Journal of Agriculture and Agricultural Technology

(JAAT)

Journal of Agriculture and Agricultural Technology 14 (2), 2025

PHYTOREMEDIATION OF HEAVY METALS POLLUTED WASTEWATER USING HYBRID CONSTRUCTED WETLANDS

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ABSTRACT

A hybrid Horizontal Subsurface flow (HSSF) and vertical subsurface flow VSSF) constructed wetlands (CWs) can be used to treat heavy metals polluted wastewater. This study evaluated the heavy metals removal efficiency of hybrid CWs using plant species in horizontal subsurface flow (HSSF) and vertical subsurface flow (VSSF)constructed wetlands. . The Hybrid CWs had an average removal efficiency of 86 % copper, 73% for lead 70% for nickel, 68% for cadmium, 79% for zinc and 68% for arsenic. The CWs planted with Typha latifolia further reduced the contaminant load giving an additional removal efficiency of 74, 65, 43, 65, 58, 50 and 75% for, respectively, copper, lead, cadmium, nickel, zinc and arsenic. The combined hybrid CW showed, therefore, an improved effluent quality with overall removal efficiencies of, respectively, 94%. Typha latifolia thrived well in the VSSF and HSSF CWs, which may have contributed to the high contaminant removal efficiency compared to other CWs planted with other macrophytes. Typha latifolia is thus a suitable plant species for treatment of heavy metal polluted wastewater. It is recommended that two or more of these macrophytes be combined in a hybrid CWs to evaluate *how viable it will be for the treatment of heavy metals polluted wastewater under the prevailing arid climatic conditions.*

Keywords: heavy metals, macrophytes, phytoremediation and transfer factor.

INTRODUCTION

The scarcity of irrigation water is a growing problem with natural water resources becoming inadequate to fulfill demand (Githuku *et al*., 2018). This problem especially in developing world is related to the quantity and quality of irrigation water (Mustapha *et al*., 2015). According to Prabhat (2013) the number of developing countries facing water scarcity during the last four decades, is expected to increase to 34% by the year 2030. In recent times, the development in technology and industries has led to more production and the wastes produced by these industries causes numerous problems due to unprocessed industrial effluents into fresh water system, which are mainly sources of irrigation water (Shafaqat *et al*., 2020) waste water from several industries such as electroplating, pharmaceutical tannery, mining and steel contains elevated levels of heavy metals when discharged directly constitute great risk not only to the aquatic ecosystem but also to soil and plant when used for irrigating. These metals ingested through water or food has been widely reported as one of the major causes of cancer, cardiovascular disease and death (Rizzo et *al.,* 2020), the toxins are also capable of causing serious damage even at low concentration (Síma *et al*., 2019).

Due to the aforementioned health related problems associated with the metal, a number of methods have been reported for their removal. The currently used method for removal of heavy metals includes solvent extraction, ion exchange, membrane separation, reverse osmosis, chemical precipitation and electrodialysis. (Nazir *et al*., 2020). Aside the health challenges, increasing environmental awareness coupled with cost efficiency has triggered researchers to finding appropriate wastewater treatment technologies (Khan *et al.,* 2019). Due to cost implication of using the conventional wastewater treatment methods, coupled with the need for further treatment of resultant sludge before disposal, the use of plant remediation as a preferred alternative is becoming more popular in recent time, (Mustapha *et al*., 2018). This system is relatively cost effective, energy sensitive and self-sustaining for wastewater treatment (Cristaldi *et al.,* 2021). In this treatment, microscopic aquatic animals have been utilized by some, while in some others, aquatic plants (hydrophytes) are involved.

Phytoremediation is the use of plants to clean up or control many kinds of pollutants including metals, pesticides, and oil. Wetland plant species are capable of moving many contaminants from the wastewater (Mustapha *et al.,* 2015). In addition, wetland plant roots provide a habitat that is conducive for the growth of a great diversity of microbial communities which enhance the pollutant removal efficiency in Constructed Wetlands (CWs) (Atta *et al.,* 2022). These green plants in construction wetland provide substrate (roots, stems and leaves) upon which microorganisms can grow as they break down organic and inorganic materials from the environment or render them innocuous (Jeevanantham *et al.,* 2019). It provides an alternative to more aggressive and intrusive conventional form of remediation. It is less expensive, safe, environmentally friendly and green technology for industrial wastewater treatment. A number of aquatic and terrestrial plants including grasses, herbs, shrubs and trees have been discovered to have high tolerant for water toxicity stress and possess an excellent ability for significant extraction of non-metabolic elements from contaminated environments (Alsghayer *et al.,* 2020). *Typha latifolia, Phragmites australis, Cypperus alternifolius and Cynodon dactylon,* have been identified in the last two decades as highly effective in absorbing and accumulating various toxic trace elements and are being evaluated for their role in this research.

The present trend of population growth and the need for more agricultural food production to cater for the increasing population has led to intensive farming activities globally (Arjus *et al*., 2022). For instance, in order to satisfy the world's food demands, a lot of irrigation farming have been established in most parts of the world, Nigeria inclusive. In recent years, the amount of wastewater produced from several industrial activities has increased as a result of the improvement in technology, unplanned industrialization, urbanization practices and agricultural by- products. However, farmers who want to take advantage of the evolution are confronted with contaminants when dealing with wastewater to irrigate, which is generated in huge amount and contains wide varieties of bacteria, parasites and other materials which makes its direct reuse harmful. Reuse will have negative impact on population and environment because of the presence of pathogens, degradable and non-degradable materials which can lead to gastro intestinal diseases. There is therefore the need to treat the wastewater to acceptable standards before it can be released for irrigation. Since most of the conventional wastewater treatment systems are very intensive and expensive. Effective but simple, cheap, and reliable wastewater treatment alternative like the CWs need to be employed in the treatment of wastewater. This will adequately treat contaminants and reduce heavy metal accumulation before reuse. The objectives of this research are therefore to develop a hybrid constructed wetland for treatment of the contaminant of concern in the discharged wastewater and to evaluate the performance of the constructed wetland in remediating heavy metals polluted wastewaters.

MATERIALS AND METHODS

Background of the Study Area

The Federal Capital Territory (FCT), location for this experiment, is situated in the central region of Nigeria and serves as the nation's capital. It encompasses the city of Abuja, along with surrounding areas, and is strategically located between latitudes 8.25° and 9.20° North and longitudes 6.45° and 7.39° East (Figure 1). This central positioning makes it accessible from various parts of the country according to Niger State Bureau of Statistics. The FCT covers an extensive land area of approximately 7,315 square kilometers. According to the National Population Commission (NPC), the area includes diverse landscapes, ranging from urban developed to peri-urban and rural zones. The territory is characterized by various land uses, including residential, commercial, industrial, agricultural, and natural reserves.

The climate in the FCT is classified as tropical savanna, featuring distinct wet and dry seasons. The rainy season extends from April to October, with the peak rainfall occurring between July and September. During this period, the FCT receives an average annual rainfall ranging from 1,000 to 1,500 mm (Mustapha *et al.,* 2015). This substantial rainfall is crucial for replenishing local water bodies, supporting agriculture, and maintaining the ecological balance of the region. Due to high level of municipal and industrial activities, FCT generates significant quantity of wastewater. The wastewater generated can be categorized into domestic, commercial, and industrial wastewater. Domestic wastewater is primarily derived from households and residential areas, while commercial wastewater comes from business establishments, offices, and service providers. Industrial wastewater, on the other hand, is produced by various manufacturing and processing industries located within the territory. These industries include food and beverage production industries, pharmaceuticals, textiles, chemicals, and construction industries. The volume of industrial wastewater generated in the FCT varies depending on the industrial activities and their scale of operations. On average, the FCT produces several thousand cubic meters of industrial wastewater daily. This wastewater contains a mixture of organic and inorganic pollutants, necessitating effective treatment and management to prevent environmental contamination and health risks.

Collection of wastewater from different locations

Sample of wastewater were collected weekly from six locations; tannery wastewater, abattoir, textile, electroplating, battery production and pharmaceutical industrial effluents. prior to discharge into the environment and mixed together to form composite wastewater. At the collection points, jerricans were thoroughly rinsed three times with that particular wastewater before samples were fetched. Composite of the six different wastewaters collected was formed by mixing equal volume in a separate container. From each sample, small sampling containers were first used to take a sample for analysis to ascertain the concentrations of heavy metals present in the wastewater samples. before the compositing and introducing into the hybrid constructed wetland.

Characterization of individual and composite wastewater

The heavy metal content; copper, zinc, lead, cadmium, chromium, nickel and arsenic. in the collected wastewater were investigated. These were determined using Atomic Absorption Spectrophotometer (AAS) according to the procedures described in the standard methods for the examination of water and wastewater (APHA, 2002).

Design of the constructed wetland.

The vertical subsurface flow (VSSF) wetland and horizontal subsurface flow (HSSF) wetland were constructed in this research based on certain design criteria as described by Prabhat *et al.* (2013). The entire set-up of the hybrid system has a total of 30 cells, which was determined using random but rotational design (15 vertical cells and 15 horizontal cells), and a replicate for validation. The VSSF were cylindrical shaped aluminum containers with dimensions 29 cm and 70cm as diameter and height respectively, while the HSSF were cuboid shaped polyvinyl chloride (PVC) containers of dimensions 50 cm, 30 cm, 30 cm as length, width and depth respectively.

