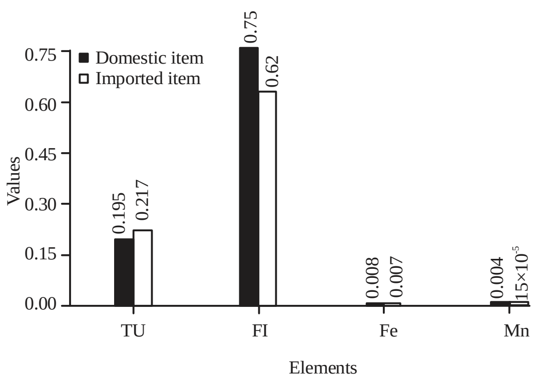

Fig. 2: Values of the arithmetic mean for elements reading of the domestic and imported items for low readings

when compared to the upper limit allowed in the specifications of the World Assembly for bottled drinking water and the United States Food and Drug Administration as shown in Table 2 and 3, respectively. There was no significant ($p<0.05$) differences between them as shown in Fig. 2.

Total Dissolved Solids (TDS): Concentration of total dissolved solids for domestic items ranged from (19.8-204, 123.5 mg L^{-1} as a mean value) and for imported items from (50-159, 114.8 mg L^{-1} as a mean value). This means that all measured concentrations are below the upper limit allowed in Saudi Arabian specifications, noting that there is a domestic item and an imported item below the minimum limit In Saudi Arabian specifications as shown in Table 2 and 3 and Fig. 1. There was no significant ($p<0.05$) differences between them.

Calcium (Ca): The concentration of the Calcium in domestic varieties ranged from (4.1-68 37.62 mg L^{-1}) as a mean value) and in items imported from 5.5-38, 16.72 mg L^{-1} as a mean value). These values did not exceed the upper limit allowed in Saudi Arabian specifications as shown in Table 2 and 3 and Fig. 1. There was no significant ($p<0.05$) differences between them.

Total hardness ($CaCO_3$): The concentration of total hardness in domestic varieties ranged between (20-184 , 110.8 mg L^{-1} as a mean value) as a calcium carbonate while the concentration of total hardness in imported varieties ranged between (45.5-121.5, 79.9 mg L^{-1}) as a mean value) as calcium carbonate as shown in Fig. 1 and by comparing the median values, the researchers found that the median value of the items Domestic (110.8 mg L^{-1}) higher than the median value of imported items (79.9 mg L^{-1}) by 38% with note that all domestic and imported items did not exceed the upper limit allowed in Saudi Arabian specifications as shown in Table 2 and 3. There was no significant ($p<0.05$) difference between them.

Magnesium (Mg): The concentration of magnesium in domestic varieties ranged between (2.35-6.1, 4.09 mg L^{-1} as a mean value) while the concentration in the imported varieties ranged from (1.8-17.6 9.61 mg L^{-1}) as a mean value) as shown in Fig. 1 with a significant ($p>0.05$) difference between them. All domestic and imported items are identical to Saudi Standard Specifications as they do not exceed the maximum allowable limit as shown in Table 2 and 3.

Sulphate (SO_4): Domestic varieties contained sulfate with concentrations of 31-210, 67.8 mg L^{-1} as a mean value) and imported items between (2.2-54, 27.03 mg L^{-1} as a mean value) as shown in Fig. 1 and the median value of the domestic items (67.8 mg L^{-1}) is approximately three times the median value of the imported items (27.03 mg L^{-1}) with no significant ($p<0.05$) difference between them. All domestic and imported items are identical to Saudi Standard Specifications as they do not exceed the maximum allowable limit as shown in Table 2 and 3.

Sodium (Na): Domestic sodium varieties contained a higher concentration of imported items with concentrations in domestic varieties ranging from (13-50, 32.98 mg L^{-1} as a mean value) while in imported items between (1-22 15.1 mg L^{-1} as a mean value) as shown in Fig. 1 with no significant ($p<0.05$) difference between them. Saudi standard specifications did not specify a value for the allowable level of sodium in bottled drinking water but Iraqi Standard Specifications for bottled drinking water stipulate that sodium concentration shall not exceed (200 mg L^{-1}).

Fluoride (F): Fluoride concentration in domestic varieties ranged between (0.095-1.15, 0.75 mg L^{-1}) as a mean value) and in imported items (0.2-0.9, 0.62 mg L^{-1} as a mean value) as shown in Fig. 2 with no significant ($p<0.05$) difference between them. It is clear that two imported items do not conform to Saudi Standard

Specifications while all domestic items are not matched Saudi Standard Specifications as shown in Table 3. Lewis, 1996 and Spellman, 2014 demonstrated that drinking water containing fluoride with a concentration of 1.0 mg L^{-1} helps in reducing the incidence of tooth decay by 65% for children between 12-15 years while increasing fluoride concentration more than 2 mg L^{-1} the grey or black spots appear on the children's permanent fingers are accompanied by a necrosis of the tooth and this is known as fluorosis. Furthermore, if fluoride concentration in drinking water is more than (8.0 mg L^{-1}) this leads to hardening of cartilage and bones because of its accumulation, leading to hardening of the spine and difficulty bending.

Nitrate (NO_3): Nitrate concentrations in domestic varieties ranged from (0.115-9, 3.32 mg L^{-1} as a mean value), higher than nitrate concentration in imported items (0.1-9, 3.58 mg L^{-1} as a mean value) as shown in Fig. 2 with no significant ($p<0.05$) difference between them. The nitrate concentration in the domestic and imported varieties did not exceed the upper limit allowed in Saudi Arabian specifications as described in Table 2 and 3, respectively.

Chlorides (Cl): Concentrations of chlorides in domestic varieties are higher than in imported items where the concentration in domestic varieties ranged between (16-197,93.6 mg L^{-1} as a mean value) and imported items between (4.5-56, 24.26 mg L^{-1} as a mean value) as shown in Fig. 1 with no significant ($p<0.05$) difference between them. The concentration of chlorides in both of domestic and imported items did not exceed the upper limit allowed in Saudi Standard specifications as shown in Table 2 and 3, respectively.

Iron (Fe): The concentration of iron in the domestic varieties ranged between (0.0-0.015, 0.004 mg L^{-1} as a mean value) and in the imported items between (0.0-0.015, 0.0002 mg L^{-1} as a mean value) as shown in Fig. 2 with a significant ($p<0.05$) difference between them. All domestic and imported items did not exceed the upper limit allowed in Saudi specifications as shown in Table 2 and 3, respectively.

Manganese (Mn): Domestic varieties, the manganese concentration ranged between (0.0-0.01, 0.004 mg L^{-1} as a mean value) and in the imported items between (0.0-0.001, 0.00015 mg L^{-1} as a mean value) as shown in Fig. 2, noting that the median value of the domestic items (0.004 mg L^{-1}) is equivalent to about two and a half times the median value of the imported items (0.00015 mg L^{-1}) with a significant ($p<0.05$) difference between them. All domestic and imported items did not exceed the maximum allowable limit in Saudi Arabian specifications as shown in Tables 2 and 3, respectively.

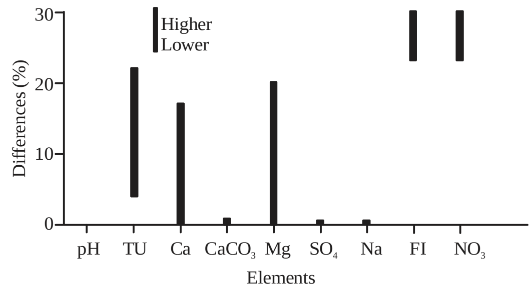

Fig. 3: Percentages difference for each element in case of domestic packaging

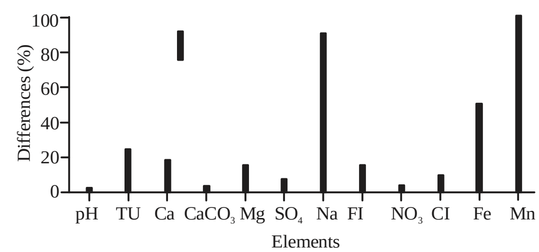

Fig. 4: Percentages difference for each elements in case of imported packaging

Comparison of the quality of the packaging water for one element: Laboratory tests of the two items tested for each of the different domestic and imported bottled water showed that the test results for some of the elements (from 5-10 variables) were equal in the two packages while the other elements differed in varying proportions ranging from (0-35%) in case of domestic items and (0-100%) in case of imported items as shown in Fig. 3 and 4, respectively. In the domestic varieties, the percentages difference between the two packaging for the pH, TU, Ca, Hardness, Mg, SO_4, Na, Fl and NO_3 were 0-2, 4-22, 0-17, 0-1, 0-20, 0-0.5, 0-0.5, 10-35 and 23-30, respectively. While laboratory tests for TDS, Cl, F and MN did not record any percentages of difference . In case of imported items, the percentages difference between The two packaging of pH, TU, TDS, Ca, Hardness, Mg, SO_4, Na, Fl, NO_3, Cl, F and Mg were 0-3, 0-25, 0-3, 0-18, 0-4, 0-15, 0-8, 0-90, 0-16, 0-4, 0-10, 0-50 and 0-100, respectively.

Comparison of the quality of water packaged with the content on the packaging: Of the 13 quality standards carried out, two of them did not mention any concentration in the domestic and imported varieties, namely, TU and Mn. All domestic and two of imported varieties did not mention Fl concentration. Total hardness was not mentioned in Four domestic and six imported items. the nitrate concentration was not mentioned in four domestic varieties and two items of imported. TDS was

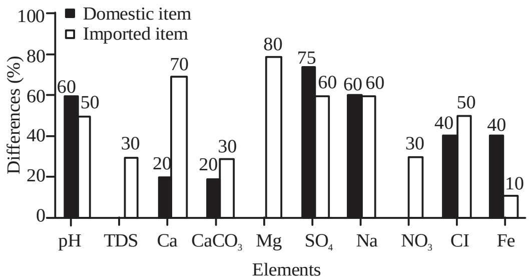

Fig. 5: Comparison of the percentage of domestic and imported varieties that exceeded the mentioned concentrations in their packaging for each element

not mentioned in three domestic varieties and two imported items. Iron concentration was not mentioned in three domestic varieties and four imported items. Figure 5 compare the proportion of domestic and imported bottled water that have exceeded their packaging for element.

CONCLUSION

Domestic and imported items have achieved good quality in all physical and chemical standards except for two criteria, namely, total dissolved solids and fluoride where one domestic item and one imported item did not achieve the level of concentration of dissolved solids and all domestic items were not specifications are achieved in fluoride concentration while two imported items did not achieve the specifications in fluoride concentration.

Statistical analysis using the Mann-Whitney test at the significance level 0.05, the quality of the domestic varieties is no different from the quality of the imported items in the following criteria: turbidity, dissolved solids, total hardness, calcium, sodium, fluoride, nitrate, sulphate and chlorides, the quality varies in the following criteria: pH, magnesium, iron and manganese.

There is a variation in the water quality of the same item as the measurement of the two packaging's tested for each item revealed a difference in the values of the standards in varying proportions ranging (0-35%) for domestic items and (0-100%) for imported items which indicates the irregularity of the efficiency of the water purification processes in the factory, the difference in the quality of the raw water over time and the multiplicity of water sources of one factory.

There is a significant discrepancy between the values of the measured standards and their stated values on the packaging's except for dissolved solids, magnesium and nitrates in domestic varieties and iron in imported items.

RECOMMENDATIONS

Some domestic items need to be added fluoride and others need to be reduced in fluoride either for imported items, some of which need to be increased fluoride.

