

Two-stage hybrid constructed wetlands system for industrial wastewater treatment

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ABSTRACT

A two-stage hybrid system with a continuous flow rate was constructed to treat highly polluted industrial wastewater of Al-Najibiyia Power Plant in Basrah city, Iraq. The system consisted of a Horizontal Subsurface Flow System (HSSF) planted with *Phragmites australis* (Cav.) followed by a Surface Flow System (SF) planted with *Hydrilla verticillata* (L.f.) Royle in series and there were two similar systems on the opposite side representing the contaminated control system. The experiment was conducted for 72 days. Several water quality parameters were measured in both raw and treated wastewaters, including pH, Biological Oxygen Demand (BOD₅), Chemical Oxygen Demand (COD), Total Suspended Solids (TSS), and turbidity, in addition to some ions such as Sulfate ion (SO₄²⁻) and Chloride ion (Cl⁻). The system performance results showed that the pollutants removal rate in the presence of plants was high reached to: BOD₅(65.85%), COD (91%), TSS (88.83%), turbidity (83.65%), SO₄²⁻ (55.11%) and Cl⁻ (23.88%) as compared to the systems without plants which achieved BOD₅(44.72%), COD (64.54 %), TSS (68.73%), turbidity (66.05%), SO₄²⁻ (33.41%) and Cl⁻ (9.9%). The results revealed that constructed wetlands (CWs) are a good option for industrial wastewater treatment compared to other expensive conventional treatment technologies. It is also revealed that the presence of plants has an influential role in wastewater treatment. Moreover, it could be concluded that a hybrid CWs system could enhance the pollutants removal efficiency better than single constructed wetlands.

Keywords: Constructed Wetlands; Industrial Wastewater; Water Quality; Hybrid System, Pollution.

Introduction

The great industrial revolution, in conjunction with the rapid increase in population growth and use of water in various industries such as power plants and chemical industries, has led to the discharge and production of considerable amounts of toxic wastes into the environment, which

causes serious environmental problems (Danh *et al.*, 2009). The organic pollution of water is always obvious, which is noticeable in the form of increased acidity and higher concentrations of salts, sediments, nutrients, trace metals, chemicals, and other toxins and harmful pathogenic organisms. Globally, the harmful substances found in the industrial

effluents represent one of the largest sources of water pollution, which affect aquatic ecosystems and humans which include algal blooms, abnormalities and death of aquatic lives, habitat deterioration, and increased water flow with the accumulation and toxicity of chemicals within the bodies of organisms and magnification at higher levels of the food chain (Gazette, 2010). Many industries use natural water bodies, especially freshwater reservoirs as a sink for their effluents, which often rendered these natural resources unsuitable for both primary and/or secondary usage. River systems are the primary base for the disposal of waste from industries that are near them. Depending on the type of industry, various levels of pollutants can be discharged into the environment directly or indirectly through public sewer lines (Kanu and Achi, 2011).

These pollutants may provide the water sources with suspended solids, both degradable and nonbiodegradable organics; oils and greases; dissolved inorganics; heavy metal ions; acids, alkaline and colouring compounds. Industrial effluents are usually characterized by their contents of abnormal turbidity, conductivity, oils and greases, hydrocarbons, biological oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), total hardness, and variable metal concentrations (Kanu and Achi, 2011; Cubillos *et al.*, 2014).

The collection, monitoring, and treatment of wastewater in population areas is of a great importance and often problematic (Kouki *et al.*, 2009); therefore, they need an integrated wastewater treatment technology which are environmentally sustainable, cost-effective and with very high treatment performances, such as constructed wetlands (CWs) (Ansola *et al.*, 2003; Hench *et al.*, 2003).

Constructed wetlands (CWs) are developed engineering systems that combine the green plants along with their associated microorganisms in the presence of an appropriate substrate (Vymazal, 2005) to improve the quality of point and nonpoint sources of water pollution, including stormwater runoff, domestic wastewater,

agricultural wastewater, industrial wastewater, and land renovation after mining or refineries (Sidek *et al.*, 2018). Compared with conventional treatment systems, CWs are cost-effective, easy in management and maintenance, applicable for various wastewater treatment, in addition to other benefits, like green areas for recreation, study purposes, and wildlife habitats (Patel and Dharaiya, 2013).

CWs can be classified according to certain bases as the type of vegetation (Free-floating, Floating-leaved, Submerged, and Emergent), hydrology (surface flow systems or subsurface flow systems), and finally, the subsurface flow constructed wetland (SSFCW) are subdivided into two types according to direction and pattern of wastewater flow that passes through the substrate as a horizontal and a vertical subsurface flow (Imfeld *et al.*, 2009; Sayadi *et al.*, 2012). The combination of two CW types such as (VS-HS, HS-VS, VS-SF, or HS-SF) is known as Hybrid system, this system is often used to achieve high treatment efficiency since it utilizes the advantages of each single CW system (Vymazal, 2010). On the other hand, the information on the mechanics of CWs in wastewater treatment is still limited because it is influenced by many factors, including design dimensions, pollutant concentrations, loading flow rate, microbial populations, and plant species concerned (De Busk, 1999).