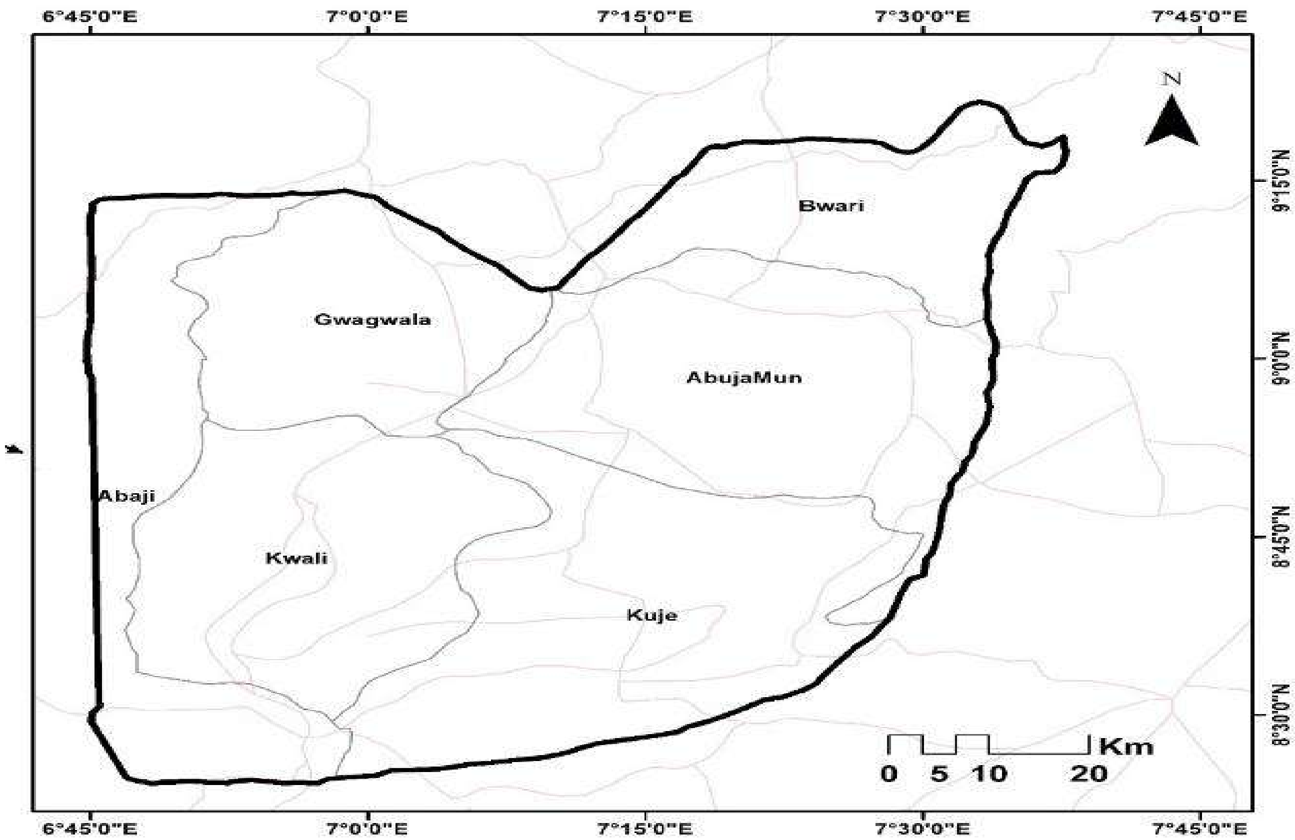


Figure 1: Aerial map of FCT

Determination of flowrate (F_r)

The flowrate into the wetland was determined as liters per day, considering of the Darcy's law. The tap heads were carefully regulated with a flow meter and flow monitored and tightly locked immediately it was 24 hours. Quantity of water collected was measured and value noted and recorded. The volume determined using this Darcy's method was found to be 9l/day.

Determination for volume of the constructed wetland (VSSF)

The wetland is cylindrical, and so the volume was determined using equation1:

$$V_{vssf} = \pi r^2 * h \qquad 1$$

Where;

R is radius of circular surface

H is the height of the cylindrical shape

Equation 1 can be re-written as:

$$V_{vssf} = \frac{\pi D^2}{4} * h \quad 2$$

Where, D is the diameter of the circular surface.

Determination for volume of the constructed wetland (HSSF)

The wetland is in the shape of a cuboid, and so the volume was determined as follows:

$$V_{hssf} = L_c * B_c * H_c \quad 3$$

Where;

$L_c$is length of cuboid shape, B_c is the breadth of cuboid shape and H_c is the height of cuboid shape.

Determination of the hydraulic retention time (HRT)

Hydraulic retention time is the time taken for the influent fed to undergo treatment in the cell by the activities of its component. It was determined as follows:

$$HRT = \frac{V_{wetland}}{F_r} \quad 4$$

Where;

$V_{wetland}$ is the volume of wetland, and F_r is the flowrate.

Determination of the hydraulic loading rate (HLR)

This is the ratio of the flowrate to the surface area of the wetland. In this research, it was determined as follows:

$$HLR = \frac{F_r}{A_s} \quad 5$$

Where;

F_r is flowrate, and As is surface area of wetland.