Relevant agencies must take their oversight role from monitoring all stages of packaged water factories and their water sources and carrying out periodic checks on the water produced from these factories for domestic items. Imported items must be subject to laboratory tests from time to time to ensure that they conform to standard specifications.

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Environmental Science and Pollution Research (2018) 25:6870–6889
https://doi.org/10.1007/s11356-017-0992-0

RESEARCH ARTICLE

Treatment of artificial wastewater containing two azo textile dyes by vertical-flow constructed wetlands

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Received: 10 July 2017 / Accepted: 10 December 2017 / Published online: 21 December 2017

Abstract

The release of untreated dye textile wastewater into receiving streams is unacceptable not only for aesthetic reasons and its negative impacts on aquatic life but also because numerous dyes are toxic and carcinogenic to humans. Strategies, as of now, used for treating textile wastewaters have technical and economical restrictions. The greater part of the physico-chemical methods, which are used to treat this kind of wastewater, are costly, produce large amounts of sludge and are wasteful concerning some soluble dyes. In contrast, biological treatments such as constructed wetlands are cheaper than the traditional methods, environmental friendly and do not produce large amounts of sludge. Synthetic wastewater containing Acid Blue 113 (AB113) and Basic Red 46 (BR46) has been added to laboratory-scale vertical-flow construction wetland systems, which have been planted with *Phragmites australis* (Cav.) Trin. ex Steud. (common reed). The concentrations 7 and 208 mg/l were applied for each dye at the hydraulic contact times of 48 and 96 h. Concerning the low concentrations of BR46 and AB113, the unplanted wetlands are associated with significant ($\rho < 0.05$) reduction performances, if compared with planted wetlands concerning the removal of dyes. For the high concentrations of AB113, BR46 and a mixture of both of them, wetlands with long contact times were significantly ($\rho < 0.05$) better than wetlands that had short contact times in terms of dye, colour and chemical oxygen demand reductions. Regarding nitrate nitrogen (NO_3-N), the reduction percentage rates of AB113, BR46 and a mixture dye of both of them were between 85 and 100%. For low and high inflow dye concentrations, best removals were generally recorded for spring and summer, respectively.

Keywords Acid Blue 113 · Basic Red 46 · Chemical oxygen demand · Common reed · Environmental pollution control · Textile wastewater

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s11356-017-0992-0) contains supplementary material, which is available to authorized users.

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Abbreviations

AB	Acid blue
ABSA	3-Aminobenzenesulfonic acid
ANOVA	Analysis of variance
ANSA	5-Amino-8-(phenylamino)naphthalene-1-sulfonic acid
AO	Acid orange
AY	Acid yellow
BR	Basic red
CASRN	Chemical Abstracts Service Registry Number
COD	Chemical oxygen demand
DAN	1,4-Diaminonaphthalene
DO	Dissolved oxygen
DY	Disperse yellow
EC	Electric conductivity
HF	Horizontal-flow
N	Nitrogen
N/A	Not applicable

NBNMA	*N*-Benzyl-*N*-methylaniline
NBNMD	*N*-Benzyl-*N*-methylbenzene-1,4-diamine
NH_4-N	Ammonia nitrogen
NO_2-N	Nitrite nitrogen
NO_3-N	Nitrate nitrogen
PO_4–P	Ortho-phosphate-phosphorus
RB	Reactive black
SD	Standard deviation
SE	Standard error
TDS	Total dissolved solids
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorus
TSS	Total suspended solids
VF	Vertical-flow
VY	Vat yellow
λ_{max}	Maximum absorbance

Introduction

Textile dyeing processes are one of the most environmental-unfriendly industrial processes, because the reagents used are very rich in chemical compounds comprising both inorganic and organic products (Juang et al. 1996; Robinson et al. 2001). Furthermore, the presence of colour in the effluent textile wastewater is one of the most important problems. Sultana (2014) stated that coloured wastewaters, produced from dyeing processes, are heavily polluted with chemicals, textile auxiliaries and dyes. The properties of textile wastewater depend on the production, technology and chemicals used (Wang et al. 2011).

Textile industries devour gigantic amounts of water and generate vast volumes of wastewater through different steps in the dyeing and finishing processes, and the discharged wastewater is an overwhelming blend of various polluting substances such as organic, inorganic, elemental and polymeric products (Babu et al. 2007; Kant 2012). Dye wastes are the most dominating materials in textile wastewater, and these materials are often toxic to the biological world as well as the dark colour of some of these materials blocking sunlight, which causes acute problems in biological communities (Ratna and Padhi 2012; Dey and Islam 2015).

The use of constructed wetlands in azo textile dye wastewater treatment is still at an experimental stage (Nawab et al. 2016). Although many researchers investigated the performance of constructed wetlands to treat textile wastewater in terms of dye, chemical oxygen demand (COD), phosphorus and nitrogen reductions, all corresponding results related to short-term operation and the data rarely covered all seasons (Table 1).

Two azo textile dyes [Acid Blue 113 (AB113) and Basic Red 46 (BR46)] were selected in this research with two different concentrations: low with a target concentration of 5 mg/l and high with a target concentration of 200 mg/l. Typically, textile industry-processing effluents contain dyes in the range between 10 and 200 mg/l (Lavanya et al. 2014). Most textile dyes can be detected at a rather low concentration of even < 1 mg/l by the human eye (Chung 1983; Lavanya et al. 2014; Pandey et al. 2007). Furthermore, Van der Zee (2002) stated that algal growth was not inhibited at dye concentrations < 1 mg/l. Both of which are commercial dyes, which are extensively used in the textile industry (Chung et al. 1992; Riu et al. 1997; Pervez et al. 1999; Olgun and Atar 2009; Ong et al. 2010; Deniz and Karaman 2011; Deniz and Saygideger 2011). AB113 is an acid dye, and BR46 is a basic dye. An acid dye is defined as a negatively charged dye at a chemical level, which contains one or more acidic groups such as a sulfonic group (Akbari et al. 2002; Martínez-Huitle and Brillas 2009). A basic dye is defined as a positively charged stain at a chemical level (Martínez-Huitle and Brillas 2009; Brillas and Martínez-Huitle 2015), which means it reacts well with negatively charged materials (Sun and Yang 2003).

The aim of this project is to evaluate the effectiveness of vertical-flow constructed wetlands in reducing azo textile dyes contaminated with artificial wastewater, aromatic amines and other water quality variables including COD and ortho-phosphate-phosphorus (PO_4-P). The corresponding objectives are to assess (a) the role of plants in reducing azo textiles within artificial wastewater, (b) the influence of the mixture of both of these two dyes on the performance of vertical-flow constructed wetlands, (c) the ability of this type of constructed wetland to reduce aromatic amines and (d) the influence of seasonal variation and operational parameters such as resting and contact times on dye reduction.

Materials and methods

Wetland set-up and operation

The research has been performed between 1 June 2016 and 31 May 2017. This system has been used for treating azo textile dye wastewater since 1 May 2015 (Hussein and Scholz 2017). The constructed wetlands have been located within a university greenhouse (Supplementary Material 1) and operated to treat artificial wastewater treating two azo textile dyes. The rig consisted of 18 vertical-flow constructed wetlands. Wastewater drained vertically to enhance aerobic biodegradation of nitrogen and organic matter (Fuchs 2009). The experiment evaluates the wetland performance by simulating processes occurring within large-scale reed beds. The filters were located at random within the system set-up. Resting and contact times as well as hydraulic loading rate impacts on dye removal were evaluated. The period of time when a wetland is empty (no liquid inside) is known as resting time, while contact time is known as the duration of the

Table 1 Previous studies (listed in order of date) on textile wastewater treatment by constructed wetlands

Dye used	Type of wetland	Design characteristics	Plants used	Removal performance	Duration (days)	Country of operation	References
AB113, *RB171	VF	Gravel-sand	*P. australis*	98% colour	70	USA	Pervez et al. (1999)
AO7	VF	Gravel-sandy clay soil	*P. australis*	74% colour, 64% COD and 71% TOC	77	Portugal	Davies et al. (2005)
AO7	VF	Gravel-sandy clay soil	*P. australis*	99% colour, 93% COD and TOC	48	Portugal	Davies et al. (2006)
Various dyes in real wastewater	HF	Gravel-sand	*Typha* and *cocoyam*	77% colour, 72% COD and 59% sulfate	84	Tanzania	Mbuligwe (2005)
**RB5, DY211, VY46	VF-HF	Gravel-sand-tuff	*P. australis*	90% colour, 84% COD, 93% TSS, 52% TN, 87% $N_{organic}$, −331% NH_4-N, 88% sulfate, 80% anion surfactant and 93% TSS	60	Slovenia	Bulc and Ojstršek (2008)
RR22, VR13, **RB5	VF	Gravel-sand-zeolite-peat	Without plant	70% dye, 60% EC, 88% COD and TOC	90	Slovenia	Ojstršek et al. (2007)
AO7	UF	Gravel-glass beads	*P. australis*	98% dye, 90% COD, 67% TN, 28% TP, 98% NH_4-N, 100% NO_3-N	365[a]	Japan	Ong et al. (2010)
RR141	VF	Gravel-sand	*Typha*	49% colour, 60% COD, 86% TDS		Thailand	Nilratnisakorn et al. (2009)
AO7	VF	Gravel-sludge	*P. australis*	94% colour, 95% COD and 86% NH_4-N	27	N/A	Ong et al. (2011)
	FWS-SSF SSF-FWS	Shale	*P. australis*	98% COD, 97%, colour 91% COD, 99% colour		Thailand	Cumnan and Yimrattanabovorn (2012)
Mixture dyes into different metabolites	VF	Coconut shavings-soil with bacteria	*Gaillardia pulchella*	70% COD, 74% TOC, 70 BOD	0.042	India	Kabra et al. (2013)
Mixture dyes into different metabolites	VF	Coconut shavings-sand-gravel-soil with bacteria	*Portulaca grandiflora*	59% COD, 38% BOD, 37% TOC, 41% turbidity, 71% TDS, 60% TSS	0.05	India	Khandare et al. (2013)
Various dyes in real wastewater	VF	Coconut shavings-gravel-sand-soil	*Typha*	79% COD, 77% BOD, 59% TDS, 27% TSS	3	Pakistan	Shehzadi et al. (2014)
AY 2G E107	VF	Gravel-sand-zeolite	*Canna* and *Typha*	95% colour, 64% COD, 94% PO_4-P, 77% NH_4-N	90	Turkey	Yalcuk and Dogdu (2014)

AB acid blue, **RB* reactive blue, *VF* vertical-flow, *AO* acid orange, *COD* chemical oxygen demand, *TOC* total organic carbon, *HF* horizontal flow, ***RB* reactive black, *DY* disperse yellow, *VY* vat yellow, *TSS* total suspended solid, *TN* total nitrogen, *N* nitrogen, *NH_4-N* ammonium nitrogen, *RR* reactive red, *VR* vat red, *EC* electrical conductivity, *UF* upper flow, *NO_3-N* nitrate nitrogen, *TDS* total dissolved solids, *FWS* free water surface, *SSF* subsurface flow, *BOD* biochemical oxygen demand, *AY* acid yellow, *PO_4-P* ortho-phosphate-phosphorus, *N/A* not applicable

[a] The experimental work was under control condition (indoor)

wastewater when it is in touch with the aggregates and/or plants in the system.

