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OPERATIONAL PERFORMANCE OF VERTICAL UPFLOW ROUGHING FILTER FOR PRE-TREATMENT OF LEACHATE USING LIMESTONE FILTER MEDIA

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Abstract: This study was conducted to investigate the removal of COD, BOD, turbidity and colour from leachate using vertical upflow filtration technique. Limestone media with a density of 2554 kg/m^3 was crushed and graded in sizes of 4–8 mm, 8–12 mm and 12–18 mm. Trial runs were done before the main experiment at an interval of 24 h analysis. Leachate was between pH 7.94 to 8.12 before experiments but increased to pH 8.42 after the filtration process. Maximum headloss at steady flow rate 20mL/min was 0.5 cm. The optimum treatment was achieved with 4–8 mm, 8–12 mm & 12–18 mm media size in combination and removal efficiency was 22 to 81%, 22 to 75%, 32 to 86%, and 36 to 62% for BOD, COD, turbidity and colour, respectively. Vertical upflow roughing filter can be used for pre-treatment of leachate before further treatment.

Keywords: Limestone; vertical upflow roughing Filter; leachate; landfill.

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INTRODUCTION

A leachate stream can be compared to a complex wastewater stream with varying characteristics not only because of the different kinds of waste present, but also varying according to the landfill age (Onay & Pohland, 1998). The leakage of leachate from landfill which contains high organic, inorganic, suspended solids, heavy metals and other pollutants can contaminate the ground water and surface water sources (Qasim & Chiang, 1994). The high level of chemical oxygen demand (COD), biochemical oxygen demand (BOD), turbidity, colour and other impurity components of landfill leachate make it very important to devise appropriate treatment methods.

Leachate used in this research was collected from Pulau Burung Landfill Site (PBLS) which is situated in Penang, Malaysia. PBLS has a semi-aerobic system and it is one of the only three sites of its kind found in Malaysia. PBSL has been developed semi-aerobically into a sanitary landfill Level II by establishing a controlled tipping technique in 1991. It was further upgraded to a sanitary landfill Level III employing controlled tipping with leachate recirculation in 2001. It has been found that the leachate from a semi-aerobic system has slightly lower organic contaminants compared with an anaerobic landfill in terms of BOD and COD (Aziz *et al.*, 2001). This site receives 1500 tons of solid waste daily. **Table 1** gives the composition of the leachate.

There are many different landfill leachate treatment options. These include complex and expensive methods from physico-chemical to biological processes for the treatment of high strength organics and inorganics. These could result in large costs on the long term. Alternative methods are continually sought to minimize expenses on leachate treatment (Tchobanoglous *et al.*, 2003).

Roughing filtration can be considered as a major pre-treatment process for wastewater and surface water, since the process efficiently separates fine solid particles over prolonged periods without addition of chemicals. Roughing filters mainly act as physical filters and reduce the solid mass. However, the large filter surface area available for sedimentation and relatively small filtration rates also supports adsorption as well as chemical and biological processes (Nkwonta, 2010). Roughing filters consist of differently sized filter media decreasing successively in the direction of flow. Most of the solids are separated by the coarse filter media near the filter inlet, with additional removal by the fine granular media in subsequent compartments.

Table 1. Composition of Leachate from Pulau Burung Landfill Site

Parameters	Range of Values, mg/L
BOD_5	48–1120
COD	1533–3600
Suspended Solid	159–1220
pH value (no unit)	7.8–9.4
Zinc	0.1–1.8
Manganese	0.6–1.1
Iron	0.32–7.5
Copper	0.1–0.4
Cadmium,	< 0.04
Colour, Pt.Co units	2430–8180

Source: Aziz *et al.* (2006).

Minitab is a statistical package having a suite of computer programs that are specialised for statistical analysis. It enables data to be processed to obtain results of standard statistical procedures and statistical significance tests, without requiring low-level numerical programming. Most statistical packages also provide facilities for data management (Quentin, 2010).

Limestone is a sedimentary rock, which is primarily composed of the mineral calcium carbonate ($CaCO_3$). As a result of the effectiveness of limestone in various treatment processes, it has been used for removal of contaminants from leachate. Sun (2004) reported 100% removal of iron from leachate in 150 min during batch experiments in which limestone was used as a filter medium to treat an iron acid solution (27.9 mg iron/L). Aziz *et al.* (2004) reported 90% removal of iron by limestone filter from landfill leachate containing 19.5 mg/L iron. The concentration of iron remaining in solution was 0.1 mg/L which was lower than the standard guidelines of 0.3 mg/L for the protection of aquatic life. A literature review indicated that limestone is capable of removing 90% of heavy metals such as Cu, Zn, Cd, Pb, Ni, Cr, Fe and Mn through a batch process and filtration technique (Aziz *et al.*, 2004). In another research work, Smith *et al.* (1994) used limestone filter to treat contaminated groundwater containing iron with concentration of 5 mg/L and reported a final concentration of 0.2 mg/L. The limestone and the limestone/sandstone filters successfully removed an average minimum of 97.60% of the iron from solution on a daily basis. Treatment of landfill leachate under aerobic batch conditions containing 6.6 mg/L iron and 1.8 mg/L manganese were also investigated (Ghaly *et al.*, 2007). It was observed that, the removal of manganese from solution was not as efficient as iron removal. In a related work, Xu *et al.* (1997) conducted batch experiments using calcite and quartz grains as filter media and reported iron removal of 99.8% from the acid mine. Adlan *et al.* (2008) evaluated the removals of turbidity, suspended solids, BOD and coliform organisms from wastewater using different sizes of limestone roughing filter. Results indicated that removal efficiencies depended on the size of the filter medium and applied flow rates. Turbidity, suspended solids, BOD and coliform organism removals were between 75 and 92%, 79 and 88%, 51 and 67%, and 67 and 96%, respectively, with particle sizes between 1.91 and 16.28 mm.

The roughing filter principle has also been used for the pre-treatment of water before supply to communities. **Table 2** gives a summary as reported by various researchers.

There is no report on the use of vertical upflow roughing filter for pre-treatment of leachate. In this study, limestone being a low cost material is used as filter media in the filtration process for the treatment of leachate. The objective of this research was to evaluate limestone as filter media for the pre-treatment and polishing of leachate and to examine the performance of the upflow vertical roughing filter for the removal of COD, BOD, turbidity and colour from leachate.

MATERIALS AND METHODS

Leachate used in this research was collected from Pulau Burung Landfill Site (PBLS), transported and stored at 4˚C in a cold room since weekly sampling was adopted. Limestone samples used in this study were obtained from the quarry industry located at Ipoh, Malaysia. The limestone chips composed of 95.5% $CaCO_3$, 3.0% $MgCO_3$ and 1.5% impurities (Aziz *et al.*, 2006). They were crushed and graded into sizes (12–18 mm, 8–12 mm and 4–8 mm) using motorized sieves. COD and colour were analyzed using DR/2800 Hach Spectrophotometer, method 8000 and 8025 respectively. BOD was analyzed using standard methods (APHA, 1992 and APHA, 1998) Method 5210B for wastewater analysis. The pH was measured by pH meter (CyberScan 20) while turbidity was measured using 2020 Turbidimeter (LaMotte). