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TREATMENT ALTERNATIVES FOR WASTEWATER EFFLUENTS FROM A MEDIUM SCALE PHARMACEUTICAL INDUSTRY

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Abstract:- *This research work examined the application and capacity of using constructed wetlands, as alternative secondary treatment system on industrial wastewater from a medium scale pharmaceutical company. Four parallel pilot-scale sub-surface flow constructed wetland systems with dimensions: 1800 mm (L); 900 mm (W); 900 mm (D) each, were installed at the Department of Civil Engineering, LAUTECH Ogbomoso, Nigeria for conducting the proposed phytoremediation study. Four different emergent wetland plant species including Wild Sorghum (Sorghum arudinaceum), White Lotus (Nymphaea lotus), Yerba De Jicotea (Ludwigia erecta), and Ginger-leaf morning-glory (Ipomoea asarifolia) were used. The averaged influent contains approximately 160 FTU turbidity, 140 mg/l total hardness, 3.1 mg/l Iron, 8.9 mg/l Nickel, 12 mg/l Zinc, 5465 mg/l oil and grease, 0.4 mg/l chemical oxygen demand (COD), 0.26 mg/l biochemical oxygen demand (BOD), and 0.27 mg/l ammonium. The wetland cells were fed with wastewater collected from a pharmaceutical industry nearby the University campus and treated effluents were collected for analyses at 7- day interval for a retention period of 28 days. The results obtained showed that phytoremediation reduced over sixty percent (64.1%) turbidity, 71.4 % total hardness, 42 % chloride, about 34 % sulphate. Nitrate and phosphate were removed by 38.8 % and 57.8 % respectively. Metal contents reduction in treated wastewaters such as 61.5 % aluminum, about fifty percent (49.4 %) Iron, 38.2 % Nickel, 49.8 % Zinc, were recorded. Yerba De Jicotea (Ludwigia erecta) reduced BOD and COD by 65.4 % and 65 % respectively, while Ginger-leaf morning-glory (Ipomoea asarifolia) removed 8.3 % of oil and grease as the highest removal efficiency. Averagely, wild Sorghum (Sorghum arudinaceum) possessed the optimal phyto-remedial efficiency of the four emergent wetland plants. Treated effluents proved the efficacy of constructed wetland, using locally available macrophytes as an effective secondary treatment technology for pharmaceutical wastewater. The treated effluents meets the current industrial wastewater discharge standards in Nigeria.*

Keywords: *Constructed wetland, Industrial wastewater, Removal efficiency, Sorghum arudinaceum, Nymphaea lotus, Ludwigia erecta, Ipomoea asarifolia*

1. INTRODUCTION

Around the world, large number of Pharmaceuticals of different therapeutic classes are consumed annually, in order to diagnose, prevent and mitigate or cure diseases in humans as well as animals (Jones et al., 2007; Uslu et al., 2013). Studies in analytical techniques have showed trace pharmaceutical residues occurrences in water environment (Verlicchi et al., 2012). Municipal wastewater remains the main channel through which pharmaceuticals get into the water environment. Some of these pharmaceuticals in humans and animals bodies are partially metabolized and ingested, as a result, are excreted with urine and feces into sewage system. Expired and abandoned pharmaceuticals in houses are usually disposed into toilets, and were subsequently introduced into the sewage system. (Zhang et al., 2008). Pharmaceutical loadings in municipal wastewater were also influenced by wastewater from hospitals or pharmaceutical manufacturers (Jones et al., 2001; Santos et al., 2013). In conventional wastewater treatment technologies, pharmaceuticals were not completely removed (Petrie et al., 2013; Repice et al.2013; Vidal-Dorsch et al., 2012), and were released via wastewater treatment plants effluents, contaminating various water sources such as surface water, ground water, coastal water, and even drinking water (Uslu et al., 2013; Vidal-Dorsch et al., 2012).

Domestic effluents from communities and households, surface runoff, wastewater from animal husbandries and agricultural effluents from rural areas contributed direct channels of pharmaceuticals contaminations to fresh and marine water environments (Anderson et al., 2013;), and on reaching the water bodies became potential risks to the health of aquatic life and human beings even at trace levels in the water environment (Pomati et al., 2006).
Advanced technologies have been studied to evaluate their effectiveness for removal of pharmaceuticals contaminants from wastewater. These have been reported by Trinh et al., 2012. However, considering their expensive nature at large-scale applications, selecting low-cost alternative technologies for pharmaceutical treatment becomes of great significance, especially in the low income regions. Constructed wetlands have proven to be an exciting alternative for several types of

industrial wastewaters that are to be treated by biological means (Cristina et al., 2009). Hence, constructed wetlands has been recognized as an accepted affordable eco-technology (Jing et al., 2001).

