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# BENZENE, TOLUENE, ETHYLBENZENE, XYLENE (BTEX) REMOVAL IN EXPERIMENTAL VERTICAL-FLOW CONSTRUCTED WETLANDS

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**Abstract**

Increase in the use of petroleum hydrocarbon as a source of energy has resulted in a tremendous increment in discharge of a wide diversity of hydrocarbon pollutants to the water and soil bodies. These pollutants are usually harmful and carcinogenic. Wetlands are a sustainable and cost-efficient technology to treat large quantities of these pollutants. The aim of this study is to explore the efficacy of different laboratory-scale vertical-flow constructed wetlands (VFCWs) filled with gravel and planted with common reed in degrading and removing BTEX (Benzene, Toluene, Ethylbenzene, Xylene) organic compounds as a function of different organic loading rates, media size, and contact and rest time. The wetlands were operated between September, 2013 and April, 2014 in a greenhouse located on top of Newton building, University of Salford, Manchester, UK. To evaluate the hydrocarbon pollution, approximately 130 grams of diesel fuel was poured into each of four wetland filters which is equivalent of a one-off inflow concentration of 20,000 mg/l. A range of hydraulic loading rates was applied across the systems using real urban wastewater. Analysis of BTEX hydrocarbon concentrations of effluent waters along with other water quality parameters were carried out to monitor both removal efficiency and treatment performance variations. The results indicated that all the BTEX hydrocarbon components treated in the wetland filters were highly degraded with high removal efficiency (above 90%) in all contaminated filters with some even attenuated below detection limit. However, with regards to water quality parameters, filters contaminated by hydrocarbon performed worse in terms of COD and BOD, but considerably better regarding nitrate-nitrogen, ortho-phosphate phosphorus and ammonia-nitrate removal. This result has shown the robustness of VFCWs in eliminating BTEX organic compounds in urban waste-water. The successful removal of the hydrocarbon compounds and other pollutants will make constructed treatment wetlands very attractive and sustainable technology in environmental pollution control.

**Keywords**: Benzene, toluene, ethylene, xylene

## Introduction

Global geometric population increase complemented with vociferous increase in urbanization, industrial and agricultural land use has ensured an enormous increase in a discharge of a wide diversity of pollutants including petroleum hydrocarbons to receiving eco-systems. Example of these hydrocarbons are BTEX, an acronym of Benzene, Toluene, Ethylbenzene, and Xylene. These are common pollutants found in petroleum industries effluents. BTEX is one of the members of Polyaromatic hydrocarbons (PAH) and when released to the receiving environments leads to negative impacts such as hydrophobicity, fish kills, reduced dissolved oxygen on water ecosystem. On the other hand, in soils, it leads to a reduction in aggregate stability with poor soil structure and texture, susceptible to wind and water erosion, reduced soil organic carbon and nitrogen on soil ecosystem respectively (Sani, 2015; Almuktar et al., 2018).

Furthermore, in the BTEX, benzene is a human carcinogen which promotes myeloid leukemia, toluene exposure causes reproduction effects, ethylbenzene affects the blood, liver and kidneys, and finally xylene exposure affects the central nervous system leading to cardiovascular and respiratory problems (Cho et al., 2009; Mathur and Balomajumder, 2013) when ingested by animals via food chain. In addition, the water and soil quality deterioration by these BTEX compounds makes the water unsuitable for irrigation and soil poor for crop production. Hence, their removal is very imperative.

The common technologies available for BTEX elimination includes adsorption, absorption, condensation, thermal incineration and membrane separation (Sani, 2015). However, these technologies are not applicable everywhere due to high cost and generation of unwanted by-products during the process of the depuration. Unlike these conventional technologies, phytoremediation treatment technology such as constructed wetlands could be environmentally friendly, easy to operate, less energy-intensive, and cost-effective.

Several studies have documented that constructed wetlands (CWs) are engineered systems used worldwide as a result of the aforementioned qualities (Sani et al., 2013; Al-Isawi et al., 2016; Almuktar et al., 2018) and most preferred efficient tool for water and soil pollution control to treat variety of waste waters including agricultural and urban runoff, industrial effluents, animal waste waters, sludge and mine drainage, petroleum wastewaters and recently applied successfully to treat domestic waste water contaminated with hydrocarbons (Sani, 2015; Al-Isawi et al., 2016; Almuktar e al., 2018). The systems are now gaining reputation and acceptance globally (Almuktar et al., 2018) as a tool for environmental pollution control as their function principle is akin to that of natural wetlands and the purification process involves the combination of wetland plants, soils, and other microbial organisms to aid in the treatment of the waste water (Scholz, 2010; Sani, 2015).

Many studies revealed that BTEX compounds in wetlands are attenuated via aerobic and anaerobic degradation (Chen et al., 2012) while some reported removal as a result of microbial rhizodegradation (Chen et al., 2012; Sani, 2015) which is carried out efficiently by the wetland microbes in wetland environments because the wetland plant roots aggregate the soil particles and provide a suitable surface area for bacteria growth and attachment (Chen et al., 2012) that feed on and remove the hydrocarbon compounds. However, some authors in their researches attributed the BTEX compounds depuration to volatilization and phytovolatilization in addition to biodegradation since they are volatile in nature (Imfeld et al., 2009), though, the volatilization removal is lower than the other elimination processes (Al-Isawi et al., 2014, 2015).

This study may provide useful information to petroleum, agricultural and related water industries to incorporate wetland systems in their wastewater treatment technologies particularly for abatement of BTEX compounds in petroleum hydrocarbon spills that may be released in sewage alone or discharged with industrial wastewater, from small factories, public utilities, and with domestic sewage that will otherwise pollute the soil and water ecosystems.

Considering the negative implication of these BTEX compounds to human health and environment, and wetland significance in water and soil pollution control, this study is aimed to assess the efficacy of these eco-technology systems in removing BTEX compounds in temperate climates. Many studies have been conducted in the UK on hydrocarbon treatment in wetlands, but only few, if any, have focused on depuration and degradation of these BTEX compounds in vertical-flow systems treating urban wastewater contaminated with hydrocarbon spill (Eke and Scholz, 2008; Tang et al., 2010; Al-Isawi et al., 2015, 2016; Almuktar et al., 2018). Therefore, the aim of this study is to evaluate the performance of vertical-flow constructed wetlands in removing different BTEX compounds with the following objectives;

1. To determine the effect of different wetland systems in removing and degrading BTEX compounds and other pollutants;
2. To assess the effect of different operational and design variables on the BTEX and other contaminants removal from the wetland systems.

## Materials and Methods

Ten laboratory-scale vertical-flow constructed wetlands were constructed from Pyrex tubes with an inner diameter of 19.5 cm and a height of 120 cm. The filters were filled with siliceous (minimum of 30%) pea gravel up to a depth of 60 cm and planted with Phragmites australis (Cav.) Trin. ex Steud. (Common Reed) to investigate performance of different filters in terms of aggregate size, hydraulic and contaminant loading rate, contact time and resting time .The preliminary treated urban wastewater used for the inflow water was obtained from the Davyhulme Sewage works, Greater Manchester. Fresh wastewater was collected approximately once per week, and was stored and aerated by standard aquarium air pumps in a cold room before use.

In order to simulate a one-off hydrocarbon fuel (100% pure; no additives) spill, 130 gram (equivalent to an inflow concentration of 20 g/l) of diesel fuel (100% pure; no additives) were poured into Filters 1, 3and 7, and into one of the two columns (Control A) on 26 September 2013 to assess BTEX compounds as the model hydrocarbon. The fuel was obtained from a petrol station operated by Tesco Extra (Pendleton Way, Salford, UK). Note; Filters 2, 4 and 8 are replicates for the most common operational scenarios. Furthermore, the COD was used as the criterion to differentiate between low and high loads. An inflow target COD of about 273 mg/l (usually between 122 and 620 mg/l) was set for wetlands with a high loading rate (Filters 7 and 8). The remaining Filters 1, 2, 3, 4, 9 and 10 received wastewater diluted with de-chlorinated tap water. The target inflow COD for these filters was approximately 139 mg/l (usually between 43 and 350 mg/l).