Table 1 presents the summary of the design factors considered for the hybrid constructed wetland. Each vertical cell has an effective volume of 43 litres with porosity of 0.40, flow rate of 9 l/day, a hydraulic load rate of 14 l/day and a hydraulic retention time of 1.9 days. While, each horizontal cell has an effective volume of 45 liters with a porosity of 0.40, flow rate of 9 l/day, a hydraulic loading rate of 6 l/day, and a hydraulic retention time of 2 days.

Experimental set-up of the hybrid wetland

Vertical subsurface flow

The cylindrical shaped component of the wetland was made with Aluminum sheets that measured 70 cm and 29 cm, length and diameter respectively, placed on 50 cm platform that was made of a 2 by 2 cm hollow pipes. This was so in consideration of the rooting depth of the macrophytes as stressed by Mustapha (2016). An opening was made 10 cm from both top and bottom to allow for inflow and outflow of wastewater in the wetland. The wastewater (influent) enters from the top, flows through the cell and flows out as (effluent) from the bottom. At the bottom, a flexible perforated pipe was fitted to avoid blockage and to allow water sink through into the horizontal section of the setup. The vertical wetlands were then connected in series to each other and to the wastewater reservoir tank, which is a PVC tank of 1,000 litres capacity installed to about 2 m above the ground. the experimental set up is as presented in

Figures 2 and 3.

Horizontal subsurface flow

The horizontal component of the hybrid wetland (HSF), which measured 50 by 30 by 30 cm were bored on the top part at one end, and at the opposite lower end, then placed on a 25 cm height platform. The two ends of the HSF were fitted with plumbing materials and fitted with control valves and other PVC fitting materials, then connected in series. These two containers were then connected laterally using flexible PVC pipes and control valves to regulate the inflow into the vertical wetland and outflow through the horizontal wetland to the discharge point.

Table 1: Summary of the Design Factors Considered for the Hybrid Wetland

S/No	Design factors	Wetland - A	Wetland – B
1	Nature of flow	Vertical substrate flow (VSSF)	Horizontal substrate flow (HSSF)
2	Number of hybrid wetland	15 including control with one replicate for each	15 including control with one replicate for each
3	Shape	Cylindrical	Cuboid
4	Dimension of wetland	29 cm by 70 cm	50 cm by 30 cm by 30 cm
5	Volume	43 L	45 L
6	Substrate and its	Gravel:	Gravel:
7	Loading method	Continuous	Continuous
8	Flowrate	9 l/day	9 l/day
9	Hydraulic retention time	1.9 day (approximately 2 days)	2 days
10	Hydraulic loading rate	14 L/day	6 L/day
11	Porosity	0.40	0.40