The removal of contaminants from wastewater sources such as domestic, industrial, municipal, agricultural, mine drainage, leachate, polluted groundwater and urban runoff in the recent times, has been practiced through the engagement of constructed wetlands (Oladejo and Olanipekun, 2018; Oladejo et al., 2015; Oladejo 2018; García et al., 2010; Vymazal, 2009). However, treatment of pharmaceuticals pollutants via constructed wetland treatment methods is a new idea and a fresh application field. Hence, need for more attention on present and future research studies.
The objective of this paper is to examine the efficiency of using constructed wetlands for the treatment of wastewater from pharmaceutical industry and application of constructed wetlands as alternative secondary wastewater treatment system.

2. MATERIALS AND METHODS

A. Experimental Site and Vegetation Description

The Four parallel pilot-scale sub-surface flow constructed wetland systems used in this study was designed to be 1800 mm (Length) x 900 mm (Width) x 900 mm (Depth) using substrates such as washed granite of 12 mm diameter and covering a total depth of 200 mm, washed sand covering a depth of 400 mm and humus soil covering a depth of 150 mm which were all properly filled from bottom to the top so as to support the growth of the macrophytes. A 150 mm freeboard was also allowed (Oladejo and Olanipekun, 2018). The treatment units were constructed within the Civil Engineering Department of Ladoke Akintola University of Technology, Ogbomoso, Oyo State Nigeria. Effluent wastewater collected from Sofak pharmaceutical Industry Aroje, Ogbomosho, Oyo State was fed to each wetland unit from elevated reservoir and circulated to the wetland units via pipes networks.
The plants used in this investigation (Figure 1) i.e. Wild Sorghum *(Sorghum arudinaceum),* White Lotus (*Nymphaea lotus*), Yerba De Jicotea (*Ludwigia erecta*), and Ginger-leaf morning-glory (*Ipomoea asarifolia*) were homogeneous in their weight and height. They were initially cultivated in tap water for a month to allow development of extensive root system after which they were transplanted into the constructed wetlands fed with the wastewater samples.
Figure 2 shows the wetland units with the vegetation growth after two months of study and four replicates were maintained per treatment. All experiments were set up at ambient temperature and light.

B. Effluent Sampling

Collection of water samples were done at 7, 14, 21, 28 days of transplanting the macrophytes and this was followed by analysis for determination of the various physicochemical parameters in wastewater including pH, Nitrate, Phosphate, Turbidity and Biochemical oxygen demand (BOD) which were determined using standard methods of the American Public Health Association (APHA, 1995; Dahunsi et al.2012). Determination of others (Calcium, Magnesium, Potassium, and Iron and heavy metals) was done with the aid of the atomic absorption spectrophotometer, and BOD as described earlier (Dahunsi et al., 2014; Ayandiran and Dahunsi, 2017).

C. Statistical analysis

Mean value and standard error were calculated for all analyzed parameters.

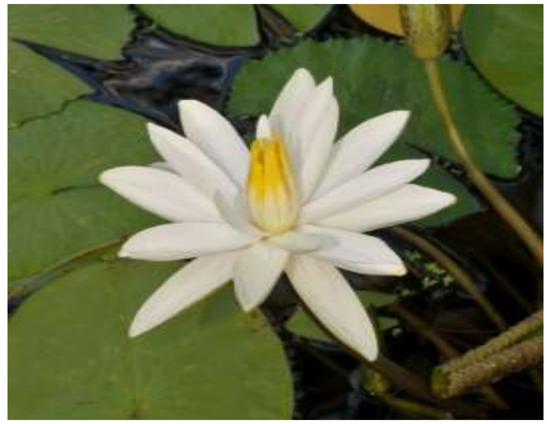
3. RESULTS AND DISCUSSION

A. Characteristics of Run-off before phytoremediation

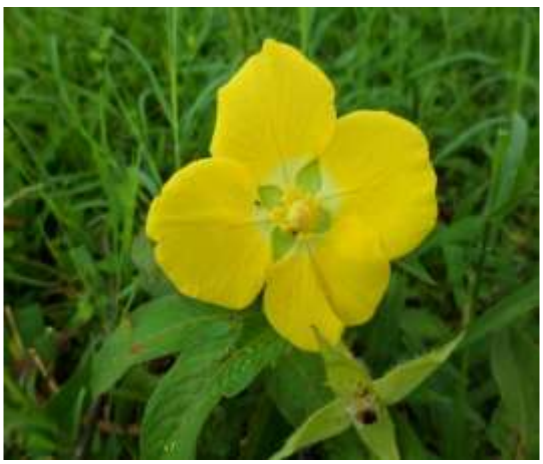
As shown in table 1, the pharmaceutical wastewater used in this study is of neutral pH with value of 7.2 and a high turbidity value of 13.5 NTU. The organic loading of the wastewater was quantified by the concentration of Nitrate and Biochemical Oxygen Demand (BOD) which gives the values of 4.05 and 185 mg/L respectively. The water if further heavily contaminated with heavy metal with the values 0.02 mg/L for Manganese, 5.33 mg/L for Iron, 0.63 mg/L for Lead while Phosphate recorded a concentration of 0.17 mg/L. For the cations, the value recorded for Magnesium, Sodium and Potassium are 16.85 mg/L, 20.1 mg/L and 8.73 mg/L respectively while Calcium recorded the highest concentration of 332.5 mg/L.

