Landfill leachate treatment by an experimental subsurface flow constructed wetland in tropical climate countries

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Abstract Municipal leachate was treated in an experimental unit of constructed wetlands of subsurface flow type. The parameters studied were organics (BOD and COD), solids and heavy metals (Zn, Ni, Cu, Cr and Pb). Using two types of emergent plants of *Scirpus globulosus* and *Eriocaulon sexangulare*, more than 80% removal was achieved for all the parameters. *E. sexangulare* removed organics and heavy metals better than *Scirpus globulosus*. A higher concentration of heavy metals in the influent did not change the removal efficiency.

Keywords Constructed wetlands; emergent plants; landfill leachate

Introduction

Constructed wetlands can be used to treat leachate from sanitary landfills (Rash and Liehr, 1999). Christensen et al. (1994) noted that leachate mainly consisted of heavy metals, organics with different biodegradation and inorganic matters such as ammonia, sulphate and cationic metals. Heavy metal removal using constructed wetlands ranged from 20 to 100%, depending on the metal types and wetland arrangements and Cu, Zn and Cd removal efficiency exceeded 97% (US EPA 1988). More than 50% of the removal was achieved through screening while the remainder was based on settling (Muller, 1988). Flyhammar and Hakansson (1999) and Lu et al. (1985) elaborated that the heavy metal removal included complexation, oxidation-reduction, sorption and precipitation. According to Surface (1993) and Suthersan and Suthan (2001), oxygen was transferred to the root zone (rhizosphere) to oxidize the substrate. The plant's root changed the biogeochemical environment of the soil, which affected the redox potential and pH and regulated the cation movement (Dunbabin and Bowmer, 1992) and the release of oxygen to the soil increased the redox potential (Amstrong, 1967). Mickle (1993) and Sundaravadivel and Vigneswaran (2001) observed that heavy metals were mainly retained in the root zones. Plants such as cattails had a lesser oxygen transport (Reddy et al., 1990 and Hunt and Poach, 2001). Heavy metal removal, may be initiated from the leaves in submergent plants (Matagi et al., 1998). Lead for example could only be retained in many plant leaves at a concentration limit of $1 \mu g/g$ (dry weight) as it is strongly bound to soils, and many plants are unable to uptake this metal. Other heavy metals like Cr, Cu and Fe have the same character as Pb. However, Zn, Cd, Mn and Ni have the opposite character (Calmano et al., 1993). Certain plants, however, may hyper accumulate certain heavy metals. *Minuartia verna*, for example, could retain lead up to $1,000 \mu g/g$ in its leaves (Kumar et al., 1995) however, it has been shown that the greener the leaves, the lower the metal content (Dunbabin and Bowmer, 1992 and Matagi et al., 1998).

In tropical climates, emergent plants treat wastewater throughout the year and nitrogen as the key element in wetland biogeochemical cycles, can be easily transferred as temperature and solar radiation play an important role in nitrogen volatilization (Vymazal 1995). Plants must grow in order to minimise direct sunlight to the soil as it may affect their survival IWA (2000). As the plants age, the rate of BOD removal increases (Brix, 1998).

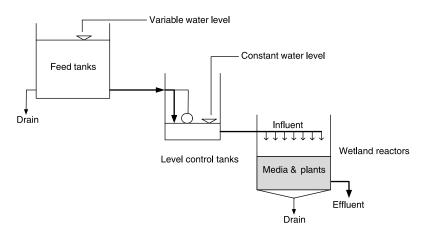
The objectives of this study were (a) to evaluate the performance of the constructed wetlands for organics (BOD and COD) and solids removal using municipal leachate and (b) to study the heavy metal removal for different concentration using emergent plants in constructed wetlands under tropical conditions. A laboratory scale experimental unit was constructed to carry out this study.

Materials and methods

A laboratory scale of sub-surface flow constructed wetland (SFCW) was constructed to carry out two stages of experiments: (a) organics and solids removal tests and (b) heavy metals removal tests. The configuration of the constructed wetland experiment is shown in Figure 1. There were 5 (five) reactors that were constructed in one unit of perspex glass. Each reactor had a dimension of $0.32 \text{ m} \times 0.32 \text{ m} \times 0.45 \text{ m}$.