Generally, the treatment of industrial wastewaters depending on the type of industry such as petrochemical, petroleum and refineries effluents, pulp and paper, textile, mining, and food processing wastewater (Lavigne and Jankiewicz, 2000; Khalil *et al.*, 2005; Yang and Hu, 2005; Bulc *et al.*, 2006). The aims of this study were to assess the locally built constructed wetland systems as an effective way in removal of organic matters (BOD₅ and COD) and TSS, to evaluate the quality of treated water by comparison with the final discharge with the Iraqi standards to enter the river systems as clean water, and assessment of native species of aquatic plants to reducing the number of pollutants.

2-Materials and methods

2.1- Constructed wetlands system design

Two series of two-stage hybrid wetland system with continues flow rate was constructed; the system included three parts started with a storage tank, a series of glass basin with horizontal subsurface flow (HSSF) system followed by a glass basin with surface flow system (SF), and a series of similar two glass basins as a replicate which represented as contaminated control horizontal subsurface flow (CC HSSF) and contaminated control surface flow (CC SF). The two basins were located on a concrete stand at about (1.4 and 1) m in height from the ground and each basin has about (0.5-1)cm slope (Patel and Dharaiya, 2013) of its base. The horizontal subsurface flow system (HSSF) was filled with a local substrate to about 20 cm height. The substrate consists of two gravel layers (coarse + fine) with a height of 3cm, followed by a fine sand layer at the top at a height of 9 cm. The HSSF system was planted with *Phragmites australis* (Cav.) while the SF system was planted with *Hydrilla verticillata* (L.f.) Royle.

The industrial wastewater samples were collected from Al- Najibiyia Power Plant in Basrah city, Iraq and transferred to the treatment unit at Basrah University for the purpose of treatment. Later, the treated water samples were collected after 7, 14, 28, 42, and 72 days of the experiment period.

Water quality monitoring

During the phytoremediation period from January to March 2019, treated water was collected for 72 days at 0,7, 14, 28, 42, and 72 d to evaluate the system's performance.

Some measurements were conducted directly on the field, such as pH, EC and temperature. All physical parameters were measured by their particular meters such as conductivity (EC), pH. In contrast, DO and BOD₅ were measured according to azide modification Winkler method (Lind, 1979), Total Suspended Solids (TSS) were measured according to the method described in APHA (2005), Chloride ion was measured according to APHA (2005), Sulfate ions were measured as described in (Eaton *et al.*, 2005), and Chemical Oxygen Demand (COD) was measured according to APHA (2005).

Statistical Analysis

All experimental data were statistically analyzed using SPSS version 23 (SPSS Inc., USA) to examine variance (ANOVA). All the analyses were performed in duplicates to substitute for experimental errors and were confirmed as mean \pm standard deviation (SD). The water quality parameters, pollutants, plant growth and bacterial populations (dependent variables) according to system type and day (independent factors) were analyzed using the compare means test with LSD test to determine the significant differences presence among means. Statistical significance was defined at 95% confidence level or ($p < 0.05$).

Results and discussion

Constructed wetland treatment performance

Table1 shows the characteristics of raw industrial wastewater before treatment which does not agree with the Iraqi standards for industrial wastewater.

Table1: Characteristics of the raw industrial wastewater of Al- Najibiya Power Plant

Parameter	Unit	Industrial wastewater (Mean value±SD) Recent study	Iraqi limits for industrial water(2009)
Temperature	°C	21.15±3.46	<35
PH	-	8.65±1.2	6-9.5
EC	mS\cm	10.75± 2.27	-
TSS	mg\L	647±66.47	60
Turbidity	NTU	17.75±1.69	-
BOD ₅	mg\L	13.55±1.77	40
COD	mg\L	355.00±77.78	100
Cl	mg\L	1571.42±39.72	600
SO ₄	mg\L	1848±11.31	400

Physical and chemical parameters

All physical and chemical parameters were recorded throughout the experiment period (72 days) illustrated higher results in the systems with the plant than the systems without plant.

Temperature

In general, the results showed that the temperature mean values ranged between (13.7-23.6) °C in the raw wastewater (WW) while it ranged between (8.05-20.65) °C in the treatment systems throughout the 72 days with no significant differences among system types. However, there were significant differences among days of retention time (Fig.1). This temperature is

normal for the moderate regions like Basrah city in the winter season. Luise *et al.*(1993) reported that temperature has a significant effect on wetland treatment efficiency since the wetland is shallow water and open to the atmosphere; therefore, it is subjected to the surrounding environment and climate changes. Al-Maliky (2018) also reported that the system and plant type could impact the wetland system's temperature. This is clear in systems with plant (HSSF and SF), which have kept the temperatures in a normal level compared to systems without plant as demonstrated by Panrare *et al.* (2015) that engineered ecosystems can reduce the temperature of the surrounding environment by three times

