

Modelling Assisted Phytoremediation of Landfill Leachate using Surface Flow Constructed Wetland Enhanced by *Pistia stratiotes* and *Salvinia molesta*

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ABSTRACT

The current study is aiming to expose the efficiency of surface flow constructed wetland (CW) assisted by *Pistia stratiote* and *Salvinia molesta* in the remediation of landfill leachates. A laboratory-scale surface flow constructed wetland was constructed to imitate the characteristic of a natural pond. Composite sample of leachates was collected and transported to the laboratory for further analysis and studies. The removal efficiency of phenol, pesticides, sulphate, chloride, colour, turbidity, total suspended solid (TSS), total dissolved solid (TDS), biological oxygen demand (BOD), chemical oxygen demand (COD), ammonia nitrate and heavy metals (Pb, Cr, Cu, Cd, Ni, Hg). The removal of heavy metal ions in the CW was determined by using a phyto-system dynamic (phyto-SDA) model while the composite design (CCD) type of response surface methodology (RSM) was employed in this study for the optimization of pesticides and phenol removal from the landfill leachates by the constructed wetland (CW). The study also predicts that the deviation from the linearity between the heavy metals in the leachates and heavy metals in the sediment and in the *plant tissues* is influenced by the physicochemical status of the leachate and the mixed cultivation of *Pistia stratiote* and *Salvinia molesta*. The study reaffirms the role of sediments in the determination of the fate of heavy metals due to its crucial role in the bioavailability of heavy metals for uptake by *P. stratiotes* and *S. molesta* in a CW. The study also shows a positive effect of concentration and exposure time on the reduction efficiency of both pesticides and phenol. The result shows that exposure time and concentration of phenol and pesticides are useful in the optimization of the removal efficiency of pesticides and phenol.

Keywords: bioconcentration factor, phyto-system dynamic, macrophytes, constructed wetland

INTRODUCTION

Rapid development, population growth, rural-urban movement, wealth, and consumption rates have all contributed to a rise in the waste generation and pollution, which has harmed both man and the environment (Ferronato and Torretta 2019; Ugya 2015). When rainfall combines with the waste in a landfill, leachate is produced (He et al. 2005). The waste composition, water budget, biological, chemical, and physical conditions in the landfill body influence the quality leachate (Ehrig and Stegmann 2018). Leachate is a type of wastewater that comes in a wide range of quality and quantity. The rate of leachate formation

is determined by climatic variables (e.g., precipitation, evaporation) as well as landfill characteristics (e.g., infiltration, storage) (Ehrig and Stegmann 2018). It has a high percentage of organic materials (both biodegradable and non-biodegradable carbon), ammonia-nitrogen, heavy metals, and chlorinated organic and inorganic salts, which can cause significant environmental degradation if it gets into groundwater and surface water (Makhatova et al. 2020). Some of these contaminants, however, can be dissolved by microorganisms (Veiga et al. 2014; Taş et al. 2018), while others may not decompose and remain in the landfill for an extended amount of time (Wojciechowska 2013).

Leachate treatment has become a major challenge as a result of the large pollutant loads, and numerous treatments have been studied (Mojiri et al. 2020). Biological methods including aerobic bioreactors, anaerobic bioreactors, anammox, bioremediation, phytoremediation, nitrification, and denitrification processes have been employed in the treatment of leachates (Xu et al. 2010; Ugya 2021). The limitation of biological methods is in the removal of heavy metals and other non-biodegradable pollutants (Miao et al. 2019). The physical and chemical methods of landfill leachate treatment include adsorption, membrane, coagulation, flocculation, adsorption, and ion-exchange (AOPs) (Tatsi et al. 2003; Ugya et al. 2019b). However, none of these methods can be said to be the most efficient because landfill leachates tend to vary in composition, volume, and migration of pollutants (Szymańska-Pulikowska and Wdowczyk 2021). Several physical, chemical and biological treatment technologies have been combined to treat landfill leachate to improve removal efficiency and reduce energy usage (Xiang et al. 2019; Pan et al. 2019). This method, which includes AOPs + membrane, AOPs + coagulation, AOPs + adsorption, membrane + adsorption, nitrification + denitrification + anammox, biological + AOPs, biological + coagulation, biological + adsorption, biological + membrane, and constructed wetland, is considered an emerging method for the effective remediation of landfill leachate.

The current study employs the use of surface flow constructed wetland with the aim of removing both biodegradable and non-biodegradable pollutants in landfill leachates due to the complexity of the system to support ecological interaction. The introduction of *Pistia stratiotes* and *Salvinia molesta* into the surface flow constructed wetland is to assist in the removal of non-biodegradable via phytoextraction and also plays a crucial role in the phytodegradation of biodegradable pollutants. This is due to the fact that a surface flow wetland allows water to flow above ground, where it is exposed to the atmosphere and direct sunlight. Simultaneous physical, chemical, and biological processes filter sediments, decompose organics, and remove nutrients from the landfill leachates as water gently flows through the wetland (Hassan et al. 2021). Macrophytes are important biological components in constructed wetland that contribute to landfill leachate via direct and indirect methods by enhancing the

rhizosphere's environmental diversity and extraction of heavy metals. As a result, selecting macrophytes with acceptable survival and development rates in a given environment, as well as tolerance and efficient pollutant accumulation ability is a key factor in the phytoremediation method (Opitz et al. 2021; Ugya et al. 2019c). Although constructed wetland have been successfully utilized to treat wastewater in temperate nations, the experiences and design criteria used in temperate countries may not be appropriate in tropical countries (Guittonny-Philippe et al. 2014). Climate and other local factors influence leachates properties in the constructed wetland particularly microbiological processes that may be enhanced by high temperatures (Varma et al. 2021). There is a paucity of literature showing the use of surface flow constructed wetland in the remediation of landfill leachate (Arliyani et al. 2021; Ugya 2015). The available literature has not reported the use of *Pistia stratiotes* and *Salvinia molesta* despite the fact that both plants are good hyperaccumulators and can assist in increasing the efficiency of constructed wetland in the remediation of landfill leachates. The current study is aimed at accessing the efficiency of surface flow constructed wetland assisted by *Pistia stratiotes* and *Salvinia molesta* in the remediation of landfill leachates.

MATERIAL AND METHOD

Description of the surface flow constructed-wetland system

A laboratory-scale surface flow constructed wetland was constructed to imitate the characteristic of a natural pond. The wetland was constructed using a set of glasses of 50.0 cm length, a height of 50.0 cm, and 30.0 cm width with a glass thickness of 3 mm. The base of the wetland was filled with gravels of <20 mm size to the depth of 5.0 cm, followed by sand of <0.5 mm to the depth of 10.0 cm and then covered with gravels of <20 mm size.

Experimental setup

Composite sample of leachates was collected in 50 litres containers between 9 to 11 am. The samples were transported to the laboratory for further analysis and studies. The leachate was tested for parameters (phenol, pesticides, sulphate, chloride, colour, turbidity, TSS, TDS, BOD, COD,

ammonia nitrate and heavy metals (Pb, Cr, Cu, Cd, Ni, Hg)) before pouring the leachate into the constructed wetland. *P. stratiotes* and *S. molesta* was introduced into the wetland and the leachate was re-tested weekly for the period of five weeks. The reduction efficiency was thus determined using the formula below:

$$\frac{B - A}{A} \times \frac{100}{1} \quad (1)$$

where: A – initial concentration,
 B – final concentration.

Determination of Parameters

The parameters such as sulphate, chloride, colour, turbidity, TSS, TDS, BOD, COD, ammonia, nitrate were determined using the standard methods according to APHA, 2005. The section for TDS is 2540-gravimetric method, TSS is 2540-gravimetric method, chloride is 4500-Cl-calorimetric method, sulphate is 4500-SO₄²⁻ – gravimetric method, COD is 5220-colorimetric method, BOD is 5210-calorimetric method, nitrate is 4500-NO₃⁻-spectrophotometric method, ammonia is titrimetric method-4500-NH₃, turbidity was by nephelometric method-2130 B and colour is spectrophotometric method-2120 C. Phenol was determined by liquid-liquid extraction method- EPA method 625 while pesticide was determined using liquid-liquid extraction method- EPA method 1699. The concentrations of heavy metals (Pb, Cr, Cu, Cd, Ni, Hg) were determined by digesting the water sample and by using an Atomic Absorption Spectrophotometer (varian AA240FS, USA).

Bioaccumulation factor

The macrophytes were removed from the CW after 6 weeks and divided into the root and the shoot. The heavy metals (Pb, Cr, Cu, Cd, Ni, Hg) in the root and shoot were then determined separately. The concentration of the heavy metals in the root was thus used to determine the bioconcentration and biotranslocation factor using the formular below:

$$BCF = \frac{A}{B} \quad (2)$$

$$BTF = \frac{C}{A} \quad (3)$$

where: A – metal concentration in root,
 B – metal concentration in water,
 C – metal concentration in shoot.

Modelling and statistical analysis

The prediction of the heavy metal removal in the CW was determined by using a phyto-system dynamic (phyto-SDA) model. The test for correlation between heavy metals (Pb, Cr, Cu, Cd, Ni) in the leachates, sediment, and plant tissues was done using linear regression. The result of the correlation was assumed to be significant at $P < 0.05$ (Zhang et al. 2010). The composite design (CCD) type of response surface methodology (RSM) was employed in this study for the optimization of pesticides and phenol removal from the landfill leachates by the CW. The independent variables in this study are retention time (week), pesticide concentration, and phenol concentration. The efficacy of pesticides and phenol reduction indicates treatment response. The total number of experiments conducted to test the two factors is 10. The data were fitted into empirical second-order polynomial model in order to optimize the efficiency of pesticides and phenol removal in the leachates. The polynomial model used in the study is demonstrated in equation (4).

$$Y = b_0 + \sum_{i=1}^n b_i x_i + \sum_{i=1}^n b_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n b_{ij} x_i x_j \quad (4)$$

where: Y is the predicted response by the model (pesticide and phenol removal efficiency), b_0 is constant, b_i are linear coefficient, b_{ii} are interaction coefficients, x_i and x_j are the coded values (Kalali et al. 2011).

The results were statistically analysed using one-way analysis of variance (ANOVA). RSM was used to predict the relationship between the retention time (week), concentration of pesticide, concentration of phenol and the corresponding responses. The different retention times include 1, 2, 3, 4, and 5 weeks (Mohamad Thani et al. 2020).

RESULTS AND DISCUSSION

Efficiency of CW in landfill leachate treatment

The result shown in Figure 1 shows the high heavy metal removal rate from the landfill leachates by the CW. This heavy metal removal efficiency increases with an increased retention time. The increased removal of heavy metals by the system is attributed to the presence of macrophytes and the CW sediments (Yan et al. 2020; Ugya 2021). Macrophytes *P. stratiotes* and

S. molesta are able to accumulate heavy metals and concentrate them in their shoots (Nedjimi 2021). The concentrated Cu and Ni in the shoot is utilized for growth and development, hence the biomass of both plants increases (Fig. 4a). The increase of biomass shows that the macrophytes are resistant to the effects associated with the uptake of Pb, Cr, Cd, and Hg. The resistance of *P. stratiotes* and *S. molesta* to the accumulation of Pb, Cr, and Cd is due to immobilization and sequestration while the ability of *P. stratiotes* and *S. molesta* to remove Hg is by phytovolatilization (Garbisu and Alkorta 2001; Ugya et al. 2019c). The sediment from the CW also plays a significant role in the heavy metal removal from landfill leachates. This is because the heavy metal concentrations in the landfill leachates tends to affect the heavy metal status of the sediment due to the precipitation of the heavy metals on the sediment

(Ugya et al. 2021a; Qasaimeh et al. 2015). This is the reason why the concentration of heavy metals was higher in the sediment after treatment of the landfill leachate (Shanbehzadeh et al. 2014).

Figure 2 shows the high reduction efficiency of phenol, pesticide, sulphate, and chloride present in the landfill leachate. The reduction efficiency of phenol, pesticide, sulphate, and chloride present in the landfill leachate increases with increasing retention time. The mechanism involved in the high removal efficiency of phenol and pesticides in the CW is degradation. The degradation of phenol and pesticide present in the landfill leachate could be due to the plant-microbe interactions caused by the presence of *P. stratiotes* or *S. molesta* in the water, or due to the provision of an adequate carbon source and electron donation by phenol and pesticide to the microorganism in the soil, which in turn causes the degradation of

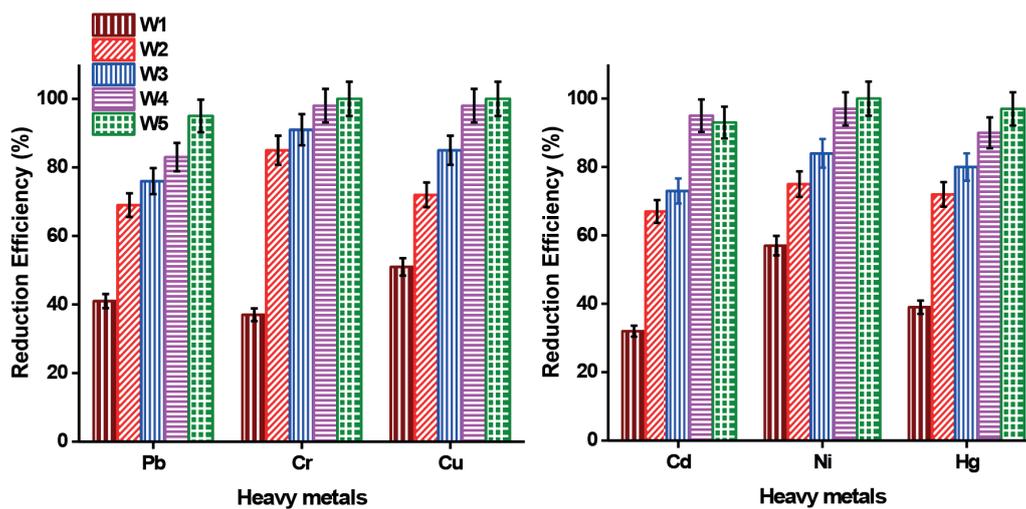


Figure 1. The removal of heavy metals from landfill leachates

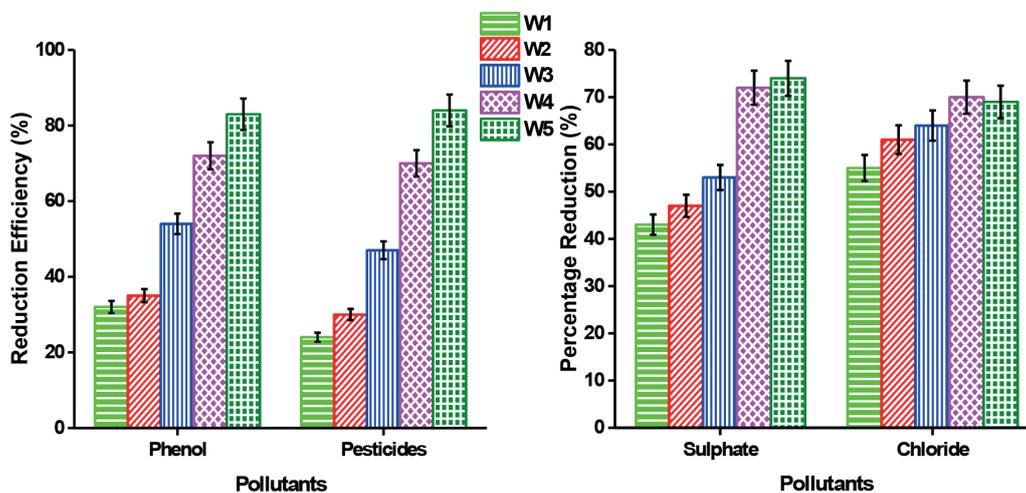


Figure 2. The removal of phenol, pesticides, sulphate and chlorides from landfill leachates