In this study, artificial wastewater containing two azo dyes (BR46 and AB113) was assessed at the concentrations of 7 and 208 mg/l for the contact times of 48 and 94 h with respect to their impact on the constructed wetland performance. All artificial wastewater chemicals (Wießner et al. 2005; Ong et al. 2009) were bought from the Scientific Laboratory Supplies (Wilford Industrial Estate, Wilford, Nottingham, UK). Details of each dye and the composition of artificial wastewater including its chemical concentrations used in the experimental work are shown in Supplementary Material S1 and Table 2, respectively. BR46 has a maximum absorbance (λ_{max}) of 530 nm (Khataee 2009) and was sourced from DyStar (Am Prime Park, Raunheim, Germany). AB113 had a λ_{max} of 566 nm (Shirzad-Siboni et al. 2014) and was purchased from Sigma-Aldrich (The Old Brickyard, New Road Gillingham, Dorset, UK). Both dyes were used without further purification. The wavelength for the maximum absorbance of the dye mixture had been determined experimentally by using a WPA Biowave II Spectrophotometer (Biochrom, Cambourne Business Park, Cambourne, Cambridge, UK). At first, λ_{max} of the mixed dye was determined by scanning the absorption of different dye mixture concentrations for wavelengths between 300 and 800 nm. The λ_{max} for the mixed dye was found to be 511 nm.

Plastic drainage pipes were used for wetland construction (Supplementary Material 1). All 18 wetlands had heights of 100 cm and diameters of 10 cm. All wetlands were filled to 90 cm with washed gravel, applying two layers of aggregates. Large gravel (diameter; 10–20 mm) was used at the bottom, preventing clogging. Pea gravel (diameter; 5–10 mm) was at the top of each wetland. The outlet valves were at the centre of the bottom plate of each filter.

All wetlands contained *Phragmites australis*, which was monitored for health and growth. Dead plants were cut to about 13 cm in terms of height. The corresponding cuttings were recycled within the filters.

The aquatic fertiliser TNC Complete was purchased from TNC Limited (Spotland Bridge Mill, Mellor Street, Rochdale, UK) and applied in the experimental research as a nutrient for the plants and microorganisms. The associated key ingredients were phosphorus (0.2%), nitrogen (1.5%), iron (0.08%), manganese (0.018%), potassium (5%), magnesium (0.08%), copper (0.002%), molybdenum (0.001%), boron (0.01%) and zinc (0.01%). TNC Complete also provides ethylenediaminetetraacetic acid (EDTA) that is a source of the elements copper, iron, manganese and zinc. One millilitre of fertiliser was added to 10 l of tap water.

The packing order of the experimental constructed wetland set-up treating artificial wastewater containing two azo textile dyes is shown in Table 3. All wetlands were filled with the same washed gravel.

Analytical methods and equipment

Measurement of physical parameters

The physical parameters included dye concentration, colour, total suspended solids (TSS), dissolved oxygen (DO), turbidity, pH, redox potential, electric conductivity (EC) and temperature. Dye concentration, colour and TSS were measured by the spectrophotometer Hach Lange DR2800 (Pacific Way, Salford, UK). Dye concentrations were quantified through a selective wavelength at maximum absorbance for each dye. Colour was measured using a unit Pt/Co scale. The TSS were measured in milligrams/litre. Samples were filtered by using Whatman grade 1 qualitative filter paper (standard grade; circle, 320 mm), which was bought from the Scientific Laboratory Supplies (Wilford Industrial Estate, Wilford, Nottingham, UK).

The DO was estimated using a Hach Lange HQ30D Flexi Meter (Pacific Way, Salford, UK) promptly after taking samples. Turbidity (NTU) was measured by using a TurbiCheck Portable Turbidity Meter (Lovibond Water Testing, Tintometer Group, Division Street, Chicago, IL, USA). The pH (−) and redox potential (mV) were determined by applying a portable WTW VARIO pH meter (Wissenschaftlich-Technische Werkstätten, Weilheim, Germany). The

Table 2 Details of artificial wastewater compositions use in the experimental work

Material	Chemical structure	Molecular weight (g/mol)	CAS number	Purity of dye (%)	Concentration (mg/l)
Sodium acetate anhydrous pure	CH_3COONa	82.03	127-09-3	≥99	107.1
Sodium benzoate	C_6H_5COONa	144.11	532-32-1	≥99	204.9
Ammonium nitrate pure	NH_4NO_3	80.04	6484-52-2	≥99	76.1
Sodium chloride pure	NaCl	58.44	7647-14-5	≥99	7.0
Magnesium chloride hexahydrate	$MgCl_2 \cdot 6H_2O$	203.30	7791-18-6	≥99	3.4
Calcium chloride dehydrate	$CaCl_2 \cdot 2H_2O$	147.01	10035-04-8	≥99	4.0
Potassium phosphate dibasic trihydrate	$K_2HPO_4 \cdot 3H_2O$	228.22	16788-57-1	≥99	36.7

CAS Chemical Abstracts Service, *C* carbon, *Cl* chlorine, *H* hydrogen, *K* potassium, *Mg* magnesium, *N* nitrogen, *Na* sodium, *O* oxygen, *P* phosphorus equipment was calibrated with standardised buffer solutions of pH 4, 7 and 9, whenever required. The acceptable range of pH is from 6.5 to 9 (Boyd and Gautier 2000).

The EC (μS/cm) was determined applying a portable Mettler Toledo Education Line Conductivity Meter (Boston Road, Leicester, UK). Although EC itself is not of aquatic or human health concern, its value gives an indication, if there is any other water quality problem. A sudden increase in EC values indicates that there is a source of dissolved ions in the wetland filter (Kumar and Chopra 2012). Furthermore, the site temperature was noted each day, applying a thermometer which was located alongside the wetland filters.

Measurement of chemical parameters

The chemical parameters included COD, ammonia nitrogen (NH_4-N), nitrate nitrogen (NO_3-N), PO_4-P and amines. The spectrophotometer Hach Lange DR2800 was applied for the water quality analysis for parameters such as COD, PO_4-P, NO_3-N and ammonium nitrogen (NH_4-N) with milligrams per litre (mg/l) as a unit. The aromatic amines were measured as absorbance by using a WPA Biowave II UV/visible spectrophotometers (Cambourne, Cambridge, UK). Specific wavelengths for the absorbance of every type of aromatic amine exist. Also, samples were filtered by using a specific filter paper (Whatman grade 1 qualitative filter paper, standard grade, circle, 320 mm). The water quality analysis was performed according to APHA (1995), if not clarified otherwise. Liquid samples were taken between 10:00 and 11:00 a.m.

The Shapiro-Wilk test (Shapiro and Wilk 1965; Razali and Wah 2011) was applied to judge data normality. A one-way analysis of variance (ANOVA) test was performed with the help of the Statistical Package for the Social Sciences software to analyse normally distributed data. The Mann-Whitney test was applied to evaluate non-normal data (Stoline 1981; Kasuya 2001). The ANOVA and Mann-Whitney tests compared averages between various treatments (e.g. Table 4).

Table 3 Packing order of the experimental constructed wetland set-up treating artificial wastewater containing two azo textile dyes

Wetland number	Plants	Dye			Resting time (h)	Contact time (h)
		Type	Mean (mg/l)	SD		
1	No	BR46	6.15	0.75	2	94
2	No	AB113	7.50	1.66	2	94
3	Yes	AB113	7.50	1.66	2	94
4	Yes	Mix	0.153		2	94
5	Yes	BR46	6.15	0.75	2	94
6	Yes	Mix	0.153		2	94
7	Yes	BR46	6.15	0.75	48	48
8	Yes	Mix	0.153		48	48
9	Yes	AB113	7.50	1.66	48	48
10	Yes	Mix	0.153		48	48
11	Yes	BR46	206	9.60	48	48
12	Yes	Mix	5.331		48	48
13	Yes	AB113	207	13.70	48	48
14	Yes	Mix	5.331		48	48
15	Yes	BR46	206	9.60	96	96
16	Yes	Mix	5.331		96	96
17	Yes	AB113	207	13.70	96	96
18	Yes	Mix	5.331		96	96

Mix, mixture between BR46 and AB113, and the reading is in a wavelength

SD standard deviation, *BR* basic red, *AB* acid blue

Results and discussion

Test of normality for plant and liquid samples

Test of normality findings concerning the dimensions of *P. australis* and effluent water quality variables is shown in Supplementary Material S3.

Table 4 Application of the statistical wetland filter set-up design (Table 3) to assess the impact of individual key variables

Comparison of two wetland systems with each other		Impact to be assessed
First wetland with number	Second wetland with number	
1	2	Difference between BR46 and AB113
1	5	*Phragmites australis* on BR46
2	3	*Phragmites australis* on AB113
4	6	Mixing dyes (low concentration)
5	7	Decrease in contact time (or increase in resting time on BR46)
3	9	Decrease in contact time (or increase in resting time on AB113)
7	9	Difference between BR46 and AB113
8	10	Mixing dyes (low concentration)
7	11	Increased BR46 concentration
9	13	Increased AB113 concentration
12	14	Mixing dyes (high concentration)
11	15	Increased contact and resting times
13	17	Increased contact and resting times
11	13	Difference between BR46 and AB113
15	17	Difference between BR46 and AB113
16	18	Mixing dyes (high concentration)

BR basic red, *AB* acid blue

Plant growth assessment

Plants became yellow in winter. Dead plant parts were cut and recycled within the wetlands (Stefanakis et al. 2014). Plants subjected to the dye AB113 developed well compared to those linked to BR46. Plants for systems with long contact time grew better than those plants associated with short time (Table 5). These findings support similar ones by Pagter et al. (2005).

Regarding plant growth, there was a significant ($\rho < 0.05$) difference concerning the length and diameter at low and high AB113 concentrations (wetlands 3, 9, 13 and 17). Concerning plant growth at the presence of BR46, significant ($p < 0.05$) differences for the length and diameter at the low dye concentrations were recorded (wetlands 5 and 7). No significance ($p > 0.05$) for either parameter was noted for the high dye concentrations (wetlands 11 and 15). In case of the mixed dye, there was no significant ($p > 0.05$) difference regarding the length at the low and high dye concentrations (wetlands 4, 6, 8, 10, 12, 14, 16 and 18). While with respect to the plant diameter, there was no significant ($p > 0.05$) difference for wetlands 12 and 14 (high concentration).

Redox potential and dissolved oxygen

Redox potentials above 100 mV are linked to aerobic environments. In comparison, values below − 100 mV highlight anaerobic boundary conditions (Suthersan 2001). The DO is an important parameter in constructed wetlands, since it is essential for aerobic respiration for microorganisms and it regulates the oxidation-redox potential in wastewater (Boyd 2000). Wu et al. (2011b) and Hou et al. (2016) highlighted that the main pathways for oxygen transfer in constructed wetlands such as the system in this research (tidal flow) are wetland macrophytes releasing oxygen via their roots, contact transfer at the interface of biofilm and atmosphere and DO associated with influent wastewater. In case of low concentration, redox potential values (Table 6) for the effluent of BR46, AB113 and the mixture of both of them were in the range between − 34 and − 64 mV, and for the effluent high concentrations, the values were in the range between − 56 and − 95 mV. These results show dye degradation, regardless of aerobic and anaerobic conditions. Regarding DO for both dyes (BR46 and AB113), the lowest effluent values (Table 6) were noted for planted wetlands 5 and 3 (low resting and high contact times of 2.97 and 3.37 mg/l, respectively) when compared with the unplanted wetlands 1 and 2 and planted wetlands 7 and 9 (high resting time and low contact time), respectively. Concerning the mixture between the two dyes, the value of DO for wetlands 4 and 6 (low resting and high contact times) was lower than that for wetlands 8 and 10 (high resting and low contact times) as a result of the higher contact time leading to consumption of more DO by the microbial community. The same findings for the DO between

Table 5 Dimensions of *Phragmites australis* (Cav.) Trin. ex Steud. (common reed) planted in the experimental wetlands

Dye	Wetland number	Number of stems	Characteristics					
			Length (cm)			Diameter (mm)		
			Minimum	Maximum	Mean ± SD	Minimum	Maximum	Mean ± SD
BR46	5	68	69	142	108 ± 17.8	1.1	3.1	2.2 ± 0.58
	7	20	60	122	93 ± 16.9	0.8	2.3	1.1 ± 0.39
	11	17	45	90	67 ± 11.2	0.8	1.1	0.9 ± 0.08
	15	14	44	90	60 ± 12.9	0.8	1.4	1.0 ± 0.16
AB113	3	40	79	140	107 ± 18.0	1.0	3.4	2.1 ± 0.63
	9	15	55	98	79 ± 14.8	0.8	1.2	1.0 ± 0.11
	13	20	45	98	70 ± 16.3	0.7	2.9	1.9 ± 0.59
	17	10	29	56	41 ± 9.1	0.7	1.6	1.1 ± 0.26
Mixture of BR46 and AB113	4	48	78	142	109 ± 15.9	1.0	3.9	2.4 ± 0.73
	6	55	70	134	109 ± 16.2	1.1	3.7	2.3 ± 0.59
	8	7	85	101	94 ± 5.5	0.9	2.0	1.5 ± 0.39
	10	10	80	110	95 ± 8.2	1.1	2.3	1.8 ± 0.37
	12	10	45	87	64 ± 11.3	0.8	1.6	1.0 ± 0.22
	14	16	46	80	66 ± 9.7	1.3	3.1	2.1 ± 0.48
	16	7	45	61	54 ± 5.8	0.8	1.9	1.3 ± 0.37
	18	9	44	67	58 ± 7.5	1.0	2.9	1.8 ± 0.63

BR basic red, *AB* acid blue, *SD* standard deviation

Table 6 Inflow and outflow water quality characteristics for general physical and chemical variables related to different wetlands

Dye	Type of flow	Wetland number	No. of samples	pH			Redox potential (mV)			Dissolved oxygen (mg/l)			Total suspended solids (mg/l)			Turbidity (NTU)			Electric conductivity (μS/cm)		
				Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD	Min.	Max.	Mean ± SD
BR46	In	N/A	82	7.22	7.77	7.47 ± 0.17	−56	−41	−47.72 ± 3.74	8.64	9.66	9.24 ± 0.22	1	2	1.07 ± 0.26	2.01	6.13	3.91 ± 0.85	545	575	560 ± 9.4
	Out	1	82	7.37	8.14	7.72 ± 0.23	−67	−49	−55.98 ± 3.81	2.13	5.51	3.73 ± 0.92	0	8	4.48 ± 1.56	3.43	6.61	5.24 ± 0.69	451	558	483 ± 18.0
	Out	5	82	6.94	7.36	7.17 ± 0.08	−45	−26	−35.29 ± 4.31	2.01	4.06	2.97 ± 0.46	2	45	10.68 ± 9.52	4.11	76.5	11.81 ± 17.02	487	670	644 ± 35.8
	Out	7	82	7.08	8.32	7.57 ± 0.42	−51	−30	−41.80 ± 4.74	2.38	5.21	3.71 ± 0.66	0	6	2.73 ± 1.45	3	6.53	4.07 ± 0.77	336	421	397 ± 16.6
	In	N/A	81	6.79	7.25	6.94 ± 0.14	−24	−14	−19.43 ± 2.48	8.8	9.74	9.21 ± 0.18	35	56	45.01 ± 4.51	12.6	15.9	14.62 ± 0.63	685	765	735 ± 13.5
	Out	11	81	7.16	8.01	7.49 ± 0.24	−97	−38	−56.28 ± 14.03	2.58	4.73	3.65 ± 0.46	16	46	23.68 ± 5.67	7.23	27.1	13.11 ± 3.43	563	684	604 ± 16.1
	Out	15	41	7.81	8.21	7.98 ± 0.09	−87	−67	−72.90 ± 4.22	2.41	4.01	3.03 ± 0.36	11	32	14.95 ± 3.19	8.04	23.4	12.09 ± 3.94	559	644	581 ± 16.6
AB113	In	N/A	82	7.19	7.57	7.35 ± 0.08	−59	−39	−44.85 ± 3.21	8.41	9.89	9.40 ± 0.35	4	8	5.87 ± 1.04	5.06	6.71	7.51 ± 0.42	521	575	543 ± 12.9
	Out	2	82	7.37	7.72	7.59 ± 0.07	−66	−50	−60.71 ± 3.08	2.42	5.57	4.14 ± 0.78	3	12	7.96 ± 1.72	3.91	6.99	5.37 ± 0.87	468	601	559 ± 22.5
	Out	3	82	7.21	7.77	7.57 ± 0.16	−71	−46	−64.24 ± 4.90	2.62	4.76	3.37 ± 0.54	2	22	6.71 ± 4.04	4.86	10.49	7.03 ± 1.18	497	622	583 ± 21.5
	Out	9	82	7.26	7.71	7.51 ± 0.07	−63	−44	−57.90 ± 3.42	2.36	5.45	3.59 ± 0.79	0	7	3.16 ± 1.28	2.87	6.42	4.59 ± 0.78	408	511	484 ± 16.4
	In	N/A	81	7.99	8.17	8.07 ± 0.04	−90	−75	−82.12 ± 4.27	9.00	9.51	9.31 ± 0.11	97	126	110 ± 8.59	42.31	62.34	51.79 ± 3.95	855	892	870 ± 5.6
	Out	13	81	7.83	8.29	8.16 ± 0.12	−102	−74	−92.24 ± 6.78	1.95	5.21	4.07 ± 0.61	37	78	62.64 ± 8.47	9.95	34.21	24.21 ± 6.61	702	909	765 ± 51.3
	Out	17	41	7.80	8.31	8.15 ± 0.16	−106	−78	−95.27 ± 8.27	1.98	4.41	3.62 ± 0.73	40	97	58.29 ± 14.40	12.45	25.41	18.62 ± 4.24	672	899	742 ± 60.8
The mixture of the dyes	In	N/A	82	7.2	7.39	7.32 ± 0.03	−52	−35	−45.67 ± 3.01	8.78	9.62	9.36 ± 0.19	6	10	8.32 ± 0.89	4.23	5.92	5.14 ± 0.34	510	535	522 ± 6.2
	Out	4	82	7.01	7.49	7.29 ± 0.09	−46	−28	−40.62 ± 3.02	1.67	4.47	3.12 ± 0.59	7	62	12.99 ± 10.90	3.85	64.5	10.67 ± 10.95	478	586	508 ± 14.2
	Out	6	82	6.78	7.18	7.09 ± 0.07	−39	−17	−33.61 ± 3.06	2.21	4.01	2.98 ± 0.44	1	72	11.40 ± 17.02	4.14	87.4	11.67 ± 17.43	475	568	491 ± 13.0
	Out	8	82	7.21	7.51	7.33 ± 0.07	−59	−41	−50.80 ± 3.81	2.01	5.01	3.85 ± 0.63	0	6	2.95 ± 1.22	3.54	6.66	4.41 ± 0.55	339	460	399 ± 18.6
	Out	10	82	7.01	7.51	7.27 ± 0.14	−55	−30	−46.66 ± 5.25	1.89	4.84	3.55 ± 0.68	0	6	3.07 ± 1.38	3.42	6.93	4.65 ± 0.61	335	439	413 ± 24.2
	In	N/A	81	7.2	7.46	7.35 ± 0.04	−47	−26	−34.91 ± 5.16	8.84	9.72	9.24 ± 0.15	242	294	263 ± 16.54	131	157	144 ± 5.97	760	785	775 ± 4.9
	Out	12	81	7.71	7.9	7.78 ± 0.03	−81	−60	−72.64 ± 4.67	2.7	5.05	4.29 ± 0.62	67	239	100 ± 44.63	14.2	71.3	43.66 ± 18.77	678	734	710 ± 9.8
	Out	14	81	6.35	7.75	7.60 ± 0.15	−72	−48	−61.23 ± 4.68	2.19	4.52	2.98 ± 0.63	57	128	80.48 ± 11.56	11.3	55	31.87 ± 11.95	693	760	738 ± 9.4
	Out	16	41	7.75	8.01	7.86 ± 0.06	−86	−65	−75.32 ± 4.54	1.56	5.19	3.52 ± 0.87	67	252	109 ± 46.20	20.4	68.3	44.59 ± 13.99	670	732	717 ± 11.0
	Out	18	41	7.74	8.17	8.00 ± 0.08	−89	−71	−82.07 ± 4.58	1.42	5.50	3.86 ± 1.06	42	112	75.27 ± 18.37	14.9	51.2	32.64 ± 10.31	696	785	746 ± 13.3

BR basic red, *AB* acid blue, *SD* standard deviation, *Min.* minimum, *Max.* maximum, *N/A* not applicable wetlands 11 and 13 and wetland 12 (low resting and contact times) and between wetlands 15 and 17 and wetland 16 (high resting and contact times) concerning a high concentration for BR46, AB113 and the mixture between them were noted. The result was opposite between wetlands 14 (low resting time and low contact time) and 18 (high resting time and high contact time). Furthermore, in the case of low and high concentrations of the two dyes and the mixture of both of them during spring time, wetlands with higher resting time started to consume more DO when compared to wetlands with lower resting time, because the increase in aerobic microorganisms was greater than that of the anaerobic ones.

Conductivity, suspended solids and turbidity

The EC is commonly applied as an indicator for ion-carrying species (Islam et al. 2011), and corresponding EC values may be used as an indicator for other water quality challenges. Any sudden increase in EC value indicates that there is a source of dissolved ions in the wetland filter (Kumar and Chopra 2012). In comparison, all effluent values for all wetlands in cases of low and high concentrations for both dyes and the dye mixture were compliant with the national effluent discharge quality standards set by the Government of Bangladesh, which stated that the maximum effluent of EC for inland surface water, public sewer secondary treatment plants and irrigated land is 1200 μS/cm (Ahmed et al. 2002). Furthermore, the Sri Lanka Central Environmental Authority (2008) stated that the maximum EC discharge on land for irrigation purpose is 2250 μS/cm. Reference to standards set on the Indian sub-continent is made here, because corresponding countries produce most of the dye wastewater being discharged to the environment.

Concerning the low concentration of BR46 and AB113 (Table 6), a higher elevation was found in planted wetlands 5 and 3 (contact time 94 h), respectively, when compared to the unplanted control wetlands 1 and 2 (contact time 94 h), respectively, while a decrease in EC effluent values was found in wetlands 7 and 9 (contact time 48 h), respectively. For the high concentration for both dyes (BR46 and AB113), the EC effluent values for all wetlands were less than the influent values. Furthermore, wetlands 15 and 17 (long resting and contact times) had EC values less than wetlands 11 and 13 (low resting time and low contact time), respectively. Regarding the dye mixture for both low and high concentrations, all effluent values were less than the influent ones as shown in Table 6. Nevertheless, all previous results indicated no sudden increase in EC values for all wetlands.

The measurement of the conventional pollutant TSS is essential for water treatment works design (Dzurik 2003; Bell et al. 2011). Concerning low concentrations of BR46 and AB113, there were increases in TSS effluent for all wetlands when compared to the influent as shown in Table 6. A lower increase was found in the planted wetlands 7 and 9 (high resting and low contact times) when compared with the unplanted wetlands 1 and 2 and the planted wetlands 5 and 3 (low resting and high contact times), respectively. For the mixture of both dyes, a slight increase of TSS was found for wetland 4, while a decrease was recorded for wetlands 6, 8 and 10. In case of high concentrations for both dyes (BR46 and AB113) and the mixture of the two dyes, a good TSS reduction was recorded for all wetlands as shown in Table 6. Wetlands with high resting and contact times had a lower TSS effluent concentrations, when compared with wetlands, which have low resting and contact times.