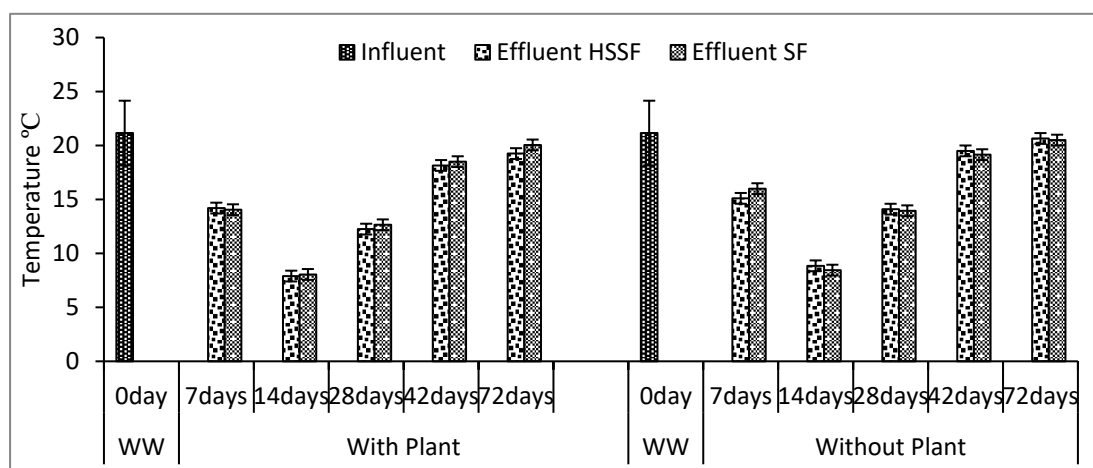


Fig.1: Temperature variations in hybrid system during the experiment period (72 days)

EC and pH

EC level fluctuated during the 72 d, where it declined during the first two weeks, but it showed increasing after that (Fig.2). However, the lowest EC value (4.23 ± 0.01) mS/cm was recorded after 14 day in the SF system while the highest value (10.75 ± 0.03) mS/cm was recorded after 42 days in CC HSSF system. Statistical analysis showed that there were significant differences in EC values among system types and among days.

In general, systems with plant (HSSF and SF) showed higher and significant EC reduction compared to systems without plant (CC HSSF and CC SF), especially during the first two weeks of the experiment due to the presence of *P. australis* and *H. verticillata*, but after that EC began to increase slightly as it reached to the end of 72 days. This is may be due to the increasing ions in water such as chloride, sulfate, and nitrate and decreasing in the plant's ability to absorb

these ions where these dissolved salts also act as a weak electrolyte and subsequently affect the conductivity of the aquatic system (Adams, 1973), in addition to the initial high level of EC in the industrial feeding wastewater. It was also mentioned by (Al-Maliky, 2018; Green *et al.*, 2004) that the high percentage of evapotranspiration represents a strong reason because of increase in temperature values which causes an increase in salinity of the final discharge of treated water, in addition to the interactions between porous media and wastewater (Gikas and Tsihrintzis, 2012).

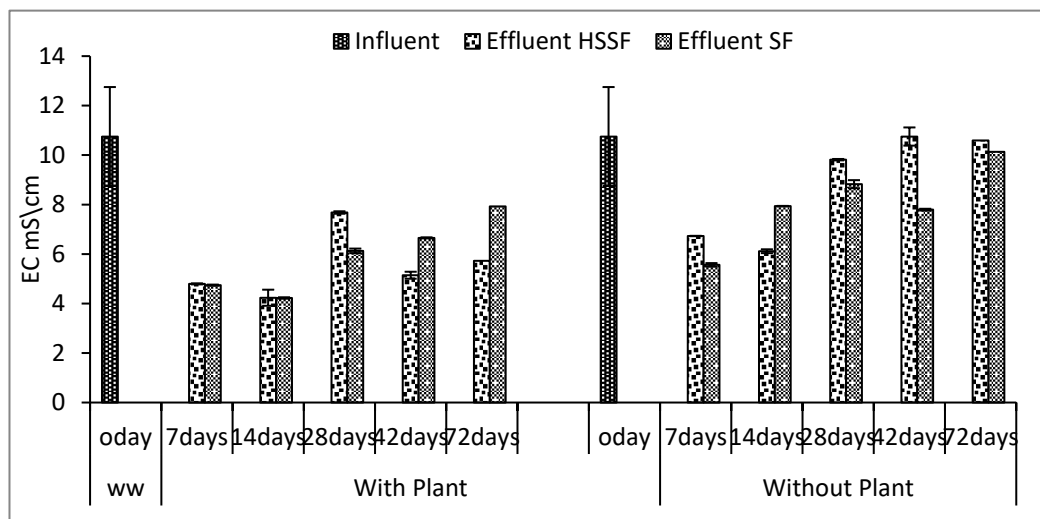


Fig.2: EC values mS/cm variation during the experiment period (72 days)

The mean pH value was in the range of (6.6-8.5) which showed the suitable conditions for removal of different pollutants and also appropriate situations for different water reuse applications (Fig.3), in addition to that pH was approximately constant in the final discharge and within the acceptable ranges compared with the influent which verified the buffering capacity of the CW (Justin *et al.*, 2009). Patel and Dharaiya (2013) demonstrated that

pH of water and soils in wetland systems has a strong impact on the path of many chemical reactions and physical and biological processes, like partitioning of ionized and un-ionized forms of acids and bases, cation exchange, solid and gases solubility, and biological transformations, moreover many metabolic activities are pH-dependent, being less effective if the pH is too high or low.