Figure 2: The Experimental Setup

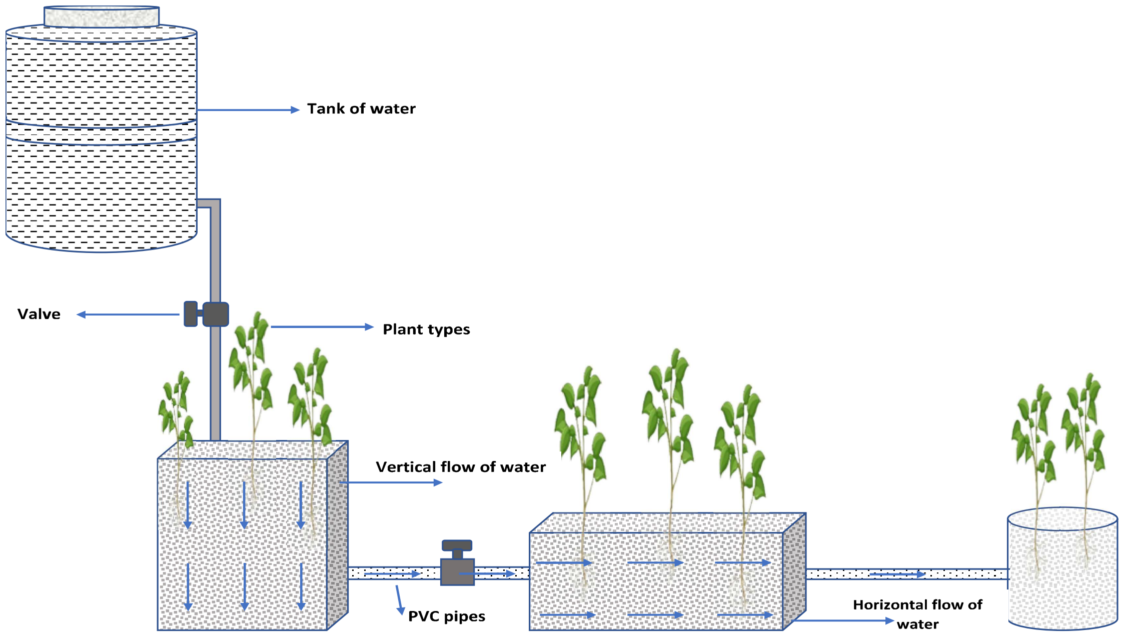


Figure 3: Graphical Representation of the Experimental Setup

Substrate arrangement in the wetland

Substrate media of different size were collected and arranged in the vertical (VSF) and horizontal (HSF) wetlands. In the VSF compartments, the largest sizes of the media (6 mm - 10 mm) were poured into the wetland to about 20 cm depth from the bottom, followed by another 20cm depth gravel of (4mm – 6mm), followed by another 20cm depth of sand particles making up to 60 cm in total depth. The 10 cm at the top was left empty to make for air circulation. In the HSF compartments also, largest sizes of the substrate media (6mm - 10 mm) were poured at both ends of the wetland making up to 20 cm at both ends, at the next two compartments from the both sides were followed by the next size of the media (4mm – 10mm), followed by the sandy particles at the middle (the exact point which the macrophytes were planted). These compartments were held in place using tile sheets and removed at the end of the construction

Arrangement and establishment of the macrophytes

The hybrid wetlands were labeled 1 - 15 are representing the different random but rotational combinations of the macrophytes. Macrophytes with different orientations were established in fourteen of the hybrid wetlands. The macrophytes used were *Typha latifolia, Phragmites australis, Cyperus alternifolia and Cynodon dactylon.*

Operation of the hybrid wetland

Wastewater was pumped up into the 1000 liters capacity over-head wastewater storage tank through pipes using a 4.5 kW electric centrifugal pump, which subsequently flow gradually by gravity into the wetland cells. This configuration of the Hybrid constructed wetland system is such that the influent (composite wastewater) from the wastewater storage tank, flows at a designed flowrate into the VSSF CWS, whereas the effluent from the VSSF CWS is referred to as the HSSF CWS influent and the effluent from the HSSF CWS is referred to as the final effluent. Treated samples from the outlet of the wetland cells were collected. The final effluent, macrophytes, substrate from the hybrid constructed wetlands and soil are taken to the laboratory for analysis.

Macrophyte sampling and analysis.

The macrophyte were collected from the hybrid wetland for analysis to determine the concentration of heavy metals in them using standard methods. the different macrophytes where cut from the hybrid containers and labeled accordingly as 1,2, 3,14, and the 15^{th}.

Treated wastewater sampling and analysis.

Treated wastewater were collected from the hybrid constructed wetland at an interval of four weeks, starting from Jan, 2024.

Determination of heavy metal in water sample.

The water sample are already in the form that heavy metal can easily be detected by analytical instrument, so the sample were transferred into sample bottle and analyzed using ICE 3000 AA021341104 V1.30 Thermo Scientific Atomic Absorption Spectrophotometer (AAS). The concentration is in mg/l

RESULTS AND DISCUSSIONS

The initial concentrations of heavy metals of concern in the wastewater samples are as presented in Table 2. This clearly showed that the concentrations of the heavy metals if compared with Mustapha *et al.,* (2015) is at elevated level which will make their usage for irrigation be hazardous to the consumers. Therefore, it has to be subjected to some forms of treatment before reuse.

Table 2: Initial Concentrations of the Heavy Metals in wastewaters for six Months Sampling Period

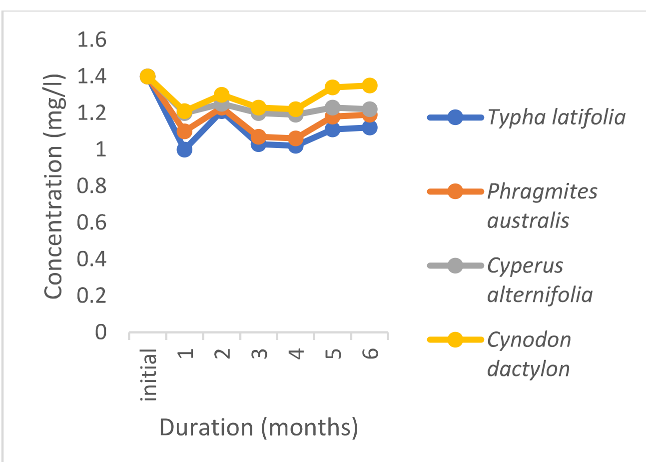
Month	Copper (mg/l)	Chromium (mg/l)	Cadmium (mg/l)	Nickel (mg/l)	Lead (mg/l)	Zinc (mg/l)	Arsenic (mg/l)
1	1.3986	3.4681	5.0581	0.2648	4.7145	0.2216	0.5163
2	1.4101	0.1718	4.1751	0.3121	3.7112	0.3121	0.5163
3	1.4017	0.1923	3.0171	0.3071	3.3142	0.4372	0.3231
4	1.2731	0.2781	3.1013	0.1211	3.0001	0.6553	0.4555
5	1.2132	1.1025	3.7300	0.3101	3.1741	0.6111	0.5163
6	1.2013	0.2107	3.1210	0.2134	4.1013	0.5910	0.4555

The results of heavy metals concentration presence in the macrophytes are as presented in Table 3. Since the plants are not edible, literatures did not quote the permissible limit which the plants can withstand. As a results, the values were just recorded and formed a baseline used to evaluate the translocations of these heavy metals from wastewater treated into the tissues of the four macrophytes under evaluation.