phenol and pesticides (Wong et al. 1990; Ribeiro et al. 2019). The high removal of sulphate from the landfill leachate is attributed to the presence of *P. stratiotes* and *S. molesta*. Both plants tend to phytoextract sulphur from sulphate for utilization. The sediment of the CW also plays a significant role in the sulphate removal from the landfill leachate because sulphate is precipitated as gypsum in the sediment (Aygun et al. 2019; Ugya et al. 2021b). The high removal rate of chloride from the landfill leachates is due to the presence of *S. molesta* and *P. stratiotes* (Page and Feller 2015). This plant phytoextracts chloride using the root and transports it via the xylem vessels to the shoot (Uraguchi et al. 2009). The high removal of chloride from the landfill leachate is due to the fact that the concentration of chloride in the landfill leachate does not exceed the cytosolic concentration of the root xylem, hence the reason why *P. stratiotes* and *S. molesta* avoided toxicity (Zalesny et al. 2008). The avoidance of the toxic effects of chloride accumulation by *P. stratiotes* and *S. molesta* is also due to the ability of both plants to release excess chloride via the process of transpiration (Nagarajan et al. 2012). The Figure 3 show the efficiency of the CW in the reduction of physico-chemical parameters such as colour, turbidity, TSS, TDS, BOD, COD, ammonia and

nitrate. The decrease in these parameters correlate positively with the increased retention time. The decrease in nitrate is due to the presence of *P. stratiotes* and *S. molesta* because both plants utilize both nitrate for growth and development. The removal of ammonia is dependent on the presence of the *P. stratiotes*, *S. molesta* and the sediments which are agents that stimulate the growth of ammonia oxidizing bacteria (To et al. 2020). The bacteria can either convert ammonia to dinitrogen gas or nitrate. The dinitrogen gas is released to the atmosphere to join the nitrogen cycle while nitrate is utilized by *P. stratiotes* and *S. molesta* for other metabolic functions (McCarty 2018; Ugya et al. 2019a). The high removal of colour from the landfill leachates is due to the ability of *P. stratiotes* and *S. molesta* to accumulate the contaminant that causes the colouration of the landfill leachates (Gowri et al. 2020). The high removal of TDS and TSS correlate positively with the high removal of turbidity and this is due to the presence of *P. stratiotes*, *S. molesta* and the CW sediment. Both the plants and CW sediment enhance particle sedimentation (Braskerud 2001; Pan et al. 2016). The high BOD and COD removal is attributed to the growth stimulation of microorganisms caused by *P. stratiotes*, *S. molesta* and the CW sediment. The microorganism uses the dissolve oxygen in