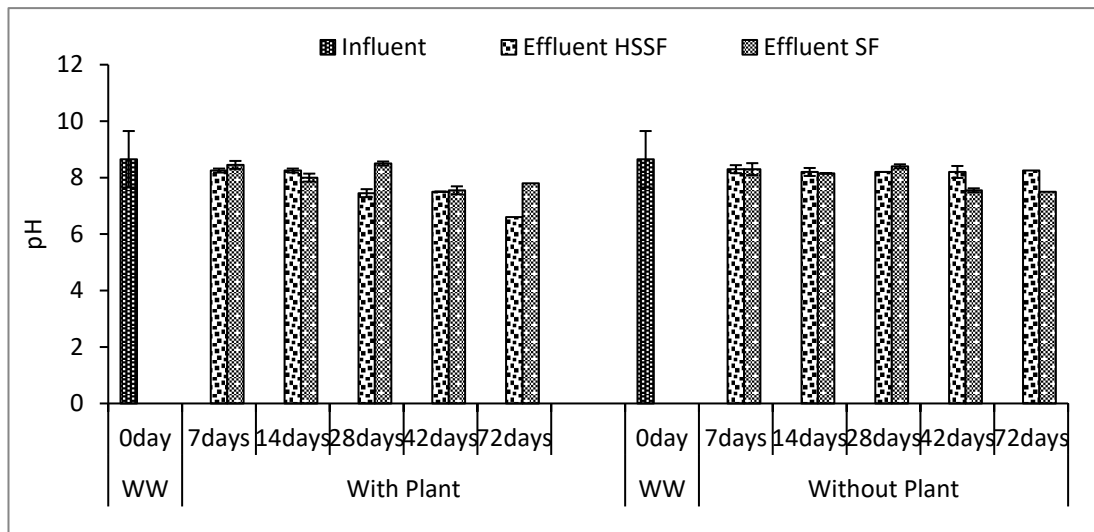


Fig.3: pH value variation during the experiment period (72 days)

Turbidity and TSS

Turbidity, the other showed a clear reduction in the systems with plant as compared to systems without plant during 72 days of the treatment (Fig.4). The minimum value 1.1 ± 0.02 NTU was recorded in HSSF after 72 days, whereas the maximum value 12.86 ± 0.02 NTU was in CC HSSF after 7 days of the treatment. Furthermore, the overall removal rate was (94.21%, 83.65%, 59.05%, and 66.05%) for SF, HSSF, CC SF, and CC HSSF, respectively. This indicates the high

efficiency of *P. australis* and *H. verticillata* to remove contaminants. Yahyapour *et al.* (2013) observed that the increase in vegetation density and decreased flow velocity resulted in more TSS and turbidity removal efficiency. Melián *et al.* (2010) also reported that the average removal for the gravel-based hybrid constructed wetland (VSSF-HSSF) was 96% for turbidity and suspended solids for urban treatment wastewater in Gran Canaria, Canary Islands, Spain.

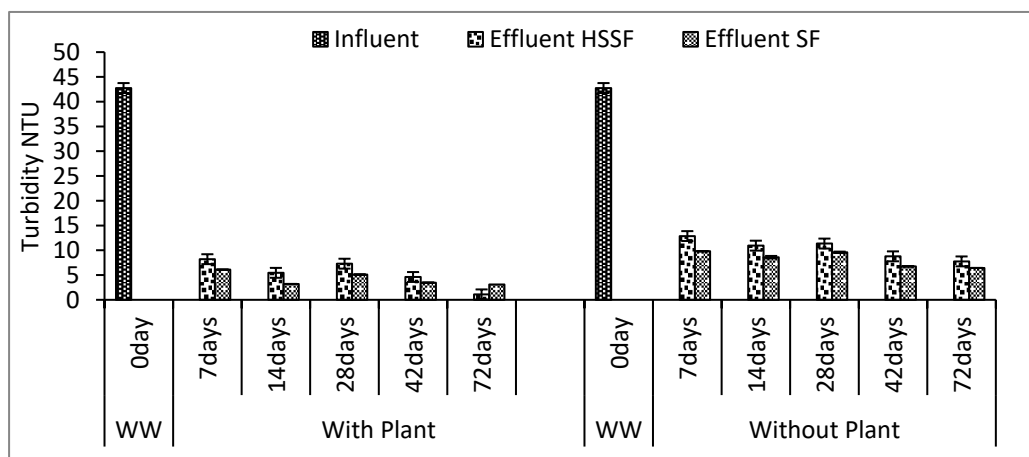


Fig.4: Turbidity NTU level during the experiment period (72 days)