Figures 4a to 8a presented the value of heavy metals reduction from wastewater which the four plants species were able to perform. The buildup of the absorbed different heavy metals was also presented in the Figures 4b to 8b. It can be seen in the figures that *Typha Latifolia* has ability to absorb more heavy metals into its tissues than other macrophytes used. The results also showed that all the macrophytes absorbed the heavy metals into their systems in the order of leaf > root > stem. This study also revealed that VSF CWs planted with *Typha Latifolia* have a high potential to reduce more of chromium with the systems achieving a removal efficiency of 70 - 80 % through (phytostimulation or rhizodegradation), suggesting the positive effect of the presence of plants on CWs performance (Shafaqat *et al.,* 2020). It was also observed that the removal rates of copper were higher than those of Nickel and cadmium suggesting that copper was easier to degrade than other heavy metals (Alsghayer *et al.,* 2020). The factors that can influence the rate of heavy metals degradation are availability of microorganisms, adequate concentrations of nutrients, oxygen and a pH between 6 and 9 (Jeevanantham *et al.,* 2019). The degradation of heavy metals in the root zone occurs through plant stimulation of microbial activity, by direct uptake by the plants as well as by phytodegradation and volatilization (Khan *et al*.,2019). These macrophytes have the ability to stimulate the growth of hydrocarbon-degrading microorganisms in the rhizosphere by releasing root exudates and sloughed-off cells (Cristaldi *et al.,* 2021), thus accelerating the degradation of heavy metals in the waste waters.

Table 3: initial metal concentration in the macrophytes

Species	Unit	Copper (Cu)	Chromium (Cr)	Cadmium (Cd)	Nickel (Ni)	Lead (Pb)	Zinc (Zn)	Arsenic (As)
Typha latifolia	mg/kg	0.1974	0.0330	0.0190	0.0517	-0.2082	-0.1599	0.0000
Phragmites australis	mg/kg	0.4720	0.0505	0.0183	0.0517	-0.1784	-0.0417	0.0000
Cyperus alternifolia	mg/kg	0.2421	0.0542	0.0061	0.0760	-0.2970	-0.1250	0.0000
Cynodon dactylon	mg/kg	0.3107	0.0417	0.0138	0.1007	-0.1793	-0.1176	0.0000



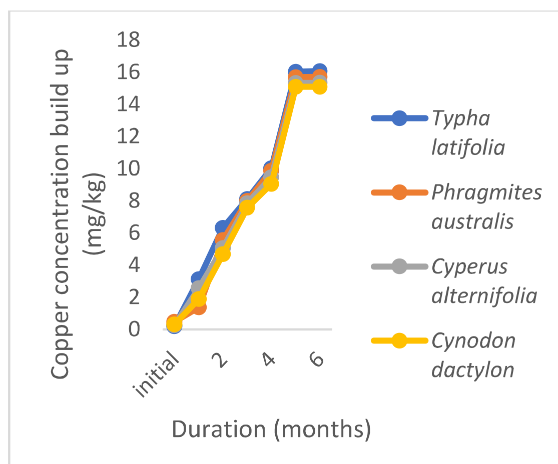
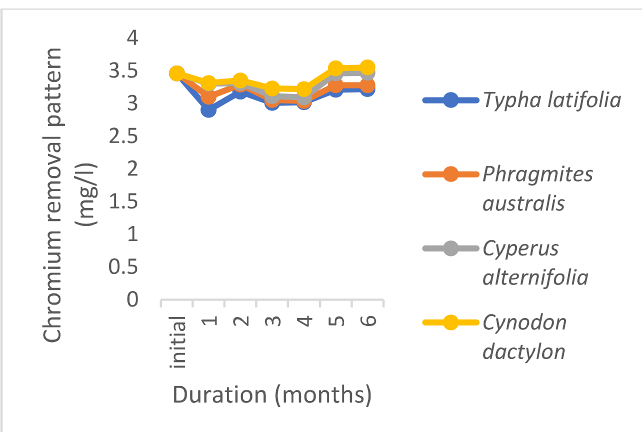


Fig. 4a: Six months treatments for copper accumulation in Macrophytes

Fig, 4b: Six months of copper

The mechanisms for heavy metals removal can be attributed to microbial degradation, sorption, plant uptake and volatilization (Nazir *et al*., 2020). *Cyperus alternifolia* and *Cynodon dactylon* were able to accumulate more of zinc and arsenic into their roots and stems. Over five times more of these contaminants accumulated into their parts compared with the initial concentrations at the start of the experiment. A plant may be considered as a hyperaccumulator if it has a bioaccumulation factor (BAF) greater than 1 (Mustapha *et al.,* 2018). In the present study, *Cyperus alternifolia* and *Cynodon dactylon* have a BAF < 1 for cadmium and chromium, indicative of low uptake of hydrophilic compounds of these two heavy metals (Shafaqat *et al.,* 2020), but a BAF > 1 for zinc and arsenic indicating their ability to uptake them from CWs through its roots. Higher values of BAF for zinc and arsenic suggest that their remediation was by phytosabilization mechanism (Shahabaldin *et al.,* 2015).