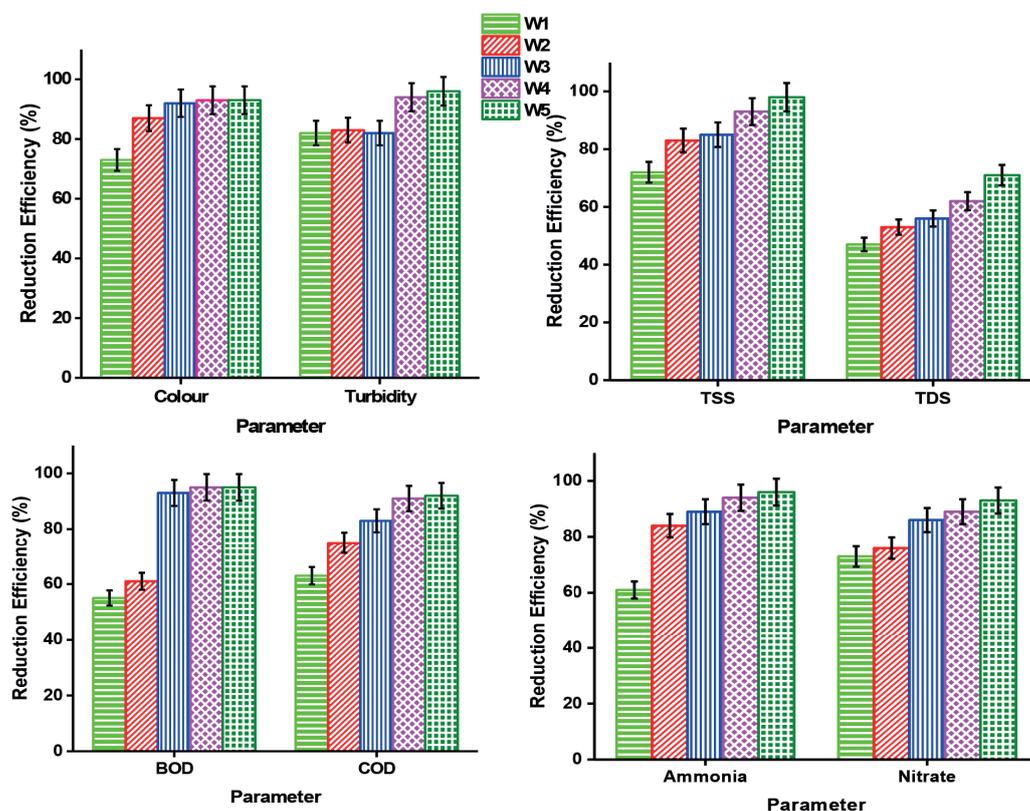


Figure 3. The removal of physicochemical parameters from landfill leachates

the breakdown of organic contaminant present in the landfill leachate (Ugya et al. 2019a).

Biomass and bioaccumulation factor of *P. stratiotes* and *S. molesta*

Figure 4 shows an increase in the biomass after the treatment of landfill leachates using both *P. stratiotes* and *S. molesta*. The increase in the biomass shows increasing carbon absorption, which is attributed to the ability of *P. stratiotes* and *S. molesta* to utilize organic and inorganic contaminants as energy and carbon sources (Kumar et al. 2017). The increase in biomass with after treatment of landfill leachates also shows the influence of the landfill leachate and CW sediment nutrient status on the rate of decomposition and respiration rate in *P. stratiotes* and *S. molesta* (Eid et al. 2021). This is because factors such as sediment quality, water chemistry, nutrient inputs, and interspecific competition play a role in the growth and establishment of macrophytes (Clarke and Wharton 2001).

Figure 4 also shows that both *P. stratiotes* and *S. molesta* have bioconcentration and bio-translocation factors greater than one for heavy

metals Pb, Cr, Cu, Cd, Ni, and Hg. This result confirms many studies that have demonstrated that *P. stratiotes* and *S. molesta* are hyperaccumulators (Mustafa and Hayder 2021). For the current study, the result displayed in Figure 4 confirms the role of *P. stratiotes* and *S. molesta* in the high reduction efficiency of heavy metals reported in Figure 1 (Suman et al. 2018).

Modelling of heavy metals in the CW

The relationship between heavy metals in the leachate, CW sediment, *plant tissues* was determined using correlation analysis. The correlation coefficient shows no linear dependence between the heavy metals in the leachate and heavy metals in the CW sediment, and plant tissues (Table 1). The results predict that the deviation from the linearity between the heavy metals in the leachates and heavy metals in the sediment and in the *plant tissues* is influenced by the physicochemical status of the leachate and the mixed cultivation of *Pistia stratiotes* and *Salvinia molesta* (Khan et al. 2009). The ability of the CW in the organic matters degradation present in the leachate causes an improvement of the leachate's

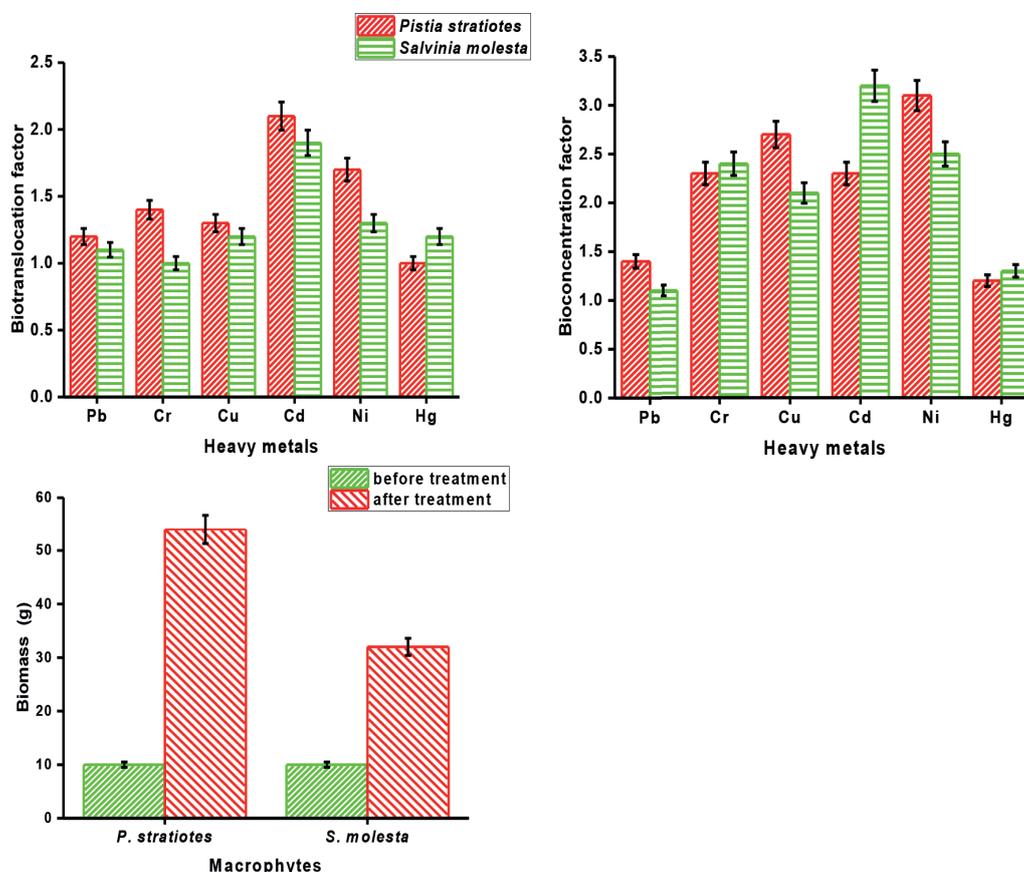


Figure 4. Biomass and bioaccumulation factor of *P. stratiotes* and *S. molesta*

physicochemical status (Strobel et al. 2005). This improvement in the physicochemical status of the leachates lowers the rate of bioavailability of the heavy metals, hence reducing the deposition rate of the heavy metals in the CW sediment (Selvi et al. 2019). The result presented in Table 1 shows a linear dependence between heavy metals in the sediment and *plant tissues*. The result shows that the bioavailability of heavy metals in the sediment enhances metal uptake by both *P. stratiotes* and *S. molesta* (Jiang et al. 2018). The result also reaffirms that the ability of *P. stratiotes* and *S. molesta* to remove heavy metal from landfill leachates in a CW is by bioaccumulation and utilization. Hence, this is the reason why the biomass of both *P. stratiotes* and *S. molesta* increases after the treatment of landfill leachates. The study reaffirms the role of sediment in the determination of the fate of metals due to its crucial role in the bioavailability of heavy metals for uptake by *P. stratiotes* and *S. molesta*. These are because the sediment is supposed to be the main sink for heavy metals in aquatic environment (Li et al. 2019).

But the current study shows that the sediment influence by its heterogeneity causes the bioavailability of metals in the system for the uptake by *P. stratiotes* and *S. molesta*. This is the reason why there is a linear dependence for heavy metals between *P. stratiotes*, *S. molesta* and sediment (Zhang et al. 2014; Tangahu et al. 2011).