According to systems, TSS gradually declined within HSSF to reach at the end of 72 day to mean value of 52.3mg/L which approximately agreed with the Iraqi standard limits for rivers (2009), which is 50mg/L while the SF system showed fluctuation in TSS values where it was dropped to its lowest point 53.33mg/L after 14 days (Fig.5). On the other hand, the highest value was recorded in CC HSSF after 7 days of the treatment. The higher removal efficiency that the hybrid system has reached during the last week was recorded in the systems with

plant (91.28% and 88.83%) compared to systems without plant (67.38% and 68.71%). The system's total removal efficiency illustrated plants' effective role in HSSF and SF systems in removing suspended solids by physical processes: sedimentation and filtration. Abdelhakeem *et al.* (2016) found that the average removal efficiency of TSS was 75% in *P.australis* planted beds compared to unplanted beds, which had 42% in HSSF CW for wastewater effluent treatment for an eight-month experiment

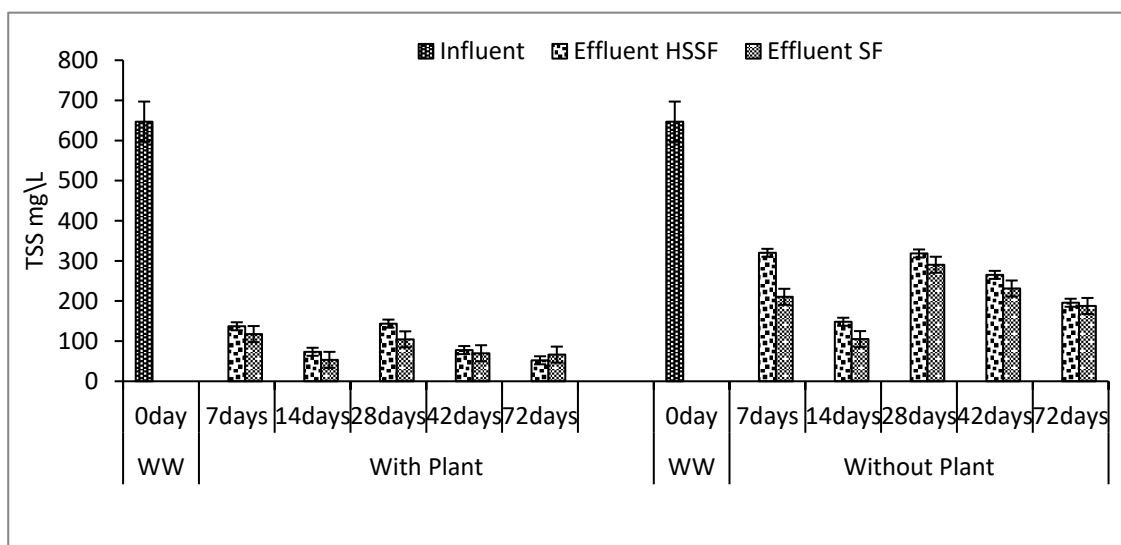


Fig.2: TSS mg/L level during the experiment period (72 days)

BOD₅ and COD

The removal efficiency of BOD depends on type of matters, where the organic compounds are degraded by aerobic and an aerobic heterotrophic microorganisms depending on the oxygen saturation conditions in the wetland bed where settleable organics rapidly removed by deposition and filtration (Abou-Elela and Hellal, 2012). Systems with plant showed higher and significant reduction of BOD₅ in comparison with systems without plant (Fig.6) where the maximum and minimum values were (10.5±0 and 1.7±0)mg/L in CC SF and HSSF. Removal efficiency which was recorded for the final discharge was 86.17%, 65.85%, 41.05% and 44.72% in HSSF, SF, CC HSSF and CC SF

, respectively. Nevertheless, SF system showed fluctuation in BOD₅ level along the period of experiment and also showed a higher value at the end of 72 day than the other planted system with mean value of 4.2 mg/L. This emphasizes the higher efficiency of *P. australis* to remove organic matter than *H. verticillata*. Haydar *et al.* (2015) showed the removal efficiency of BOD₅ was about 75% after five days of treatment using a reed (*Phragmites*) constructed wetland bed in Pakistan.

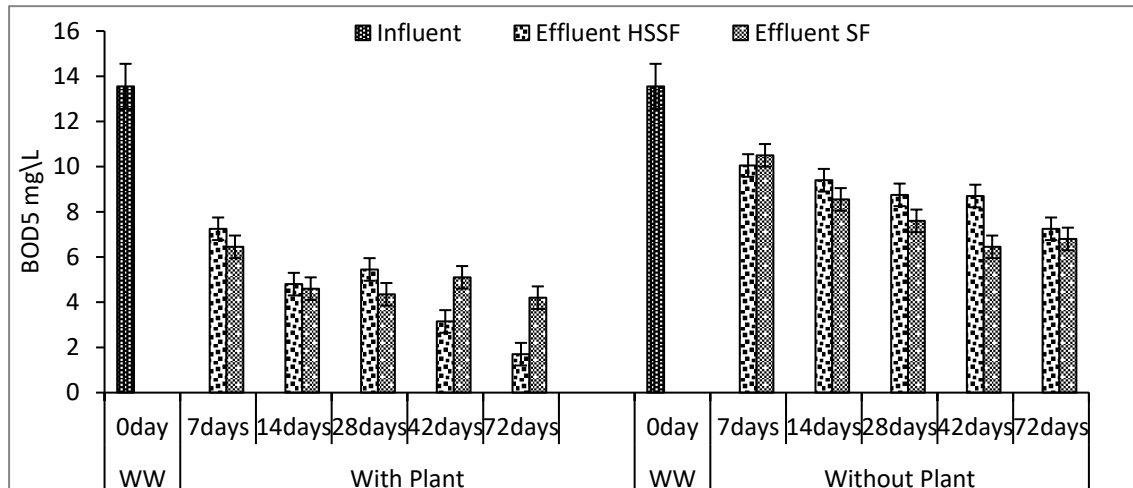


Fig.6: BOD₅ mg/L values during the experiment period (72 days)