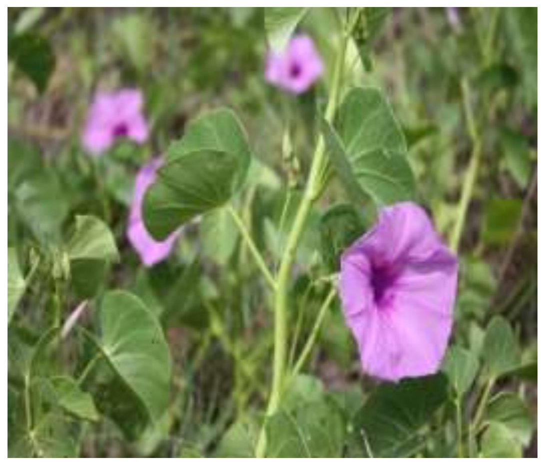
B. Physicochemical Characteristics of Pharmaceutical Wastewater treated via phytoremediation

Table 1 shows the physicochemical characteristics of pharmaceutical wastewater at the inlet and mean composition after phyto- treatment at the outlet of the pilot unit over the 28-day retention time in each case. Decrease in the values of all parameters, was observed, after remediation with *Sorghum arudinaceum,* having up to 71.4 percent removal of contaminants. The same trend was noticed with *Ipomoea asarifolia,* where up to 80.6 percent remediation was recorded, except for nickel, ammonium, total copper, BOD and COD, with increased values after remediation. There was also increase in values of all parameters after phyto-treatment with *Nymphaea lotus,* except for turbidity, aluminium, calcium and phosphate, where reduced values of parameters were recorded. However, only about sixty-five percent of parameters were reduced in values, after treatment with *Ludwigia erecta,* with the highest value of 65.4 percent removal of BOD.



(a) Wild Sorghum *(Sorghum arudinaceum)* **(b) White Lotus (*Nymphaea lotus*)**





(c) Yerba De Jicotea (*Ludwigia erecta*) **(d) Ipomoea Asarifolia** (ginger-leaf morning-glory)

Figure 1 : The wetland plants (a-d) used in this present investigation

Figure 2: Constructed Wetland System setup with vegetation growth after two months of study

Table 1: Pharmaceutical wastewater quality at the inlet and mean composition after phyto- treatment at the outlet of the pilot units (n = 4).