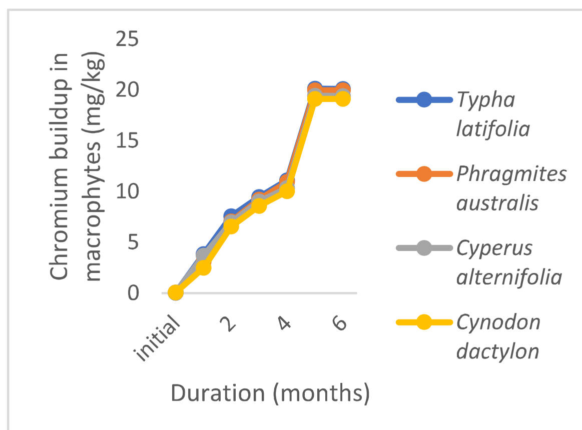
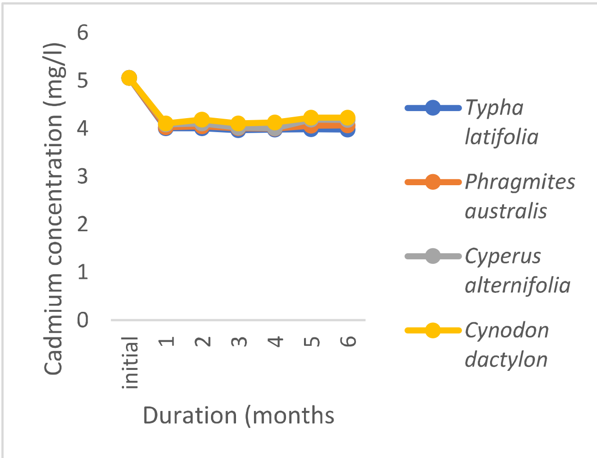


Fig. 5a: Six months treatments for Chromium

Fig. 5b: Six months accumulation of Chromium in Macrophytes

It also suggests that the two macrophytes, zinc and arsenic are hyperaccumulator as proposed by Khan *et al.,* (2019). The BAF values of *Phragmite australis* for lead and copper were observed in the order of root > leaf > stem suggesting phytoremediation potential of *Phragmite australis* through phytostabilization in root and phytodegradation in leaf samples (Arjus *et al.,* 2022). *Phragmite australis* had a good translocation ability for lead and copper, since the transfer factor (TF) > 1 (Githuku *et al.,* 2018).



Fig, 6a: Six months treatments for Cadmium

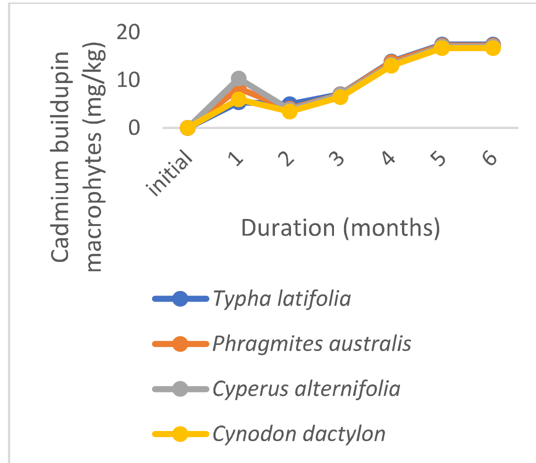


Fig.6b: Six months accumulation of Cadmium in Macrophytes

Treatment of heavy metals polluted wastewater with constructed wetlands is thus a good system when plant species for macrophyte are carefully selected because of its high productivity, high tolerance, high rate of contaminant accumulation, nutrient assimilation and support to the proliferation of soil microorganisms in the root zone (Mustapha *et al.,* 2018).