COD, on the other hand showed a significant influence by the presence of plants and type of system as the final discharge reached to 13.3 ± 0 , 31.95 ± 0.07 , 133.1 ± 0.14 and 125.75 ± 0.21 mg/L COD in HSSF, SF, CCHSSF and CCSF, respectively (Fig.7) with percentage of removal 96.25% and 91% in HSSF and SF, while 62.54% and 64.54% in CC HSSF and CC SF, respectively after 72 days. Conversely, the retention time had no significant effect on COD level at $P \leq 0.05$. The higher efficiency of the systems with plant in removing COD compared to the systems without plant indicates that plants (*P. australis* and *H. verticillata*) were able to provide the beds with the sufficient oxygen that supports

the aerobic degradation of the organic matter in the wastewater (Abdelhakeem *et al.*, 2016). The removal efficiency of COD usually improved when the microorganisms responsible for biodegradation process is adapted (De Biase *et al.*, 2011).

The BOD₅/COD ratio (>0.2) is an important factor for treatment performance because it indicates that organic carbon is easily biodegradable from wastewater (Bulc, 2006; Justin *et al.* 2009), this ratio was on average 0.07 at the wastewater and (0.13, 0.13) at effluent of HSSF and SF, respectively while it was lower for CC HSSF and CC SF which have (0.05, 0.05)

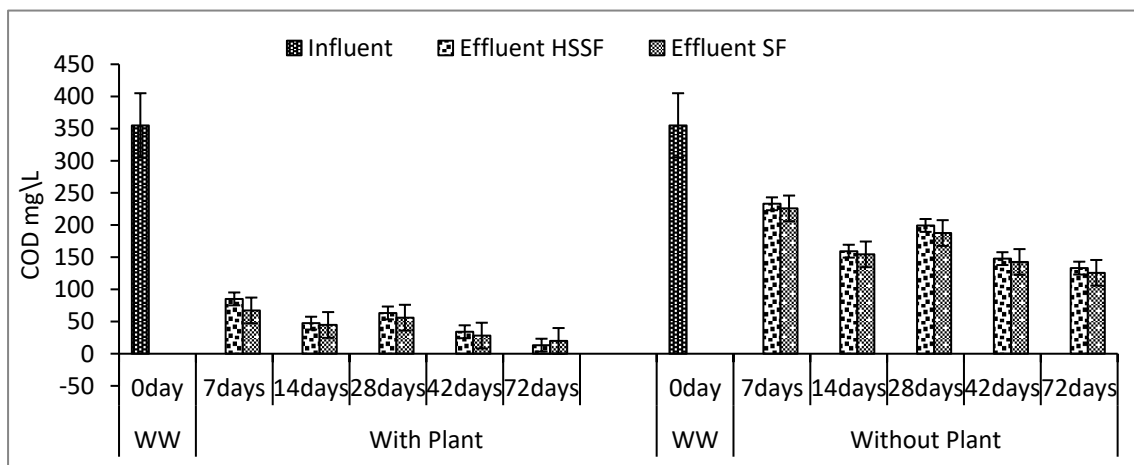


Fig.7: COD values mg/L during the experiment period (72 days)

Cl⁻ and SO₄⁻²

Chloride is the negative ion in NaCl, a naturally occurring mineral found in all water bodies in different concentrations. Generally, lower levels of Cl⁻ and SO₄⁻² ions achieved after 14 days at all hybrid system parts, but later they showed a fluctuated increase within the hybrid system during the 72 days of the experiment (Fig.8 and Fig.9). The highest value was 1480±0 mg/L and 1269.62±0.02 mg/L for Cl⁻ and SO₄⁻², respectively within CCHSSF system after 7 days while the lowest value was 610±0mg/L and 472.43±0.3mg/L for Cl⁻ and SO₄⁻², respectively within SF system after 14 days of the experiment. The significant increase in both ions after the second week could be due to the high value of the initial wastewater during that period. Another reason for that was the evaporation and physicochemical processes as the temperature has increased.