Filters 1 and 2 compared to Filters 3 and 4 tested the influence of a larger aggregate diameter. Filters 7 and 8 compared to Filters 3 and 4 checked the impact of a higher loading rate. The application of a lower contact rate is tested if Filter 9 is compared with Filters 3 and 4. Finally, a lower resting time is the difference between Filters 9 and 10. Undiluted wastewater was introduced to wetlands with a high loading rate (Filters 7 and 8). The remaining Filters 1 to 4 and Filters 9 and 10 received wastewater diluted with de-chlorinated tap water. All wetland columns received 6.5 l of inflow waste water during the feeding mode.

**Water quality analyses:** Routine obstruction observations and water quality sampling were carried out according to APHA (2005) unless stated. The spectrophotometer DR 2800 Hach Lange (www.hach.com) was used for standard water quality analysis for variables including chemical oxygen demand (COD), ammonia-nitrogen (NH4-N), nitrate-nitrogen (NO3-N) and orthophosphate-phosphorus (PO4-P). The five-day biochemical oxygen demand (BOD5) was determined in all water samples with the OxiTop IS 12-6 system, a manometric measurement device, supplied by the Wissenschaftlich-Technische Werkstätten (WTW), Weilheim, Germany. Nitrification was suppressed by adding 0.05 mL of 5 g/l N-Allylthiourea (WTW chemical solution No. NTH600) solution per 50 ml of sample water. The pH was measured with sension+Benchtop Multi-Parameter Meter (Hach Lange, Düsseldorf, Germany). Temperature data were supplied by the UK Meteorological Office (www.metoffice.gov.uk).

**BTEX analysis:** BTEX compounds were determined by gas chromatography and flame ionization by Exova Health Services (Hillington park, Glasgow, UK) according to their own accredited "Hydrocarbon in Waters (with Aliphatic/Aromatic Splitting) Method" (Exova Health Services, 2014).

**Data analysis:** After data collection, data were subjected to normality test before validation and subsequent analysis. Because of high variability, the data were not normally distributed even after transformations with transformers such as arc sine, square root, log, etc. and as a result, easy statistical tools that will fit the abnormal distributed data such as nonparametric Mann-Whitney U-test using IBM SPSS Statistics Version 20 were sought and applied, while Microsoft Excel (www.microsoft.com) was used for the general descriptive data analysis.

## Results and Discussion

### Influent and effluent water quality

Table 1 and 2 show the overall experimental set-up of the research and influent water quality before dilution for the entire experimental period. Table 2 shows the overall inflow water quality of the undiluted domestic inflow water quality evaluated from 26th September, 2013 to April 2014 when some selected filters are subjected to one-off hydrocarbon spill to assess BTEX compounds as a target hydrocarbons. The undiluted influent concentrations for COD, BOD, ammonia-nitrogen, nitrate-nitrogen and ortho-phosphate-phosphorus were 246 mg/l, 133 mg/l, 32 mg/l and 4 mg/l, respectively while the hydrocarbon spill was 130,000mg/l.

Table 1: Experimental wetlands set-up used for the research

| Wetland filters | Design and operational variables | | | |
|---|---|---|---|---|
| Filters | Aggregate size (mm) | Contact time (hours) | Resting time (hours) | Chemical oxygen demand (mg/l) |
| Filters 1 and 2 | 20 | 72 | 48 | 138.9 |
| Filters 3 and 4 | 10 | 72 | 48 | 138.9 |
| Filters 7 and 8 | 10 | 72 | 48 | 272.9 |
| Filters 9 | 10 | 36 | 48 | 138.9 |
| Filters 10 | 10 | 36 | 24 | 138.9 |
| Control A | 10 | 72 | 48 | 2.1 |
| Control B | 10 | 72 | 48 | 2.1 |

Table 2: Inflow water quality: undiluted domestic waster from 26/09/2013 to 30/04/2014 when some selected filters are subjected to one-off diesel spill.

| Parameters | Unit | Number | Mean | Minimum | Maximum | Standard deviation |
|---|---|---|---|---|---|---|
| COD | mg/l | 16 | 246.1 | 112.0 | 360.0 | 93.02 |
| BOD | mg/l | 68 | 133.3 | 10.0 | 360.0 | 98.45 |
| $NH_4$-N | mg/l | 22 | 32.4 | 3.1 | 70.0 | 24.06 |
| $NO_3$-N | mg/l | 20 | 3.7 | 0.4 | 14.0 | 4.32 |
| $PO_4$-P | mg/l | 18 | 16.3 | 9.3 | 27.6 | 113.3 |

Note: only filters 7 and 8 received the above water characteristics. The remaining filters received diluted waste water (i.e. 1 part dechlorinated tap water and 1 part waste water)

**Comparison of BTEX hydrocarbon outflow Water Qualities**

**Oxygen Demand Variables (COD and BOD)**

Table 3 shows the comparison of the outflow water quality of the wetland filters in mg/l during the period of hydrocarbon contamination. The result indicated that filters with BTEX hydrocarbon contamination (1, 3, 7 and control A) recorded poor COD removal efficiencies (< 35%) and relatively lower BOD removal efficiencies (66-81%) in comparison to their corresponding filters without BTEX hydrocarbons (35-75% of COD) and (78-87% of BOD). This indicates that BTEX hydrocarbons resulted in a sharp decline of the removal efficiency of the COD in the contaminated wetland filters possibly because of the indirect artificial contribution of the COD in the inflow water, which might have raised the outflow COD values observed (Al-Isawi et al., 2014, 2015) since it has been reported that hydrocarbon compounds including BTEX are associated with high COD values (Scholz, 2010). On the other hand, the relative high COD and BOD removal efficiencies recorded in the uncontaminated wetland filters could be attributed as a result of gradual improvement in macrophytes growth and wetland microbe's acclimation as the wetland systems mature, subsequently, leading to the recorded high pollutants biodegradation (Sani et al., 2013).

The overall mean COD and BOD removal efficiencies for Filter 8 without BTEX hydrocarbons is higher compared to filter 7 with BTEX hydrocarbons (both with high loading rate). This difference was not statistically significant as shown in Table 4. The overall removal efficiencies were also higher for filters without BTEX hydrocarbons than those with, though the difference is not much with regards to BOD (Table 3). A comparison between filters 3 and 4 has shown a statistical significant difference on hydrocarbon effect. The relative poor performance of the polluted filters can be explained by the high inflow loading rates, which could also be attributed to the influence of the high applied BTEX hydrocarbons in the influent waste water as mentioned above. However, there were no significant differences with all polluted filters observed when period of pollution was compared with the unpolluted one (Table 4).

Pertaining COD however, there was a significant difference in all filters polluted with BTEX hydrocarbons during the contamination period if compared with the corresponding period before they are contaminated (Table 4) which could probably be due to the earlier stated reason (Al-Isawi et al., 2014, 2015). The Urban Waste Water Treatment (England and Wales) Regulations (UK Government, 1994), which implements the Council Directive 91/271/EEC Concerning Urban Waste Water Treatment (European Community, 1991), set a threshold value of 125 mg/l of COD and 25mg/l of BOD for secondary waste water treatment respectively. All filters were non-compliant in the period of BTEX hydrocarbons contamination.

**Comparison of nutrients variables (N and P)**

Table 3 also indicated that overall removal rates of ammonia-nitrogen were relatively high ranging from 57% to 84% in all filters regardless of contamination (though better in low loading rate filters than high loading ones). This high removal of ammonia-nitrogen observed could be attributed to the fact that intermittent aeration, increase in aerobic bacteria and established macrophytes growth as a result of improved wetland maturation that took place overtime, might have promoted the nitrification process leading to high ammonia removal (Fan et al., 2013).

Regarding statistical difference between the variables, all filters regardless of contamination period and whether or not contaminated, showed no statistical significant differences (Table 4). A typical standard realistic guideline threshold value of ammonia-nitrogen concerning secondary wastewater treatment was set to be 20 mg/l (Sani et al., 2013). After the BTEX contamination, filters 1 and 7 were non-compliant while filter 3 complied.