All wetland effluents of low and high concentrations of BR46, AB113 and the mixture of both dyes (Table 6) were compliant with the national effluent discharge quality standards set by the Government of Bangladesh, which stated that the maximum TSS effluent concentrations for inland surface water, public sewer secondary treatment plant outflow and irrigated land application are 150, 500 and 200 mg/l, respectively (Ahmed et al. 2002).

A high turbidity of surface water may indicate cloudiness due to elevated concentrations of TSS (Postolache et al. 2007). A higher turbidity value can also increase the temperature of surface water as a result of increased absorption of heat from sunlight, as well as leading to reduced light penetration, which affects photosynthesis (Håkanson 2006).

For the low concentration of the dye BR46, there was an increase in all effluent wetlands when compared with the influents. The planted wetland 7 (high resting and low contact times) has a smaller increase when compared with the unplanted wetland 1 and the planted wetland 5 (low resting and high contact times). In case of dye AB113, a slight increase was recorded for the mean value of the planted wetland 3 (low resting time and high contact time), while a slight decrease was noted for the unplanted wetland 2 (low resting and high contact times) and the planted wetland 9 (high resting time and low contact time). For the mixture of the two dyes, an increase was recorded in wetlands 4 and 6 (low resting time and high contact time), while a decrease was noted in wetlands 8 and 10 (high resting time and low contact time). Regarding the high concentrations of BR46, AB113 and the mixture of these two dyes, all wetlands had a good effluent reduction when compared with the influent. Wetlands 15, 17, 16 and 18 (high resting and contact times) had a greater reduction when compared with wetlands 11, 13, 12 and 14 (low resting and contact times), respectively.

Lin et al. (2005) and Bulc and Ojstršek (2008) stated that the ability of vertical-flow constructed wetlands to reduce TSS and turbidity is relatively poor. In this study, for a low concentration of AB113, a short contact time (48 h) was more advantageous than a long (94 h) one for the reduction of TSS and turbidity as well as in the case of the mixture of both of the dyes (BR46 and AB113). While for high concentrations of both dyes and a mixture of both dyes, the long contact time was better than the short contact time. The percentage TSS reduction rates for BR46, AB113 and the mixture of these dyes were 69, 47 and 71%, respectively.

pH value

The measuring of pH is very important due to its impact on nutrients, COD and TSS in constructed wetlands. The pH value influences microbial populations in degrading pollutants (Eke and Scholz 2008; Lavrova and Koumanova 2013; Paing et al. 2015). Concerning the low BR46 concentration, the mean influent pH was 7.47, a minute decrease in the pH effluent value of 0.3 was noted in the planted wetland 5 (low resting time and high contact time), while there was a slight increase of 0.25 and 0.1 for the unplanted wetland 1 (low resting time and high contact time) and the planted wetland 7 (high resting and low contact times), respectively. For the dye AB113, there was a slight effluent increase of 0.24, 0.22 and 0.16 for the unplanted control wetland 2, planted wetland 3 (low resting and high contact times) and planted wetland 9 (high resting and low contact times), respectively, when compared to the influent value of 7.35 as shown in Table 6. In case of the mixture of both dyes, there was a slight decrease of 0.03, 0.23 and 0.05 for wetland 4, wetland 6 (low resting and high contact times) and wetland 10 (high resting and low contact times), respectively, if compared with the influent value of 7.32, while for wetland 8 (high resting and low contact times), there was a slight increase of 0.01. For the high concentration for both dyes (BR46 and AB113) and the mixture of the two dyes, a slight increase was found ranging between 0.08 and 0.65 for wetlands 11, 13, 12, 14 (low resting and contact times), 17, 16 and 18 (high resting and contact times), while an increase of 1.04 was recorded for wetland 15 (high resting and contact times), when compared with the corresponding influent value of 6.94. This increase in effluent pH values is due to the formation of basic aromatic amine metabolites (Chandra 2015).

Regarding the effect of plants on the pH value for the low concentration of the dye AB113, there was a slight difference of 0.02 between the unplanted control wetland 2 and the planted wetland 3 (both of them have the same conditions). This result suggests that the pH modification in vertical-flow constructed wetlands is probably as a result of interactions between the media and its biofilms, rather than due to the plants; this result confirms findings by Kadlec and Wallace (2008). Unlike the result for the dye BR46, there was a difference of 0.55 between the unplanted control wetland 1 and the planted wetland 5 (both of them have the same conditions). The different results regarding the role of plants on pH are most likely due to each dye having a different chemical structure and molecular weight as shown in Table 3. Furthermore, there were no change in pH values in contrast to the findings, which were obtained by Wieder (1989), who surveyed 128 constructed wetlands treating acid coal mine wastewater and found a difference of 0.11(influent pH was 2.5) between effluent and influent. Mitsch and Wise (1998) corroborated this finding; they found that the difference between the influent and the effluent is 0.52 (influent pH was 2.82).

Kadlec and Wallace (2008) stated that the pH value for most bacteria responsible for degradation is between 4 and 9.5. Nevertheless, findings indicate the ability of macrophytes to modify pH conditions in the rhizosphere (Brix et al. 2002). Furthermore, the effluent pH values for all wetlands in case of low and high concentrations of BR46, AB113 and the mixture of these dyes during the whole period were compared with the effluent discharge quality standards set by the Government of Bangladesh and the Sweden Textile Water Initiative, which state that the pH effluent for inland surface waters, public sewer secondary treatment plants and irrigated land should be between 6 and 9 (Ahmed et al. 2002; STWI 2012).

Dye, colour and chemical oxygen demand reductions

The degradation of azo dyes in aerobic and anaerobic environments involves enzymes and chemical reduction (Khehra et al. 2005; Pandey et al. 2007; Saratale et al. 2011). The first contaminant to be easily recognised in an effluent textile wastewater is colour, which adsorbs and reflects sunlight entering the water, thereby interfering with the aquatic species growth and hindering photosynthesis (Pereira and Alves 2012; Yadav et al. 2012).

For dye and colour reductions concerning low concentrations of dyes (BR46 and AB113), and the mixture of these two dyes, wetlands with long contact times have the best dye and colour reductions (regardless of the planting regime), when compared to wetlands having short contact times. For the high concentration of the dyes BR46 and AB113 (Figs. 1, 2, 3 and 4), and the mixture of both of them, wetlands, which have a low loading rate (high resting and contact times), have better dye and colour reductions ($p < 0.05$), if compared to wetlands with a high loading rate (low resting and contact times) as shown in Table 7, although wetlands that have a low loading rate have better dye reductions when compared with wetlands which have a high loading rate. The influent values expressed as a mass loading rate for wetlands 11 and 13 (high loading rate) were 573.71 ± 26.74 and 576.49 ± 38.15 g/m^2/day, respectively, while for wetlands 15 and 17 (low loading rate), they were 286.86 ± 13.37 and 288.25 ± 19.08 g/m^2/day, respectively (Table 8). The final decision about which loading rate (low or high) is better for a treatment system depends on the design conditions of the specific constructed wetland in the field. The effluent colour values for those wetlands of low concentrations concerning BR46, AB113 and the corresponding mixture of these dyes were compliant with the national effluent discharge quality standards set by the Government of India (1986), which stated the maximum colour value is 400 Pt/Co. In case of the high concentrations for BR46, AB113

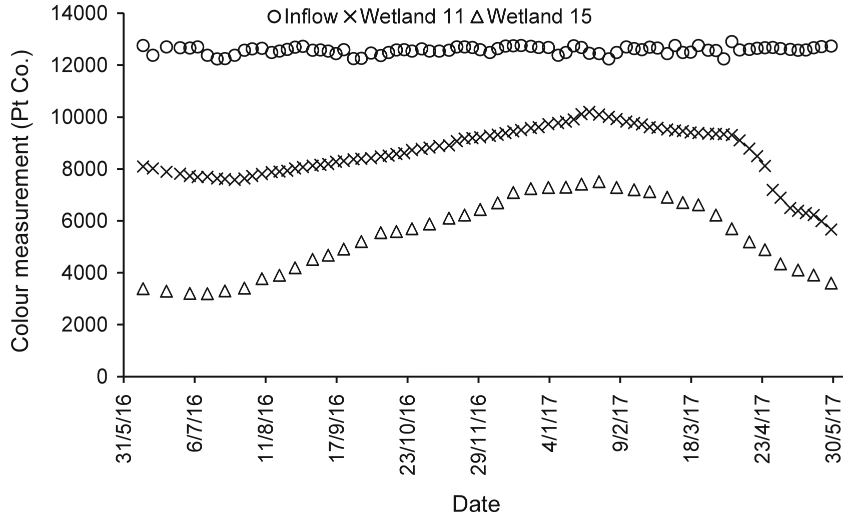

Fig. 1 Inflow and outflow colour measurements of Basic Red 46 for wetlands 11 and 15

and the mixture of these two dyes, they were not compliant even when compared to the maximum threshold for colour (550 Pt/Co.) set by the Government of Taiwan (2003).

For textile wastewater, the measurement of COD is very important to assess organic matter in wetlands. Its reduction processes may be aerobic or anaerobic and are based on filtration, adsorption and microbial metabolism processes (Vymazal et al. 1998; Song et al. 2006; Stefanakis et al. 2014). The effluent COD values for a low concentration of BR46, AB113 and the mixture of both dyes (Table 7) were complaint with the national effluent discharge quality standards set by the Government of Bangladesh, which has set the maximum COD values for inland surface water, public sewer secondary treatment plant outflow and irrigation water to be 200, 400 and 400 mg/l, respectively. In case of high concentrations for BR46, AB113 and a mixture of these dyes for both dyes (Table 7), values were compliant for public sewer secondary treatment and irrigated land (Ahmed et al. 2002). For COD reduction concerning the low concentration of dyes (BR46 and AB113) and the mixture of both of these dyes, the results showed that all wetlands demonstrated good COD reduction as shown in Table 7. Furthermore, wetlands with a long resting time had the best COD reductions, if compared to the control (unplanted wetlands) and/or other wetlands having short resting times. These results indicated that both aerobic and anaerobic environments are acceptable for COD reduction. These findings are supported by the DO values for wetlands as shown in Table 6. Wetlands 7, 9, 8 and 10 have effluent DO values higher than those for wetlands 5, 3, 4 and 6. For the high concentration, COD reductions in wetlands, which have low loading rates (high resting and contact times), were better than for those wetlands with high loading rates (low resting and contact times) in terms of COD concentration (Table 7). However, the influent mass loading rates for wetlands 11 and 13 (high loading rate) were 1423.1 ± 102.27 and 1668.2 ± 132.73 g/m^2/day, respectively, as shown in Table 9, while for wetlands 15 and 17 (low loading rate), they were 711.6 ± 51.14 and 834.1 ± 66.37 g/m^2/day. The