The highest removal efficiency of Cl⁻ was about (32.93%, 23.88%, 8.75%, and 9.9%) for HSSF, SF, CC HSSF, and CC SF, respectively at the end of the 72 days. Almeida *et al.* (2015) found that the removal percentage of Cl⁻ was 63.6% and SO₄⁻² was 99.9% in CWs vegetated used an aquatic macrophyte *Hymenachne grumosa*. They also mentioned that some anions such as chloride, sodium and potassium show raised values in the discharge of the wastewaters since chloride, in particular, travels freely through the wetlands to the outlet currents. Furthermore, soluble salts have a high ability to mobilize which leads to accumulate them in the plants, and as a result, they work on a

displacement of cations of heavy metals that accumulated in the sediments or water (Ouadjenia-Marouf *et al.*, 2010). However, statistical analysis showed that there were significant differences in Cl⁻ values among retention time and among system types at $P \leq 0.05$.

It has reported by Stein and Hook (2005) that the increased SO₄ level at moderate temperature periods could result from enhanced oxygen release by plant roots as respiration is reduced. It has also seen documented by Taylor *et al.* (2011) that plant root exudates for carbon could also contribute in SO₄⁻² concentrations reduction during warmer temperatures by stimulating sulfate reduction. The percentage of removal of SO₄⁻² in systems with plant was 58.83 and 55.11% in HSSF and SF, respectively while it reached in systems without plant to 32.89 and 33.41% in CC HSSF and CC SF, respectively at the end of the 72 days. On the other hand, sulfate reduction was declined due to carbon reduction which is a limiting factor for Sulfate-Reducing Bacteria (SRB) activity, that suggested by Bezbaruah and Zhang (2003). In our experiment, release of high SO₄⁻² concentrations in the effluent of all hybrid system was probably due to the oxidation of dissolved hydrogen sulfide as a result of dissolved oxygen from diffusion and plant release (Brune *et al.*, 2000). Table 2 illustrates the difference in quality of treated water between systems with plant and systems without plant in the hybrid system after 72 days of the treatment.

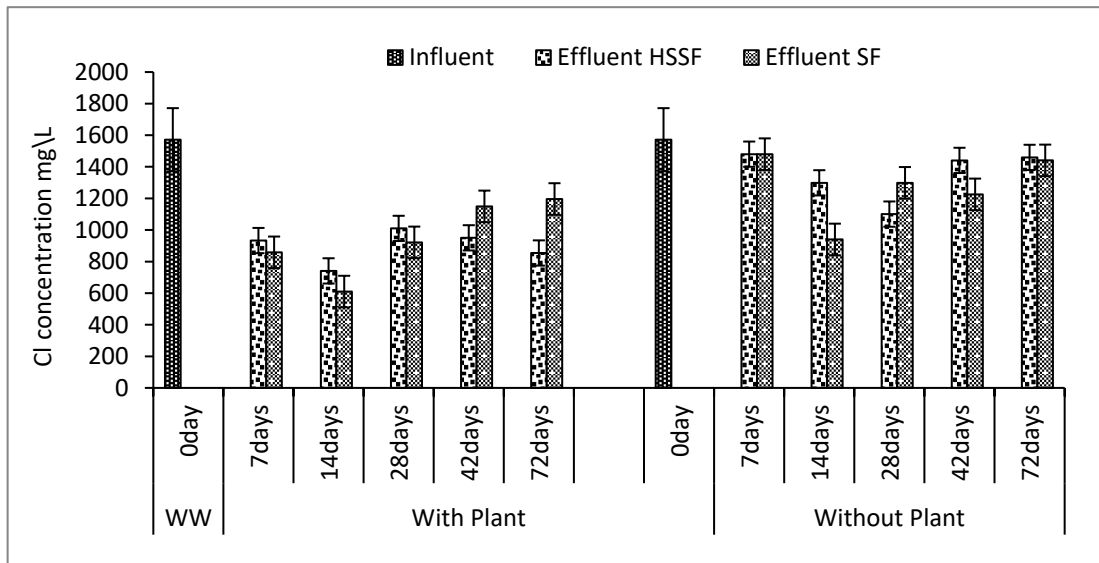


Fig.8: Cl⁻ concentrations during the experiment period (72 days)