Despite the fact that nitrate-nitrogen concentration in the influent wastewater was relatively low, the effluent concentrations were relatively high for all filters. Only Filters 1, 3, 7 and 8 had positive removal efficiencies. In contrast, all other filters functioned as sources for nitrate-nitrogen. The negative removal efficiencies for nitrate-nitrogen indicated that denitrification was likely to be only a minor removal mechanism. The overall removal efficiency is relatively high with no much difference between the filters (Table 3). This nitrogen removal could be attributed to high biodegradation processes of BTEX hydrocarbon spills in the contaminated filters, which overtime promoted the growth of some micro-organisms, provided high source of carbon and energy (Tang et al., 2010), subsequently stimulating the nitrogen reduction via BTEX hydrocarbon degradation by the wetland microbes (Scholz, 2010).

Statistically, overall daily nitrate-nitrogen mean values of all filters with or without BTEX hydrocarbons were not significantly different from each other (Table 4). A common standard set by environment agencies for the nitrate-nitrogen variable concerning secondary treatment of wastewater is 50 mg/l (Sani et al., 2013) and all filters were compliant. In ortho-phosphate-phosphorus, the results indicated that the removal efficiencies ranged between 58 and 74% for all filters regardless of the loading rate and BTEX hydrocarbon contamination (Table 3). This nitrogen removal could be attributed to high biodegradation processes of BTEX hydrocarbon spills in the contaminated filters, which overtime promoted the growth of some micro-organisms, provided high source of carbon and energy

Table 3: Comparison of the outflow water quality of the wetland filters in mg/l during the period of hydrocarbon contamination (26/09/2013 to 30/04/2014)

| Parameters | Wetland Filters | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| COD | 1 | 2 | 3 | 4 | 7 | 8 | 9 | 10 | CA | CB |
| Mean | 108.0 | 48.2 | 115.3 | 42.1 | 160.7 | 61.7 | 39.5 | 47.6 | 35.1 | 35.1 |
| Std | 94.4 | 35.82 | 87.98 | 36.72 | 113.06 | 47.47 | 36.56 | 34.2 | 99.31 | 36.8 |
| Removal% | 12.3 | 61.0 | 6.7 | 65.9 | 34.7 | 75.0 | 68.0 | 61.5 | Ned | Ned |
| BOD | | | | | | | | | | |
| Mean | 22.4 | 13.9 | 25.7 | 13.0 | 25.7 | 18.0 | 13.9 | 14.7 | 8.1 | 8.1 |
| Std | 16.34 | 8.82 | 19.31 | 10.33 | 19.98 | 13.6 | 9.80 | 8.71 | 10.58 | 8.49 |
| Removal% | 66.6 | 79.0 | 61.4 | 80.5 | 80.7 | 86.5 | 79.0 | 77.8 | Ned | Ned |
| $NH_4$-N | | | | | | | | | | |
| Mean | 6.6 | 6.2 | 4.3 | 4.9 | 14.1 | 12.9 | 4.8 | 2.8 | 2.0 | 2.0 |
| Std | 7.99 | 6.21 | 4.82 | 4.81 | 15.89 | 15.62 | 7.96 | 5.2 | 1.71 | 1.95 |
| Removal% | 63.8 | 65.7 | 76.6 | 73.2 | 56.5 | 60.1 | 73.4 | 84.2 | Ned | Ned |
| $NO_3$-N | | | | | | | | | | |
| Mean | 0.5 | 3.4 | 0.5 | 0.6 | 1.1 | 3.5 | 4.3 | 3.7 | 0.6 | 0.6 |
| Std | 0.26 | 3.24 | 0.36 | 0.41 | 0.81 | 5.57 | 3.47 | 4.38 | 0.61 | 0.41 |
| Removal% | 76.2 | -65.8 | 77.7 | -73.4 | 71.6 | 6.9 | -114.8 | 88.2 | Ned | Ned |
| $PO_4$-P | | | | | | | | | | |
| Mean | 3.3 | 3.1 | 3.0 | 3.1 | 4.6 | 4.2 | 3.2 | 3.5 | 3.5 | 3.5 |
| Std | 3.09 | 1.34 | 2.80 | 1.31 | 4.19 | 3.86 | 2.05 | 2.28 | 2.05 | 2.28 |
| Removal% | 61.2 | 63.7 | 64.7 | 63.2 | 71.6 | 74.3 | 61.8 | 58.3 | Ned | Ned |