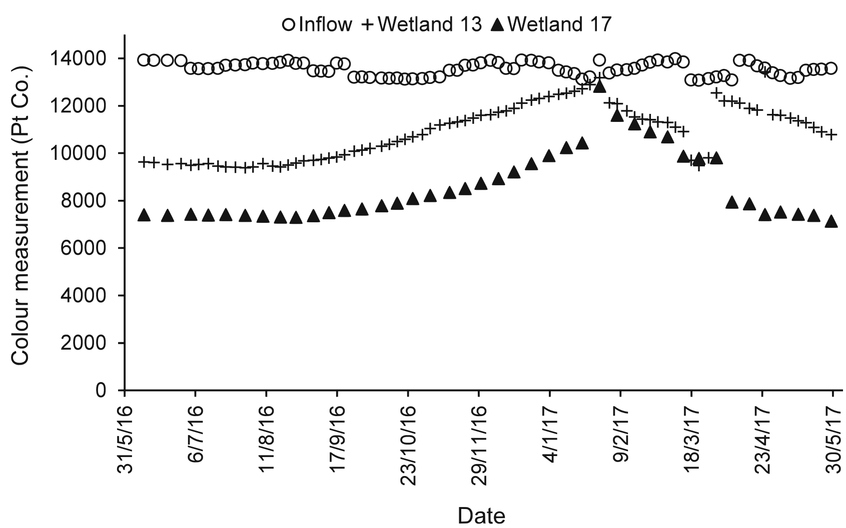

Fig. 2 Inflow and outflow colour measurements of Acid Blue 113 for wetlands 13 and 17

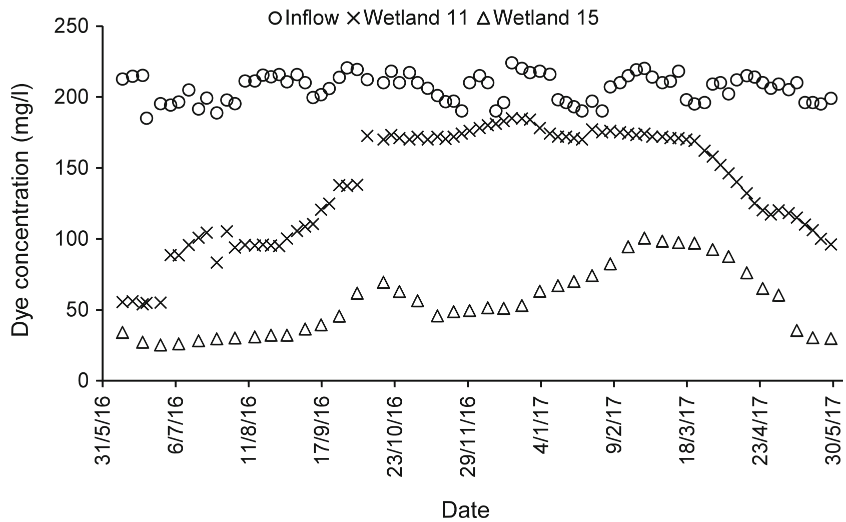

Fig. 3 Inflow and outflow dye concentrations of Basic Red 46 for wetlands 11 and 15

final decision about which wetland performs better depends on the design conditions of constructed wetlands in the field. All previous findings regarding low and high concentrations for BR46, AB113 and the mixture of these two dyes indicate that having both aerobic and anaerobic conditions will improve the COD reduction (Vymazal et al. 1998; Li et al. 2012; Lehl et al. 2016).

Seasonal comparison of effluent dye reductions

The overall seasonal comparison of the influent and effluent dye concentrations for all wetlands is shown in Table 10. In case of low concentration for BR46 and AB113, the best and significant ($\rho < 0.05$) reduction percentages were recorded for the spring season as a result of well-established microbial populations, favourable operating conditions achieved over time and plants, as confirmed by many publications (Scholz et al. 2002; Al-Isawi et al. 2015; Scholz 2015). In case of high concentrations of BR46, AB113 and the mixture of both dyes, the best and significant ($\rho < 0.05$) reduction percentages were linked to summer as shown in Table 9 as a result of the higher temperature as confirmed by several researchers, who stated that the best treatment performance occurs during higher temperatures (Song et al. 2006; Sani et al. 2013).

Nutrient reduction

The removal of ortho-phosphate-phosphorous is controlled by chemical and physical adsorption, sedimentation, plant uptake, precipitation and microbial uptake in constructed wetland systems (Brix 1997; Vymazal 2007, 2010; Johari et al. 2016). Moreover, many researchers have reported that the reduction efficiency of phosphorous compounds is generally poor within constructed wetlands (Choudhary et al. 2011; Lavrova and Koumanova 2013; Ge et al. 2016).

For low concentrations in case of AB113 and BR46, the reductions for planted wetlands 3 and 5 (low resting time and high contact time) were significantly ($p < 0.05$) better

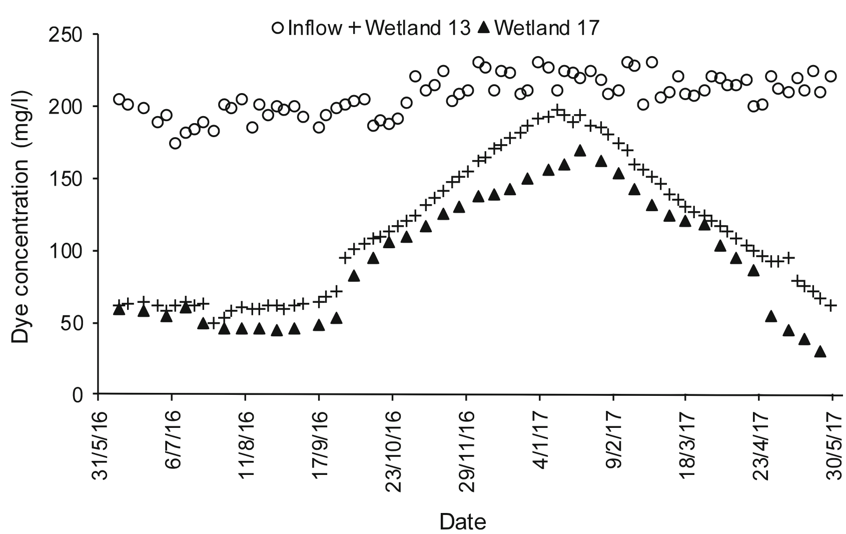

Fig. 4 Inflow and outflow dye concentrations of Acid Blue 113 for wetlands 13 and 17

Table 7 Colour, dye and chemical oxygen demand (COD) reduction for different wetlands (Tables 3 and 4)

Dye	Flow type	Wetland number	No. of samples	Characteristics											
				Colour (Pt/Co)				Dye concentration (mg/l)				COD[a]			
				Min.	Max.	Mean ± SD	Reduction (%)	Min.	Max.	Mean ± SD	Reduction (%)	Min.	Max.	Mean ± SD	Reduction (%)
BR46	In	N/A	82	410	439	422 ± 7.7	N/A	4.3	8.4	6.2 ± 0.75	N/A	200	297	248 ± 20.9	N/A
	Out	1	82	16	187	71 ± 35.3	83	0.0	1.2	0.5 ± 0.22	92	50	93	76 ± 11.8	69
	Out	5	82	26	356	97 ± 71.4	77	0.0	1.0	0.6 ± 0.24	91	54	110	74 ± 16.4	70
	Out	7	82	9	272	120 ± 80.5	72	0.1	1.9	0.7 ± 0.40	89	20	80	45 ± 13.9	82
	In	N/A	81	12,240	12,900	12,574 ± 142.2	N/A	185.0	224.0	206.0 ± 9.60	N/A	478	576	511 ± 36.7	N/A
	Out	11	81	5670	10,210	8604 ± 1053.0	32	53.8	185.0	139.0 ± 39.18	33	196	385	311 ± 52.4	39
	Out	15	41	3200	7520	5442 ± 1459.5	57	25.1	100.6	55.8 ± 24.00	73	135	301	225 ± 48.4	56
AB113	In	N/A	82	530	589	555 ± 16.1	N/A	5.3	10.9	7.5 ± 1.66	N/A	234	310	275 ± 18.1	N/A
	Out	2	82	39	386	135 ± 100.3	76	0.5	4.8	1.2 ± 0.87	85	54	125	66 ± 16.3	76
	Out	3	82	89	400	182 ± 82.5	67	0.6	4.5	1.4 ± 0.84	82	89	176	106 ± 19.4	62
	Out	9	82	224	490	310 ± 71.2	44	0.6	5.3	1.7 ± 1.08	77	40	85	53 ± 10.3	81
	In	N/A	81	13,080	13,990	13,561 ± 279.7	N/A	174.0	231.0	207.0 ± 13.70	N/A	541	710	599 ± 47.7	N/A
	Out	13	81	7230	13,420	10,699 ± 1338.9	21	50.0	197.0	115.0 ± 46.97	44	199	495	357 ± 75.5	40
	Out	17	41	7290	12,820	8707 ± 1432.6	36	30.2	169.0	95.8 ± 43.72	54	190	345	265 ± 43.3	56
Mixture of BR46 and AB113[b]	In	N/A	82	323	450	400 ± 20.8	N/A	0.108	0.182	0.154 ± 0.022	N/A	254	350	292 ± 28.0	N/A
	Out	4	82	145	311	192 ± 37.5	52	0.031	0.156	0.069 ± 0.025	55	86	270	133 ± 43.6	55
	Out	6	82	80	263	119 ± 33.8	70	0.014	0.157	0.050 ± 0.029	68	58	276	117 ± 54.4	60
	Out	8	82	114	339	233 ± 34.0	42	0.039	0.136	0.067 ± 0.021	56	36	91	57 ± 15.7	81
	Out	10	82	206	372	273 ± 31.4	32	0.044	0.138	0.073 ± 0.017	53	32	120	63 ± 24.8	79
	In	N/A	81	16,090	16,190	16,130 ± 29.9	N/A	4.652	5.781	5.339 ± 0.310	N/A	480	594	551 ± 43.5	N/A
	Out	12	81	8620	14,520	12,199 ± 1593.5	24	3.040	5.333	4.371 ± 0.606	18	145	450	347 ± 76.4	37
	Out	14	81	10,790	13,720	12,020 ± 668.2	25	1.884	5.073	3.587 ± 0.936	33	215	431	340 ± 64.2	38
	Out	16	41	9490	13,690	11,933 ± 1096.6	26	1.521	4.892	3.429 ± 0.997	36	225	397	310 ± 47.1	44
	Out	18	41	6430	12,260	10,250 ± 1680.3	36	1.100	4.511	2.819 ± 1.032	47	230	373	294 ± 34.6	46

BR basic red, *AB* acid blue, *SD* standard deviation, *Min.* minimum, *Max.* maximum, *N/A* not applicable

[a] The number of samples is 30

[b] All dye concentration measurements for the mixture are given as a wavelength

Table 8 Dye loading rate

Dye	Flow type	Wetland number	No. of samples	Characteristic			
				Dye loading rate (g/m^2/day)			
				Min.	Max.	Mean ± SD	Reduction (%)
BR46	In	N/A	82	11.98	23.42	17.13 ± 2.09	N/A
	Out	1	82	0.00	3.29	1.45 ± 0.61	92
	Out	5	82	0.00	2.76	1.59 ± 0.67	91
	Out	7	82	0.31	5.21	1.92 ± 1.11	89
	In	11	81	515.22	623.84	573.71 ± 26.74	N/A
	Out	11	81	149.83	515.23	387.12 ± 109.12	33
	In	15	41	257.61	311.92	286.86 ± 13.37	N/A
	Out	15	41	34.95	140.09	77.63 ± 33.42	73
AB113	In	N/A	82	14.82	30.44	20.89 ± 4.63	N/A
	Out	2	82	1.34	13.39	3.20 ± 2.42	85
	Out	3	82	1.69	12.50	3.82 ± 2.34	82
	Out	9	82	1.59	14.79	4.85 ± 3.01	77
	In	13	81	484.59	643.34	576.49 ± 38.15	N/A
	Out	13	81	139.25	548.65	320.28 ± 130.81	44
	In	17	41	242.29	321.67	288.25 ± 19.08	N/A
	Out	17	41	42.05	235.33	133.43 ± 60.88	54