		Wild Sorghum		White Lotus		Ludwigia		Ginger Morning Glory	
Parameters	wastewater (inlet)	Mean [a] (outlet)	% of removal [b]	Mean (outlet)	% of removal	Mean (outlet)	% of removal	Mean (outlet)	% of removal
Ph	7.35	7.00 ± 0.1	5.3	7.54 ± 0.1	- [c]	7.25 ± 0.2	1.4	7.24 ± 0.3	1.5
Turbidity	160	57.50 ± 29.2	64.1	136.50 ± 86.3	14.7	63.75 ± 35.3	60.2	72.50 ± 39.2	54.7
D.O	0.94	0.75 ± 0.4	22.2	1.19 ± 0.2	-	0.84 ± 0.3	10.6	0.84 ± 0.3	10.6
Total Hardness	140	40.00 ± 57.07	71.4	221.00 ± 52.29	-	65.00 ± 47.76	53.6	55.00 ± 69.8	60.7
Aluminium	0.26	0.10 ± 0.03	61.5	0.24 ± 0.2	7.7	0.19 ± 0.1	26.9	0.18 ± 0.1	30.8
Iron	3.10	1.57 ± 0.4	49.4	3.80 ± 1.3	-	2.59 ± 1.1	16.5	2.18 ± 1.2	29.7
Magnessium	32.00	14.00 ± 4.2	68.8	50.25 ± 15.8	-	31.75 ± 10.0	0.8	26.25 ± 13.5	18.0
Calcium	90.00	0.0000 ± 0.0	100.0	35.25 ± 16.1	60.8	36.00 ± 32.6	60.0	17.50 ± 22.1	80.6
Chloride	1.60	0.93 ± 0.1	42.0	2.25 ± 0.6	-	1.67 ± 0.4	-	1.41 ± 0.6	11.9
Sulphate	55.00	36.50 ± 5.0	33.6	68.75 ± 12.0	-	53.00 ± 12.5	3.6	47.00 ± 14.4	14.5
Nitrate	1.60	0.98 ± 0.1	38.8	2.08 ± 0.7	-	1.88 ± 0.2	-	1.49 ± 0.6	6.9
Phosphate	84.0	66.78 ± 12.4	20.5	34.35 ± 40.1	37.0	35.58 ± 41.6	57.6	39.60 ± 26.5	53.0
Potassium	3.5	2.75 ± 0.3	21.4	4.30 ± 0.5	-	3.83 ± 0.2	-	3.33 ± 0.5	5.0
Nickel	8.9	5.50 ± 1.2	38.2	16.98 ± 2.8	-	12.40 ± 1.4	-	9.13 ± 4.0	-
Zinc	12.0	6.03 ± 2.2	49.8	21.63 ± 3.1	-	14.41 ± 8.2	-	7.95 ± 1.4	33.8
Ammonium	0.27	0.07 ± 0.1	74.1	0.43 ± 0.16	-	0.30 ± 0.1	-	0.28 ± 0.2	-
Tot.Copper	1.75	0.98 ± 0.3	44.0	3.30 ± 0.4	-	2.31 ± 0.6	-	1.89 ± 0.7	-
BOD	0.26	0.12 ± 0.2	53.8	0.66 ± 0.3	-	0.09 ± 0.3	65.4	0.53 ± 0.3	-
COD	0.4	0.19 ± 0.3	52.5	1.07 ± 0.5	-	0.14 ± 0.5	65.0	0.83 ± 0.5	-
Oil and Grease	5465	5250 ± 1133.6	3.9	7302 ± 1480.8	-	5125 ± 696.0	6.2	5013 ± 1004.4	8.3

a: Mean ± standard deviation; b: % of removal = [(averaged inlet concentration - averaged outlet concentration)/averaged inlet concentration].100%; c: Not available.

In all the experiments, alkaline pH was maintained throughout. The highest removal efficiency for turbidity (64.1%) at the end of the phytoremediation period was achieved in experiment with *Sorghum arudinaceum,* followed by *Ludwigia erecta*, (60.2%), while the least removal effeciency (14.7%) was noticed with *Nymphaea lotus*. Calcium was removed with the highest efficiency (100%) using *Sorghum arundinaceum* for remediation while phyto-remedial reductions in parameter values were also recorded in other experiments. Nickel, ammonium, copper were not removed by three other plants except with *Sorghum arundinaceum*. Also *Nymphaea lotus* and *Ipomoea asarifolia* could not remove BOD and COD. Oil and grease were not removed at all by *Nymphaea lotus*, but inefficiently removed in other experiments: *Sorghum arundinaceum* (3.0%), *Ludwigia erecta* (6.2%) and *Ipomoea asarifolia* (8.3%). *Nymphaea lotus* and *Ludwigia erecta* could not remove chloride and nitrate. These contaminants were poorly removed by *Ipomoea asarifolia* (11.9 and 6.9%), but ineffectively, removed by *Sorghum arundinaceum* (42 and 38.8%) respectively.

There was removal of total hardness, iron, manganese and sulphate in three experiments, except with *Nymphaea lotus*, with highest removal percentage values recorded in *Sorghum arundinaceum*. The results of the usage of the four macrophytes revealed that *Sorghum arundinaceum* possessed the highest overall efficiency in the remediation of the parameters, except in phosphate, BOD and COD where *Ludwigia erecta* was more efficient, while *Ipomoea asarifolia* displayed highest removal capacity majorly, for oil and grease.
This result agrees with earlier submissions (Vymazal, 2007; Schwitzguébel et al. 2009; Vymazal, 2014; Ijaz et al. 2015; Lynch et al. 2015; Srivastava et al. 2016; Ng et al. 2017;).

4. CONCLUSION

The result obtained in this study showed that *Sorghum arundinaceum, Ludwigia erecta* and *Ipomoea asarifolia* macrophytes are capable in remediating wastewater from pharmaceutical industry. Phyto-treatment method proves to help stabilize wastewater from pharmaceutical industry and other wastewaters before being released to the environment, thus mitigating occurrence of eutrophication. After the phytoremediation experiments, the treated Pharmaceutical wastewater quality was improved such that it is suitable for discharge into the environment without risks to ecosystem. However, further studies were recommended to evaluate the full potential of the macrophytes in remediation using wastewaters from different sources.

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