Note; Air temperature is in ˚C, with mean of 11.3, minimum and maximum of 20.0 throughout the period of the research and Ned, no enough data.

Table 4: Overview of the statistically significant differences between P values regarding outflow water quality variables (mg/l) of different wetland filters using the non-parametric Mann-Whitney U-test during the period of hydrocarbon contamination (26/09/13 to 19/03/14)

| Parameters | Effect of BTEX hydrocarbon | | | | | |
|---|---|---|---|---|---|---|
| | a | B | c | d | e | f |
| COD | 0.126 | 0.126 | 0.089 | 0.028 | 0.002 | 0.000 |
| BOD | 0.054 | 0.003 | 0.146 | 0.052 | 0.411 | 0.049 |
| $NH_4$-N | 0.355 | 0.348 | 0.297 | 0.527 | 0.411 | 0.484 |
| $NO_3$-N | 0.149 | 0.275 | 0.275 | 0.322 | 0.105 | 0.054 |
| $PO_4$-P | 0.564 | 0.564 | 0.149 | 0.266 | 0.138 | 0.141 |

[a]comparison between filters 1 and 2; [b]comparison between filters 3 and 4; [c]comparison between filters 7 and 8; [d]comparison between filter 1 pre and post hydrocarbon; [e]comparison between filter 3 pre and post hydrocarbon; and [f]comparison between filter 5 pre and post hydrocarbon. Note P-value is a probability of getting a test statistic at least as extreme as the one that was actually observed. Filters are statistically significantly different only if the p value <0.05 for the corresponding water quality parameter.

(Tang et al., 2010), subsequently stimulating the nitrogen reduction via BTEX hydrocarbon degradation by the wetland microbes (Scholz, 2010). Statistically, overall daily nitrate-nitrogen mean values of all filters with or without BTEX hydrocarbons were not significantly different from each other (Table 4). A common standard set by environment agencies for the nitrate-nitrogen variable concerning secondary treatment of wastewater is 50 mg/l (Sani et al., 2013) and all filters were compliant. In ortho-phosphate-phosphorus, the results indicated that the removal efficiencies ranged between 58 and 74% for all filters regardless of the loading rate and BTEX hydrocarbon contamination (Table 3). The relatively high ortho-phosphate phosphorus removal can be attributed to gradual maturity and improvement of the wetland systems, established biomass as a result of macrophytes growth, and high aeration due to intermittent feeding mode and microbial acclimatization, which might have enhanced the high phosphorus reduction.

Effect of BTEX hydrocarbon contamination of the wetland filters on water quality parameters were not statistically significantly different in terms of overall ortho-phosphate-phosphorus treatment (Table 4). The regulations concerning environmental agencies set a threshold value of 1 mg/l for ortho-phosphate-phosphorus removal from secondary wastewater (Sani et al., 2013). All filters were non-compliant both the polluted and the unpolluted ones.

**BTEX hydrocarbon removal and degradation in the wetland filters**

Table 5 shows BTEX petroleum hydrocarbons evaluated in this research and its constituents with their corresponding concentration values in the influent wastewater before and after their treatment in the wetland filters. The analyzed constituents are benzene, toluene, ethylene and xylene. However, in all these BTEX compounds, toluene and ethylene were found to be below the 10 µg/l detection limit in Filters 2, 4, 8, 9, 10 and control B while benzene and xylene recorded very low in filters 9 and 10 outflow concentrations respectively indicating very little or none of the pollutant compounds in the background inflow wastewater of these low loading filters since they are not contaminated with the petroleum hydrocarbons. Furthermore, these BTEX petroleum hydrocarbons have also shown to be in the range of less than 10 µg/l in both polluted and unpolluted filters in spite of their high concentration in the hydrocarbon fuel. However, in the contaminated filters, except for the control A that recorded 92% benzene removal, they recorded 98% removal of BTEX compounds regardless of filter operation or design (Table 5). This can be explained by high degradation activity as a result of high microbial population in the wetland filters due to high amount of nutrients from the undiluted waste water received overtime in addition to that from the hydrocarbon compound as source of carbon and energy (Awe et al., 2008).