BR basic red, *AB* acid blue, *Min.* minimum, *Max.* maximum, *SD* standard deviation, *N/A* not applicable

compared to those for the unplanted control wetlands 2 and 1 (low resting time and high contact time) and the planted wetlands 7 and 9 (high resting time and high contact time; Table 11). In case of the mixture of both dyes (BR46 and AB113), wetlands 4 and 6 (low resting and high contact times) had better reduction percentages when compared with wetlands 8 and 10 (high resting and low contact times), respectively (Table 11). Assessing the high concentrations for BR46, AB113 and the

Table 9 Chemical oxygen demand (COD) loading rate

Dye	Flow type	Wetland number	No. of samples	Characteristics			
				COD loading rate (g/m^2/day)			
				Min.	Max.	Mean ± SD	Reduction (%)
BR46	In	N/A	30	557.0	827.1	690.7 ± 58.18	N/A
	Out	1	30	137.9	259.0	212.1 ± 32.84	69
	Out	5	30	150.4	306.4	205.2 ± 45.79	70
	Out	7	30	56.0	221.4	124.3 ± 38.68	82
	In	11	30	1331.2	1604.2	1423.1 ± 102.27	N/A
	Out	11	30	545.9	1072.2	866.1 ± 145.85	39
	In	15	30	665.6	802.1	711.6 ± 51.14	N/A
	Out	15	30	188.0	419.1	313.3 ± 67.42	56
AB113	In	N/A	30	651.7	863.4	765.9 ± 50.46	N/A
	Out	2	30	150.1	348.1	185.1 ± 45.34	76
	Out	3	30	247.3	490.2	294.5 ± 54.06	62
	Out	9	30	110.6	235.6	148.5 ± 28.69	81
	In	13	30	1506.7	1977.4	1668.2 ± 132.73	N/A
	Out	13	30	554.2	1378.6	994.3 ± 210.21	40
	In	17	30	753.4	988.7	834.1 ± 66.37	N/A
	Out	17	30	264.6	480.4	369.0 ± 60.31	56

BR basic red, *AB* acid blue, *Min.* minimum, *Max.* maximum, *SD* standard deviation, *N/A* not applicable

Table 10 Seasonal artificial wastewater removal (mg/l) and for the mixture dyes (absorbance)

Dye	Type of flow	Wetland number(s)	Characteristics							
			Summer[a]		Autumn[b]		Winter[c]		Spring[d]	
			Mean ± SD	Removal (%)	Mean ± SD	Removal (%)	Mean ± SD	Removal (%)	Mean ± SD	Removal (%)
BR46	In	N/A	6.26 ± 0.99	N/A	6.07 ± 0.49	N/A	6.22 ± 0.76	N/A	6.06 ± 0.66	N/A
	Out	1	0.56 ± 0.13	91	0.67 ± 0.23	89	0.47 ± 0.22	92	0.34 ± 0.08	94
	Out	5	0.58 ± 0.18	91	0.58 ± 0.29	90	0.54 ± 0.31	91	0.58 ± 0.16	90
	Out	7	1.00 ± 0.39	84	0.69 ± 0.16	88	0.66 ± 0.37	89	0.28 ± 0.27	95
	In	N/A	202.80 ± 9.52	N/A	207.47 ± 9.64	N/A	208.64 ± 11.05	N/A	204.65 ± 7.18	N/A
	Out	11	95.9 ± 11.94	53	168.30 ± 14.18	19	174.70 ± 4.63	16	128.58 ± 22.27	37
	Out	15	30.94 ± 4.41	85	54.18 ± 8.07	74	80.01 ± 16.99	62	63.71 ± 26.82	69
AB113	In	N/A	8.96 ± 1.55	N/A	6.66 ± 1.15	N/A	6.51 ± 1.36	N/A	7.78 ± 1.17	N/A
	Out	2	1.98 ± 1.13	78	0.96 ± 0.65	86	0.70 ± 0.11	89	0.80 ± 0.18	90
	Out	3	2.24 ± 1.14	75	1.18 ± 0.76	82	1.03 ± 0.15	84	0.85 ± 0.18	89
	Out	9	3.10 ± 0.94	65	1.56 ± 0.53	77	1.25 ± 0.32	81	0.69 ± 0.02	91
	In	N/A	192.33 ± 8.14	N/A	207.58 ± 13.31	N/A	208.00 ± 9.24	N/A	213.82 ± 7.04	N/A
	Out	13	60.68 ± 3.90	68	129.81 ± 27.93	37	172.16 ± 21.45	17	96.49 ± 20.34	55
	Out	17	49.81 ± 5.65	74	109.38 ± 26.88	47	146.48 ± 16.07	30	71.29 ± 33.29	67
Mixture of BR46 and AB113 (absorbance)	In	N/A	0.15 ± 0.02	N/A	0.16 ± 0.02	N/A	0.15 ± 0.02	N/A	0.15 ± 0.03	N/A
	Out	4	0.07 ± 0.001	53	0.06 ± 0.01	63	0.05 ± 0.01	67	0.09 ± 0.05	40
	Out	6	0.05 ± 0.01	67	0.04 ± 0.001	75	0.04 ± 0.00	73	0.08 ± 0.05	47
	Out	8	0.07 ± 0.001	53	0.06 ± 0.01	63	0.06 ± 0.00	60	0.09 ± 0.04	40
	Out	10	0.08 ± 0.001	47	0.07 ± 0.01	56	0.06 ± 0.00	60	0.09 ± 0.03	40
	In	N/A	5.35 ± 0.38	N/A	5.24 ± 0.28	N/A	5.34 ± 0.26	N/A	5.44 ± 0.29	N/A
	Out	12	3.60 ± 0.35	33	4.57 ± 0.21	13	4.89 ± 0.32	8	4.57 ± 0.41	22
	Out	14	2.37 ± 0.44	56	3.98 ± 0.38	24	4.51 ± 0.43	16	3.62 ± 0.59	33
	Out	16	2.09 ± 0.53	61	3.75 ± 0.45	28	4.32 ± 0.54	19	3.75 ± 0.58	31
	Out	18	1.57 ± 0.52	71	3.34 ± 0.44	36	3.96 ± 0.56	26	2.52 ± 0.0.42	54
Temperature (°C)	N/A	N/A	22.8	N/A	11.6	N/A	8.6	N/A	20.5	N/A

BR basic red, *AB* acid blue, *N/A* not applicable, *SD* standard deviation

[a] From 21 6 2016 to 21 September 2016

[b] From 22 September 2016 to 20 December 2016

[c] From 21 December 2016 to 19 March 2017

[d] From 20 March 2017 to 29 May 2017

mixture of these dyes, wetlands 15 and 17 and wetlands 16 and 18 (high resting and contact times) had lower PO_4-P effluent concentrations when compared with wetlands 11 and 13 and wetlands 12 and 14 (low resting and contact times), respectively (Table 8). The previous results for low and high concentrations indicate that the reduction efficiency for PO_4-P was relatively good, especially for wetlands, regardless of planting regime, with long contact times (and lower resting times).

A typical standard set by environment agencies for PO_4-P reduction concerning secondary wastewater treatment is 2 mg/l (Royal Commission on Sewage Disposal 1915). Effluent PO_4 *p* values were complaint to this standard for low concentrations of BR46 (planted wetland 5; 1.76 mg/l). In comparison, a slight increase in case of the low concentration for the mixture of these two dyes (wetland 6; 2.03 mg/l) was noted. However, the value was relatively high in case of low concentration of AB113 (planted wetland 3; 3.91 mg/l). For other wetlands, the effluent values of PO_4-P were much higher than the standard value of 2 mg/l.

Nitrification and denitrification are the main reduction mechanisms of nitrogen in constructed wetlands, and these mechanisms include a two-step process: ammonium is oxidised to nitrite followed by oxidisation of nitrite to nitrate (nitrification process). The subsequent denitrification process involves the reduction of nitrate to gaseous nitrogen (Schaechter 2009; Kessel et al. 2015; Song et al. 2015; Yang et al. 2016). Regarding NH_4-N reduction percentages for low concentrations of BR46 and AB113 (Table 11), planted wetlands 7 and 9 (high resting and low contact times) have better reduction percentages when compared with the unplanted control wetlands 1 and 2 as well as the planted wetlands 5 and 3 (low resting and high contact times), respectively. In case of a mixture of both dyes, wetlands 8 and 10 (high resting and low contact times) had better reduction percentages compared to wetlands 4 and 6 (low resting time and high contact time), respectively (Table 11). For the high concentrations of BR46, AB113 and the mixture of both dyes, wetlands 15, 17, 16 and 18 (high resting and contact times) have better

Table 11 Inflow and outflow water quality characteristics for nutrients related to different wetlands (Supplementary Material 1 and Table 2)

Dye	Flow type	Wetland number	No. of samples	Characteristics											
				Ammonia nitrogen (mg/l)				Nitrate-nitrogen (mg/l)				Ortho-phosphate-phosphorus (mg/l)			
				Min.	Max.	Mean ± SD	Removal (%)	Min.	Max.	Mean ± SD	Removal (%)	Min.	Max.	Mean ± SD	Removal (%)
BR46	In	N/A	30	16.9	25.7	21.98 ± 2.43	N/A	21.9	30.2	25.07 ± 2.38	N/A	5.4	7.7	6.36 ± 0.68	N/A
	Out	1	30	11.0	22.1	18.72 ± 2.48	15	0.0	0.5	0.21 ± 0.18	99	2.1	5.9	3.49 ± 0.98	45
	Out	5	30	3.7	19.0	14.41 ± 3.53	34	0.0	0.2	0.02 ± 0.05	100	0.0	3.3	1.76 ± 0.83	72
	Out	7	30	2.6	16.1	6.19 ± 4.78	72	0.0	0.7	0.21 ± 0.20	99	3.4	5.9	4.54 ± 0.69	29
	In	N/A	30	21.3	29.7	26.99 ± 2.41	N/A	28.7	38.1	33.47 ± 1.75	N/A	62.5	68.3	65.78 ± 1.75	N/A
	Out	11	30	12.5	31.4	21.67 ± 4.12	20	4.03	16.6	8.41 ± 3.99	75	33.6	52.4	45.33 ± 6.32	31
	Out	15	30	9.2	26.3	20.29 ± 4.79	25	1.4	13.9	6.23 ± 3.61	81	11.9	35.8	19.09 ± 6.70	71
AB113	In	N/A	30	20.4	24.4	23.36 ± 1.02	N/A	19.9	28.2	23.53 ± 2.19	N/A	7.6	11.8	10.34 ± 1.13	N/A
	Out	2	30	9.9	24.3	19.23 ± 4.16	18	0.0	1.0	0.11 ± 0.23	100	2.3	7.3	4.00 ± 1.62	61
	Out	3	30	13.5	36.2	24.48 ± 7.59	−5	0.0	1.5	0.55 ± 0.43	98	2.0	6.2	3.91 ± 1.07	62
	Out	9	30	9.1	17.4	13.20 ± 2.25	43	0.0	1.1	0.22 ± 0.31	99	5.1	8.3	6.22 ± 0.94	40
	In	N/A	30	26.9	29.0	28.16 ± 0.68	N/A	30.2	37.6	33.05 ± 2.10	N/A	144.0	158.0	156.00 ± 3.27	N/A
	Out	13	30	13.5	27.7	20.89 ± 4.74	26	3.1	7.3	5.59 ± 0.89	83	8.9	95.4	46.15 ± 28.22	70
	Out	17	30	9.7	24.7	18.60 ± 4.98	34	2.9	6.3	4.57 ± 0.99	86	21.2	67.0	42.29 ± 16.47	73
Mixture of BR46 and AB113	In	N/A	30	27.2	34.2	30.27 ± 1.81	N/A	22.6	29.0	25.37 ± 1.72	N/A	6.0	8.7	7.56 ± 0.81	N/A
	Out	4	30	17.4	35.0	26.01 ± 4.75	14	0.0	0.3	0.08 ± 0.09	100	2.5	8.3	4.47 ± 1.48	41
	Out	6	30	7.2	22.2	15.66 ± 4.39	48	0.0	2.1	0.13 ± 0.39	100	0.4	4.2	2.03 ± 1.02	73
	Out	8	30	5.1	17.0	9.78 ± 3.21	68	0.1	13.9	3.71 ± 4.79	85	4.2	8.2	5.85 ± 1.19	23
	Out	10	30	2.3	17.9	10.75 ± 5.39	65	0.0	2.5	0.39 ± 0.59	99	3.9	8.9	6.14 ± 1.54	19
	In	N/A	30	23.2	33.6	30.75 ± 2.81	N/A	31.2	35.1	33.91 ± 1.03	N/A	112.1	123.0	118.00 ± 2.72	N/A
	Out	12	30	10.7	24.5	20.72 ± 3.68	33	4.3	9.7	6.45 ± 1.44	81	5.2	86.8	58.37 ± 24.80	51
	Out	14	30	9.8	26.4	19.87 ± 3.85	35	3.7	7.5	5.79 ± 1.04	83	4.7	97.2	56.34 ± 26.13	52
	Out	16	30	8.0	25.2	17.41 ± 5.47	43	2.9	6.1	4.89 ± 0.86	86	19.5	74.3	57.31 ± 15.98	51
	Out	18	30	8.59	27.7	16.84 ± 5.25	45	2.4	8.4	4.71 ± 1.23	86	26.3	69.7	50.48 ± 13.16	57