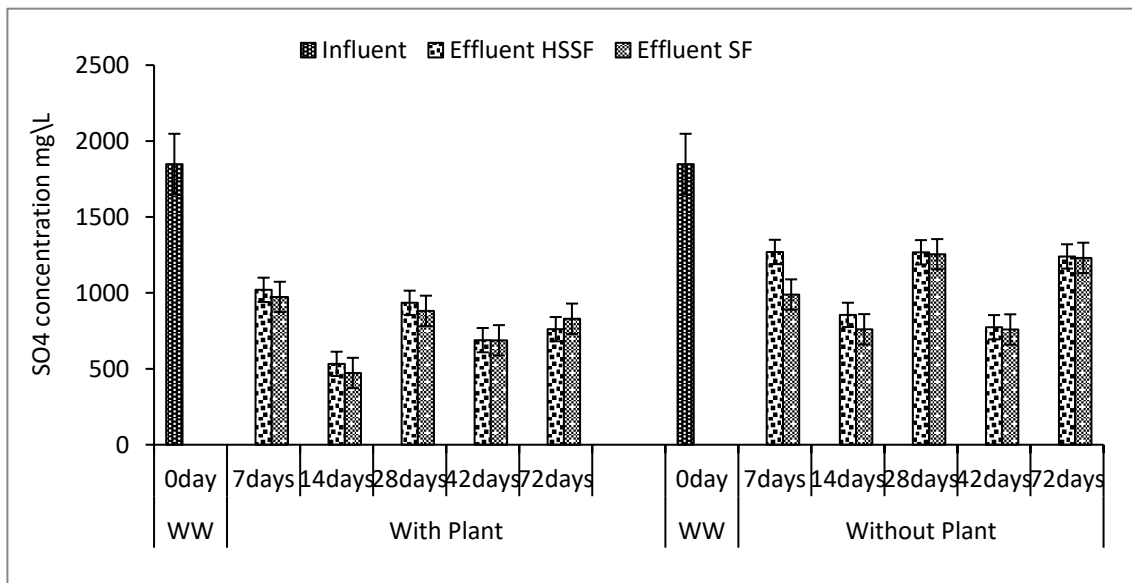


Fig.9: SO₄ concentrations during the experiment period (72 days)

Table 2: Difference in quality of treated water between systems with plant and systems without plant in the hybrid system after 72 days

Parameter	Unit	Systems with plant: mean (percentage of removal)		Systems without plants: Mean (percentage of removal)	
		HSSF	SF	CC HSSF	CC SF
Temperature	°C	14.35	14.66	15.64	15.61
PH	-	7.71	7.62	8.23	7.97
EC	mS\cm	5.5170	5.9360	9.00	8.0
TSS	mg\L	52.3(91.28%)	66.48(88.83%)	195.72(67.38%)	187.725(68.73%)
Turbidity	NTU	5.33(94.21%)	4.19(83.65%)	10.34(59.05%)	8.23(66.05%)
BOD ₅	mg\L	4.47(86.17%)	4.94(65.85%)	9.00(41.05%)	8.05(44.72%)
COD	mg\L	13.3(96.25%)	31.95(91%)	133.1(62.54%)	125.75(64.54%)

Conclusions

The performance of the continuous hybrid system was evaluated under different climatic conditions in terms of percentage of removal. It was concluded from this study that the plants had the ability to enhance water quality though a short time of treatment (72 days). It was also concluded that CW is an efficient alternative technology to remediate industrial wastewater and can deal with fluctuated organic loading conditions commonly found in industrial effluents. This study has proved the effective capability of the hybrid (HSSF-SF) system for removal high percentage of organic contaminants in terms of BOD₅ and COD in addition to particulate matters as TSS from industrial wastewater in a moderate environment area. The results demonstrated that this technology is a promising method to remediate various kinds of industrial wastewater, especially when using more than one plant species.

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نظام الأراضي الرطبة المشيدة الهجينة على مرحلتين لمعالجة مياه الصرف الصحي الصناعية

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المستخلص

تم بناء نظام هجين من مرحلتين مع معدل تدفق مستمر لمعالجة المياه المستعملة الصناعية الملوثة للغاية في محطة كهرباء النجيبية في مدينة البصرة، العراق. ويتكون النظام من نظام تدفق أفقي تحت سطح الأرض (HSSF) مزروعة بـ *Phragmites australis* (Cav.) يليه نظام تدفق السطح (SF) المزروعة بـ *Hydrilla verticillata* (L.f.) في سلسلة وكان هناك نظامان مماثلان على الجانب الآخر يمثلان نظام التحكم الملوثة. أجريت التجربة لمدة 72 يوماً. تم قياس العديد من عوامل جودة المياه في كل من مياه الصرف الصحي الخام والمعالجة، بما في ذلك درجة الحموضة (pH)، والمتطلب الحيوي للاوكسجين (BOD₅)، والمتطلب الكيميائي للاوكسجين (COD)، ومجموع المواد الصلبة المعلقة (TSS)، والعاكارة، فضلاً عن بعض الأيونات مثل أيونات الكبريتات SO_4^{2-} وأيون كلوريد

أظهرت نتائج أداء النظام أن معدل إزالة الملوثات في وجود النباتات بلغ نسبة عالية (65.85) BOD₅ (%). COD (91) (%). TSS (88.83) (%). العاكارة (83.65) (%). SO_4^{2-} (55.11) (%). Cl^- (23.88) (%). مقارنة بالنظم التي لا تعمل فيها المصانع والتي حققت (44.72) BOD₅ (%). COD (64.54) (%). TSS (68.73) (%). العاكارة (66.05) % SO_4^{2-} (33.41) (%). Cl^- (9.9) (%). (أوضحت النتائج أن الأراضي الرطبة المشيدة (CWs) هي خيار جيد لمعالجة مياه الصرف الصحي الصناعية مقارنةً بتكنولوجيات المعالجة التقليدية الأخرى المكلفة. كما تبين أن وجود المحطات له دور مؤثر في معالجة مياه الصرف الصحي. وعلاوة على ذلك، يمكن استنتاج أن نظام الزرع CW الهجين يمكن أن يعزز كفاءة إزالة الملوثات أفضل من الأراضي الرطبة التي شيدت بمفردها.

كلمات مفتاحية: أراضي رطبة مشيدة، مياه عادمة صناعية، نوعية مياه، نظام هجين، تلوث