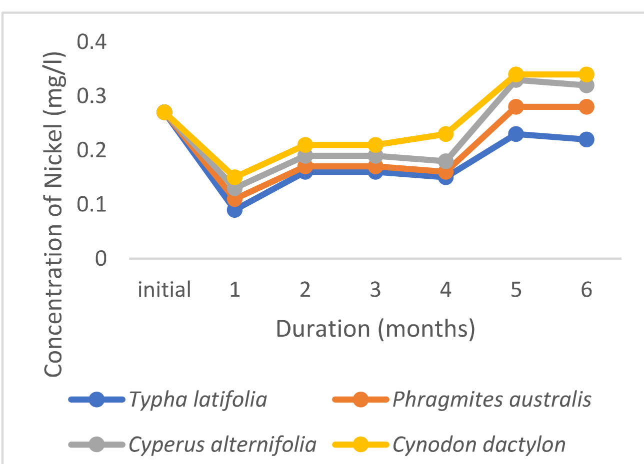


Figure 7a: Six months treatments for Nickel

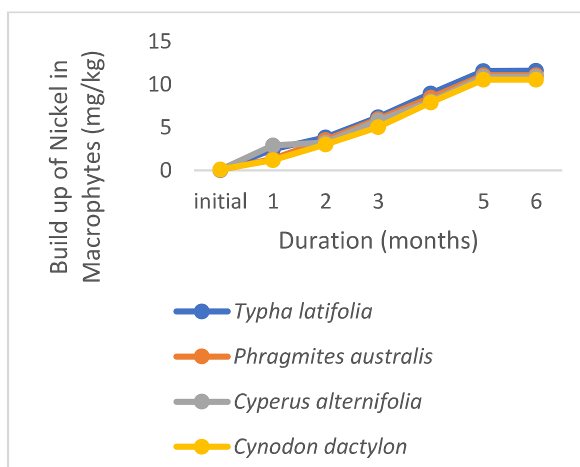
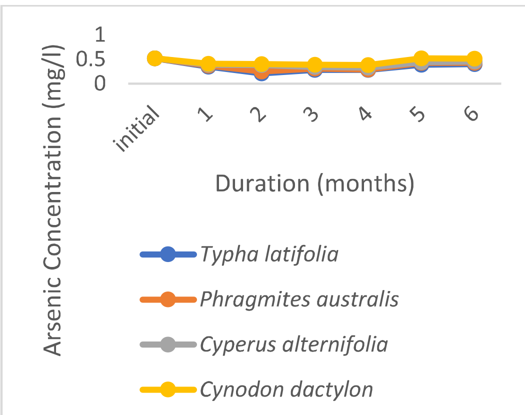


Figure 7b: Six months accumulation of Nickel in Macrophytes

The mobility of contaminants from the wastewater to the plant root (TF > 1) was higher than the mobility within the plant tissues. All the contaminants were partly transferred from the sediment to the roots of *Cyperus alternifolia*, indicating that uptake and accumulation depended on the pollutant present and its concentration (Alsghayer *et al*., 2020). It was observed that translocation within the plant tissues depended on the contaminant as well as on the tissue part.



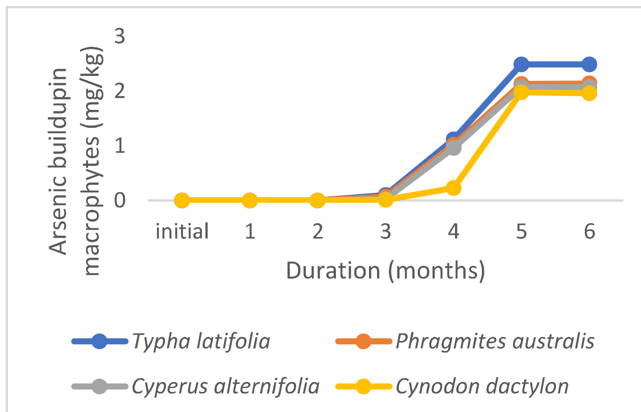


Figure 8a: Six months treatments for Arsenic

Figure 8b: Six months accumulation of Arsenic in Macrophytes

For instance, the transfer of cadmium within the *Cyperus alternifolia* tissue was higher from root to stem and from root to leaf than from stem to leaf, while copper and zinc showed higher translocation from stem to leaf. nickel was more translocated from the root of *Cynodon dactylon* to it leaf, while arsenic was more transferred from the stem to the leaf of *Typha latifonia*. The translocation of zinc from root to leaf (TF = 1.01) is due to mobility within the plant. Nickel showed the lowest translocation from root to stem and root to leaf, though it showed a higher translocation from root to stem than arsenic.

CONCLUSIONS AND RECOMMENDATIONS

A hybrid HSSF and VSF CWs planted with four macrophytes operated under tropical climatic conditions for over 150 days to examine the potential of CWs to remove heavy metals from industrial wastewater. The CWs showed effective removal of these metals (45 - 99 % copper; 79 - 80 % zinc and 70 - 80 % arsenic). *Typha latifolia* able to accumulate more of lead copper and arsenic, while *Cyperus alternifolia* and *Cynodon dactylon* absorbed more of Nickel cadmium and zinc into their plant tissue and showed a high translocation ability for the heavy metals. The variations in TF values showed that *Typha latifolia* is capable of removing heavy metals through its roots, leaves and stem. It is thus a good candidate plant species that can be used for further treatment of industrial wastewater and other types of wastewaters, especially for Nigeria or other developing countries that require low cost and low maintenance wastewater treatment systems.

ACKNOWLEDGEMENTS

The authors sincerely appreciate Nigerian Tertiary Education Trust Fund (TETFUND) who sponsored this research work through Institutional Based Research (IBRI) 2023 under the grant number TETFUND/FUTMINNA/2024/017 and the Department of Agricultural and Bioresources Engineering, Federal University of Technology, Minna where some laboratory analyses were carried out.

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