BR basic red, *AB* acid blue, *SD* standard deviation, *Min.* minimum, *Max.* maximum, *N/A* not applicable

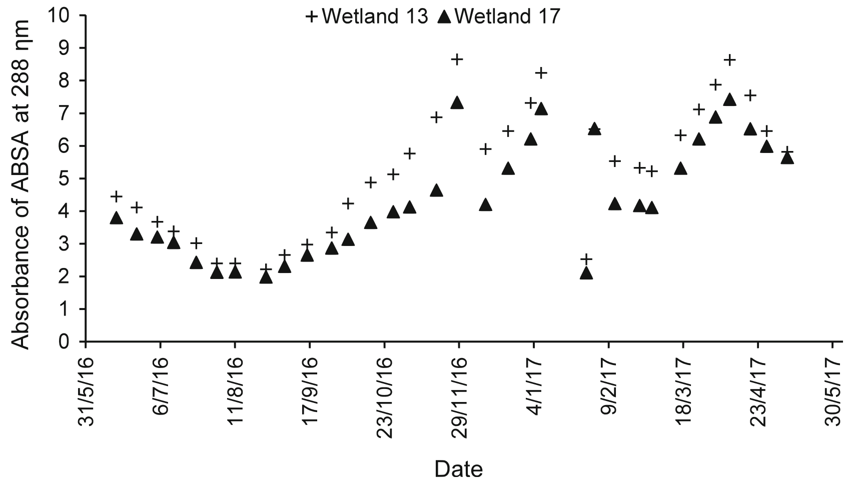

Fig. 5 3-Aminobenzenesulfonic acid absorbance for the high concentration of Acid Blue 113

reduction percentages when comparing them with wetlands 11, 13, 12 and 14 (low resting and contact times), respectively. The previous results indicate that aeration plays a major function in determining the performance of higher nitrogen reduction. These findings are confirmed by many researchers (Vymazal 2007; Wu et al. 2011a; Fan et al. 2013). The effluent NH_4-N values for all wetlands in case of low and high concentrations for BR46, AB113 and the mixture of both dyes were compared to the traditional UK standard (Royal Commission on Sewage Disposal 1915), which states that the NH_4-N outflow from the secondary wastewater should not exceed 50 mg/l. Furthermore, both the Government of India (1986) and the Government of Bangladesh (Ahmed et al. 2002) stated that 50 mg/l is an acceptable outflow threshold to protect surface waters.

Regarding NO_3-N reduction for low concentrations of BR46, AB113 and the mixture of both dyes (Table 11), the influent NO_3-N values were in the range 23.53 to 25.37 mg/l. The reduction percentages for all wetlands were in the range between 83 and 100%. For the high concentration of BR46, AB113 and the mixture of both dyes, the influent values were approximately 33.45 mg/l and the reduction percentages for all wetlands were in the range from 75 to 86% (Table 11). The NO_3-N reduction percentages indicate that vertical-flow constructed wetlands have a good ability to reduce nitrogen in high percentages, especially when there is a source of organic carbon, and both dyes have carbon in their chemical structure (Supplementary Material S1). These findings have been confirmed by Lavrova and Koumanova (2014) as well as Shen et al. (2015).

Furthermore, Lavrova and Koumanova (2013) demonstrated that vertical-flow constructed wetlands can effectively reduce NO_3-N with and without plants with a sufficient organic carbon source. The effluent NO_3-N values for all wetlands in case of low and high concentrations for BR46, AB113 and the mixture of both dyes were compared to the traditional UK standard, which states that the NO_3-N outflow concentration should not exceed 50 mg/l (Royal Commission on Sewage Disposal 1915).

Aromatic amine reductions

Azo dye decolourisation is achieved under aerobic, anaerobic and anoxic conditions (O'Neill et al. 2000; Sponza and Işik 2002; Van Der Zee 2002; Davies et al. 2006). In anaerobic conditions, the azo bond (N=N) cleaves (cutes), and this

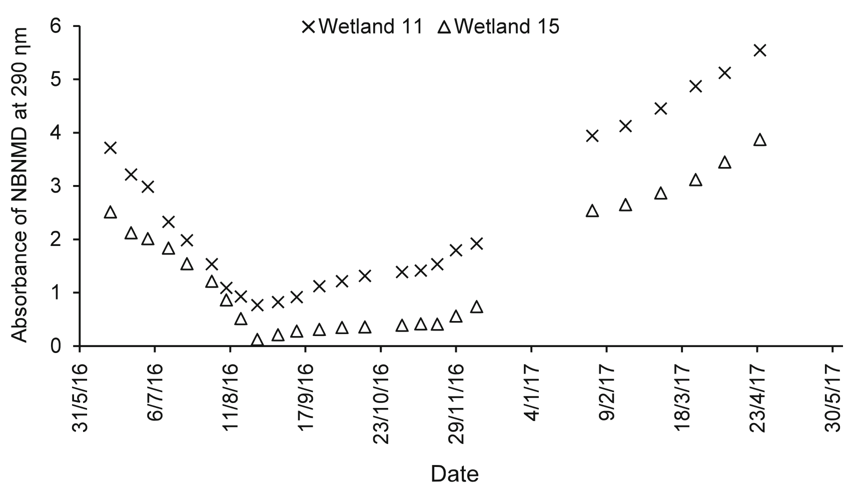

Fig. 6 *N*-Benzyl-*N*-methylbenzene-1,4-diamine absorbance for the high concentration of Basic Red 46

process releases aromatic amine, which resists any further anaerobic treatment (Brown and Hamburger 1987; Chung and Stevens 1993). Aromatic amine can be reduced under aerobic treatment (Weber and Wolfe 1987; Pinheiro et al. 2004; Ong et al. 2011). The amine compounds are toxic and negatively impact on some bacteria, leading to insufficient dye degradation (Phugare et al. 2011; Holkar et al. 2014). Each dye has one or more types of aromatic amines (Pielesz et al. 2002; Pinheiro et al. 2004). Wetlands can degrade aromatic amines under aerobic conditions (Mbuligwe 2005; Ong et al. 2010, 2011).

In this study, three types of amines were released as a result of the degradation of the dye AB113: 3-aminobenzenesulfonic acid (ABSA), 1,4-diaminonaphthalene (DAN) and 5-amino-8-(phenylamino)naphthalene-1-sulfonic acid (ANSA) (Senthilvelan et al. 2014). The corresponding wavelengths for maximum absorbance are 288, 255 and 225 nm, respectively (Koepernik and Borsdorf 1983; Paul et al. 1990). In case of BR46, two aromatic amines were released as a result of its degradation: *N*-benzyl-*N*-methylaniline (NBNMA) and *N*-benzyl-*N*-methylbenzene-1,4-diamine (NBNMD), with wavelengths of maximum absorbance of 254 and 290 nm, respectively (Fihtengolts 1969; Küçükgüzel et al. 1999). For the low concentration of AB113, in case of ABSA (Supplementary Material 4), wetland 9 (high resting and low contact times) had a significant ($\rho < 0.05$) reduction efficiency when compared with the unplanted wetland 2 and the planted wetland 3 (low resting time and high contact time). Regarding the high concentration of AB113 and for ABSA amine (Fig. 5), wetland 17 (high resting time and high contact time) has a significant ($\rho < 0.05$) reduction efficiency when compared with wetland 13 (low resting and contact times). It follows that a reduction of aromatic amine compounds requires aerobic conditions (see above). The amines DAN and ANSA were not detected for low and high concentrations by a UV spectrophotometer. This is because both of them are instable, and therefore, they were not detected in solution as confirmed by Davies et al. (2005) and Davies et al. (2006), who also found that using HPLC analysis did not detect this type of amine.

For the low concentration of BR46 concerning the NBNMD amine, wetland 7 (high resting and low contact times) has a significant ($\rho < 0.05$) reduction efficiency when compared with the unplanted wetland 1 and the planted wetland 5 (low resting and high contact times) as shown in Supplementary Material 5. The NBNMA amine was not detected by the UV spectrophotometer regarding the unplanted wetland 1, while for the planted wetlands 5 and 7, it was sometimes detected, but this amine was not dependent on the activity of microorganisms required to degrade this type of amine. For the high concentration of BR46 in case of the NBNMD amine (Fig. 6), wetland 15 (high resting and contact times) had a significant ($\rho < 0.05$) reduction efficiency when compared with wetland 11 (low resting and contact times). Furthermore, during the period between 19 December 2016 and 3 February 2017, the NBNMD amine was not detect as a result of a decrease in temperature during this period and because of the growth and development of microbial communities (Jerman et al. 2009). The NBNMA amine was not detected by the UV spectrophotometer for the same reason as stated above.

Conclusions and recommendations for further research

Regarding low BR46 and AB113 reductions, the unplanted wetlands had good reduction performances, if compared with planted wetlands concerning the removal of dyes. For the high concentrations of AB113, BR46 and a mixture of both of them, wetlands with long contact times were considerably better than wetlands which had short contact times, in terms of dye, colour and COD reductions. For low and high inflow dye concentrations, best removals were recorded for spring and summer in this order. Furthermore, aromatic amine concentrations were very low.

The vertical-flow wetland filters were linked to significantly ($p < 0.05$) good denitrification processes for both low and high concentrations of AB113, BR46 and the mixture of both dyes throughout the year. Regarding nitrate nitrogen (NO_3-N), the reduction percentage rates of AB113, BR46 and a mixture dye of both of them were between 85 and 100%.

Future wetland designs for the treatment of dye wastewater should be based on these recent more long-term research findings. The authors recommend to assess the effect of pH (low and high) on dye reduction. Aromatic amine compounds require more large-scale process investigations, especially in case of mixtures of dyes.

Acknowledgements Amjad Hussein obtained a PhD Studentship from the Ministry of Higher Education and Scientific Research of the Government of Iraq via Al-Muthanna University. The sponsor had no influence over the analysis and presentation of findings.

Compliance with ethical standards

Conflict of interest The authors declare that they have no competing interests.

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