Response surface methodology for the optimization of pesticides and phenol removal

The CCD study of the interactive effects of exposure and concentration of phenol and pesticides is presented in Table 2. The results obtained show a positive effect of concentration and exposure time on the reduction efficiency of both pesticides and phenol. The increase in removal efficiency of pesticides and phenol is due to the fact that the increase in exposure time tends to prolong the microbial cooperation of the synergistic microbial community present in the leachates (Huang et al. 2018). This microorganism tends

Table 1. Linear correlation coefficients (r) between heavy metal in leachate, sediment and plants materials

	Leachates	Sediment	Plants material
Pb			
Leachates	1	0.422**	0.646*
Sediment	0.422**	1	-0.152
Plants material	0.646*	-0.152	1
Cr			
Leachates	1	0.690**	-0.617**
Sediment	0.690**	1	0.904
Plants material	-0.617**	0.904**	1
Cu			
Leachates	1	0.192*	0.324**
Sediment	0.192*	1	-0.129
Plants material	0.324**	-0.129	1
Cd			
Leachates	1	0.432**	0.653**
Sediment	0.432**	1	-0.374*
Plants material	0.653	-0.374*	1
Ni			
Leachates	1	0.532**	0.513**
Sediment	0.532**	1	-0.347
Plants material	0.513**	-0.347	1
Hg			
Leachates	1	0.672**	0.575**
Sediment	0.672**	1	-0.192
Plants material	0.575**	-0.192	1

** Correlation is significant at the 0.01 level. * Correlation is significant at the 0.05 level.

Table 2. Composite design optimization of pesticides and phenol removal from the landfill leachates

Exposure time (week)	Concentration ($\mu\text{g/l}$)	Experimental result (%)	Predicted result (%)
Phenol			
1	4.760	32	31.798
2	4.550	35	38.220
3	3.220	54	51.382
4	1.960	72	73.197
5	1.190	83	82.774
Pesticides			
1	0.063	24	24.776
2	0.058	30	29.655
3	0.044	47	49.347
4	0.025	70	69.412
5	0.016	84	87.456

to interact with pesticides and phenol present in the leachates, this interaction causes changes in the structure of pesticides and phenol, leading to gradual and complete degradation (Jayaraj et al. 2016). This is also why the decrease in the concentration of pesticides and phenol in the leachates leads to an increase in the reduction efficiency (Lushchak et al. 2018). The result of polynomial regression between the predicted and experimental value shows a perfect fit. This result shows that the exposure time and concentration of phenol and pesticides are useful in the optimization of the removal efficiency of pesticides and phenol. Previous studies by Ting et al, (2020) had earlier shown the efficacy of RSM in the optimization of ammoniacal nitrogen removal (Ting et al. 2020).

CONCLUSIONS

The current study shows the efficacy of constructed wetland assisted by *Pistia stratiotes* and *Salvinia molesta* in the remediation of landfill leachates. The study shows high reduction efficiency of for phenol, pesticide, sulphate, chloride, colour, turbidity, TSS, TDS, BOD, COD, ammonia nitrate and heavy metals (Pb, Cr, Cu, Cd, Ni, Hg) from leachates by the system. The study further shows that the increase in the biomass of *P. stratiotes* and *S. molesta* is attributed to an increased carbon absorption, which is attributed to the ability of *P. stratiotes* and *S. molesta* to utilize organic and inorganic contaminants as energy and carbon sources. The study also predicts that the deviation from the linearity between the heavy metals in the leachates and heavy metals in the sediment and in

the *plant tissues* is influenced by the physicochemical status of the leachate and the mixed cultivation of *Pistia stratiotes* and *Salvinia molesta*. The study reaffirms the role of sediment in the determination of the fate of metals due to its crucial role in the bioavailability of heavy metals for uptake by *P. stratiotes* and *S. molesta* in a CW. The study also shows a positive effect of concentration and exposure time on the reduction efficiency of both pesticides and phenol. The polynomial regression between the predicted and experimental values shows a perfect fit. This result shows that exposure time and concentration of phenol and pesticides are useful in the optimization of the removal efficiency of pesticides and phenol.

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