Perspex glass materials were used to ease the observation and to allow light penetration for photosynthesis. The bottom media was crushed stones of 6 cm in height and 10 mm in diameter, while the media support was clean course sand of about 10 cm depth. The leachate was collected from a sanitary well inspection in Kulai. Flow to each reactor was 30 l/day that was equivalent to less than 0.03 kg BOD/day loading and was lower than proposed by the IWA Group (2000).

Reactor A was not planted for control purposes. Reactors B and D were planted with *S. globulosus* while Reactors C and E were with *E. sexangulare*. Each reactor was planted with three clusters of the grass. The grass was watered once a day in the reactors for three weeks. During the first stage, Reactors B and C were only dosed using the leachate from Kulai landfill site, Johor, Malaysia, while Reactors D and E were dosed with additional heavy metal to the leachate. Additional heavy metals included zinc (zinc sulphate), nickel (nickel sulphate), copper (copper nitrate trihidrate), chromium (potassium dichromate) and lead (lead nitrate). Heavy metal concentrations during the first stage



were the same as the initial 10 days in the second stage experiment. Analysis in the first stage included TSS, BOD, and COD of samples collected on days 3, 6, 9, 12, 15, and 18.

Heavy metal dosings in the second stage were carried out in two steps as shown in Table 1 as proposed by Mungur *et al.* (1997). The average heavy metal dosage during the first 10 days was 5 mg/l, while in the second period of 10 days was 10 mg/l, as shown in Table 1. Second 10 days of spike was only held for Reactor C and E.

Water samples were collected on days 5, 10, 15 and 20. Heavy metal analyses were carried out for the influent-effluent wastewater, leaves-steam and roots of the grass. Analysis was conducted as mentioned by Markert (1994).

Results and discussion

The characteristics of the leachate from the Kulai Landfill-site is shown in Table 2. The table shows that all the heavy metals analyzed are above the water quality standard. Based on the heavy metals concentration of raw leachate shown in Table 2, the sequence from high to low is as follows: Zn > Cr > Pb > Ni > Cu.

Organics and solids removal

After 18 days of the experiment, COD, BOD and TSS removal was higher than 85% (Figures 2, 3 and 4). The results show that the addition of heavy metals into the reactor did not affect the organics and solids removal that increased with time. The removal efficiency for organics and solids was higher than 85% after 18 days of sampling. The removal in the control reactor (without plants) was lower in comparison to the other reactors but also increased with time. Clogging within the soil matrix may increase the removal as found by Johnston (1991) and Gilliam (1994), whether the reactor was planted or not. The presence of plants resulting in more contaminants being removed was shown by Hupp *et al.* (1993) and Suthersan and Suthan (2001).

Table 1 Heavy metal dosages in the influent

Heavy metals	Concentration (mg/l) for the first 10 days	Concentration (mg/l) for the second 10 days		
Zinc (Zn)	2.9475	5.895		
Nickel (Ni)	3.3925	6.785		
Copper (Cu)	3.35	6.7		
Chromium (Cr)	4.25	8.5		
Lead (Pb)	3.0325	6.065		

Table 2 Influent characteristics of the Kulai leachate

Parameters	Mean concentration (mg/l)	Concentration according to EQA (mg/l)*
COD	950 - 1,000	_
BOD	350 - 400	-
Solids	300 - 400	-
Zinc (Zn)	1.6545	N/A
Nickel (Ni)	0.1260	0.075
Copper (Cu)	0.1038	0.075
Chromium (Cr)	0.5018	0.030
Lead (Pb)	0.2920	0.075

*Guidelines for groundwater and/or surface water supply project (DOE, 1997, and UPM & DOE, 1999) EQA stands for Environmental Quality Act (1974)