A common standard set by environment agencies for BTEX hydrocarbons concerning secondary treatment of wastewater is 300 ug/l (Grontimij, 2015), and all filters complied. However, this result of the BTEX hydrocarbon removal efficiencies for each components in different wetland filters, including natural background concentrations in the raw wastewater are based only on one-off sample. Overall, removal efficiency is relatively very high for all BTEX components with no much difference between the filters except in controls (Table 5). The observed good performance in the different wetland filters could be explained by the fact that high aeration as a result of intermittent operation mode applied to the wetland systems, established reed growth and biomass achieved overtime due to wetland maturity, and high microbial activity could have enhanced the biodegradation and removal of the hydrocarbons efficiently (Albaldawi et al., 2014; Al-Isawi et al., 2016). Moreover, the BTEX compounds have concentrations below 10ug/l in almost all filters including controls regardless of contamination, design or operation (Table 5) indicating their very high removal (>90%). This could be attributed to the combined effect of volatilization and phytovalatilization in addition to biodegradation since they are volatile in nature (Imfeld et al., 2009).

Table 5 Overall overview of the analysis of BTEX petroleum hydrocarbon compounds in different wetland filters ( 26/09/13 to 19/03/14)

| Analyte (ug/l) | | Wetland filters | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| BTEX | Method | Filter 1 | Filter 2 | Filter 3 | Filter 4 | Filter 7 | Filter 8 | Filter 9 | Filter 10 | Control A | Control B | Inflow | Hydrocarbon |
| Benzene | AN15a | <10 | <10 | <10 | <10 | <10 | <10 | 18 | 19 | 39 | <10 | 459 | 65120 |
| Removal% | | 97.8 | 97.8 | 97.8 | 97.8 | 97.8 | 97.8 | 96.0 | 95.8 | 91.5 | 97.8 | Na | Na |
| Toluene | AN15a | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | 473 | 302300 |
| Removal% | | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | 97.9 | Na | Na |
| Ethylene | AN15a | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | <10 | 532 | 9405 |
| Removal% | | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | 98.1 | Na | Na |
| Xylene | AN15a | <10 | <10 | <10 | <10 | <10 | <10 | 20 | 22 | 42 | <10 | 489 | 34890 |
| Removal% | | 98.0 | 98.0 | 98.0 | 98.0 | 98.0 | 98.0 | 95.9 | 95.5 | 91.4 | 98.0 | Na | Na |

**Conclusion**

The overall findings indicated that all the BTEX hydrocarbon components treated in the wetland filters were highly degraded (>90% removal efficiency) in all filters regardless of contamination, design or operation. However, with regards to water quality parameters, the filter with the highest COD loading but no BTEX hydrocarbon contamination performed the best in terms of COD and BOD removal. Filters contaminated by diesel performed worse in terms of COD and BOD, but considerably better regarding nitrate-nitrogen removal. However, some filters including both contaminated and uncontaminated, recorded negative values in their outflow concentration for nitrate-nitrogen, indicating that the filters served as a source for the nutrient.

The result of this research indicates the robustness of vertical-flow constructed wetlands under intermittent operation mode in eliminating BTEX hydrocarbon organic compounds and other conventional contaminants in urban waste-water even at shock loading, simultaneously protecting the environment. The successful removal of these BTEX hydrocarbon compounds and other pollutants will make constructed treatment wetland very attractive and sustainable technology capable of meeting zero discharge goal in the production, storage, refining and transportation sectors of the oil and gas industries.

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