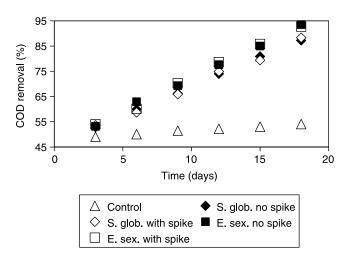


Figure 2 COD removal for municipal leachate using SFCW

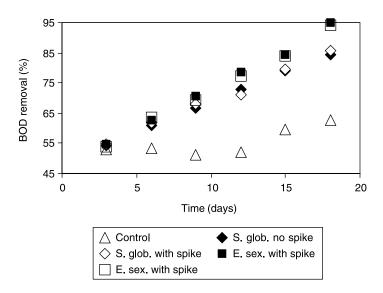


Figure 3 BOD removal for municipal leachate using SFCW

S. globulosus, the *E. sexangulare* could remove organics more efficiently than the control. The converse occurred for the removal of solids. The control reactor showed poorer removal of any contaminants analysed. The IWA Group (2000) stated that rhizomes played an important role in removing the contaminants. *S. globulosus* has more, deeper, and longer roots if compared to *E. sexangulare* which may help to trap solids.

Heavy metals removal

The results of additional heavy metal dosage (the second 10 days) as mentioned in Table 1 are shown in Table 3. Although Cr and Pb concentrations are still higher than the standard in Reactor E, the effluent results in Reactor E are lower than the one in Reactor D if compared to the standard. This shows that *E. sexangulare* can remove more heavy metals

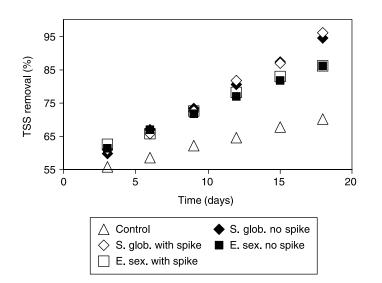


Figure 4 TSS removal for municipal leachate using SFCW

than *S. globulosus*. The results also show that effluent concentrations are increasing after the first 10 days in both reactors, which may be caused by saturation in the media. This was in contrast to the results in Reactor B and C where the effluent heavy metal concentration was relatively constant.

Heavy metal removal during the experiment is shown in Table 4. The removal in Reactors D and E were after the second 10 days addition of heavy metals. Although the results were not shown, the removal trends from each heavy metal through time (days 5, 10, 15 and 20) were not the same with even very low r^2 value. The results in Table 4, however, show that the heavy metal removals in the second 10 days were higher, especially for copper. Heavy metal removal using *E. sexangulare* was more pronounced.

Heavy metals retained in plants

Heavy metals retained in the leaves of the *S. globulosus* (Reactor B and D) are shown in Table 5. Table 6 shows heavy metals retained in the root zone of the same reactor. By comparing Tables 5 and 6, it can be seen that metal concentrations were higher in the roots than the leaves of *S. globulosus* and increased in both tissues over time. However, chromium concentrations decrease between 15-20 days in Reactor D. Tables 7 and 8 show the heavy metals retained in leaves and roots, consecutively, using *E. sexangulare* as the emergent plant in the reactor.

E. sexangulare showed similar results in Tables 7 and 8 to *S. globulosus*. The metals retained, either in the roots or leaves, were increasing over the time. More metals were also retained in the root zones and the concentration of chromium increased sharply in Reactor C. *E. sexangulare* in Reactor C could retain more Zn in the leaves or roots than in Reactor E. The converse occurred with *S. globulosus* as the emergent plants. Not only did the root of *E. sexangulare* achieve a higher concentration of zinc, but also retained more nickel and chromium. The root tissue of *E. sexangulare* in Reactor C retained more heavy metals than Reactor E indicating that its root system might not be able to cope with higher dosage of heavy metals. *E. sexangulare* retained more heavy metals than the *S. globulosus*, especially in the leaves.

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Heavy metals	Heavy metals In Reactor A, B, and C (ranges in mg/l)		Effluent concentration (range in mg/l)	ion (range in mg/l)		Effluent conc. limit (mg/l)*
		Reactor D (first 10 days)	Reactor D (second 10 days)	Reactor E (first 10 days)	Reactor E (second 10 days)	
Zn	0.106 - 0.887	0.497 - 1.228	0.86 - 1.022	0.169 - 0.257	0.218 - 0.247	1.5
ÏZ	0.017 - 0.096	0.446	0.825 - 1.558	0.052 - 0.114	0.283	1
Cu	0.008 - 0.031	0.104 - 0.336	0.112 - 0.174	0.013 - 0.031	0.019 - 0.03	-
ò	0.03 - 0.123	0.46 - 1.994	1.615 - 2.596	0.252 - 0.343	1.368 - 2.017	0.5
Pb	0.006 - 0.051	0.043	0.009 - 0.051	0.003 - 0.249	0.003 - 0.004	0.1
*According to	According to Environmental Quality Act (1974)					

Bold numbers represent higher concentrations than the standard

Table 4 Heavy metal removal in the reactors (%)

Heavy metals	Reactor A	Reactor B	Reactor C	Reactor D	Reactor E
Zn	65.8	89.8	86.9	84.6	96.2
Ni	45.7	81.3	77.6	85	96.8
Cu	72.2	86.6	83.9	95.8	99.5
Cr Pb	83.5 91.8	88.3 94.2	86.1 95.9	75.4 99.1	87.5 98.1

Table 5 Heavy metals retained in leaves of S. globulosus (μ g/g)

Heavy metals	tals Grow naturally 10 days 15 day		Grow naturally	Grow naturally 10 days	days	20 0	days
		В	D	В	D	В	D
Zn	10.81	57.8	48.47	82.81	108.6	108.6	174.71
Ni	17.18	6.25	9.99	12.5	50.45	18	55.12
Cu	1.08	12.45	5.32	12.78	21	13.67	20.43
Cr	29.75	30.07	34.44	24.65	31.64	20.67	30.43
Pb	NA	4.69	2.44	8.91	21	10.25	21.29

Table 6 Heavy metals retained in roots of S. globulosus (µg/g)

Heavy metals	metals Grow naturally 15 days		days	20 days	
		В	D	В	D
Zn	36.88	49.98	232.44	122.3	273.25
Ni	2.01	20.37	31.06	22.37	64.25
Cu	2.87	11.11	24.16	13.5	27.75
Cr	23.58	26.11	98.53	15.5	61.75
Pb	5.2	7.43	18.21	8.2	29.13

Table 7 Heavy metals retained in leaves of E. sexangulare (μ g/g)

Heavy metals	Grow naturally	10 0	10 days 15 days		lays	20 days	
		с	E	с	E	с	E
Zn	45.33	76.98	65.65	206.32	128.7	265.67	184.9
Ni	3.49	20.47	18.47	21.28	77.38	22.57	163.22
Cu	17.27	8.3	9.24	12.06	26.92	18.57	45.25
Cr	47.93	54.77	23.17	38.68	55.89	20.33	87.8
Pb	1.29	4.22	4.49	24.87	16.08	24.76	21.65

Table 8 Heavy metals retained in roots of E. sexangulare (μ g/g)

Heavy metals	Grow naturally	Grow naturally 15 days		20 days		
		с	E	с	E	
Zn	149.02	124.93	70.92	254.37	110.83	
Ni	7.34	6.58	35.39	95.5	75.58	
Cu	9.55	5.99	13.98	13.95	23.33	
Cr	93.8	28.52	69.84	150.75	64.67	
Pb	8.45	6.1	5.55	15.18	15.5	

Conclusions

Subsurface flow constructed wetland with plants reduced organic and heavy metal concentrations by more than 85%. *E.sexangulare* removed organics and heavy metals better than *S. globulosus*. The higher removal efficiency for *E. sexangulare* was probably due to a better oxygen transfer as it had a larger leaf surface area. A higher dosage of heavy metals did not change the removal efficiency of the reactors. *S. globulosus* removed solids more efficiently than *E. sexangulare* which was probably due to better roots development to intercept the particles.

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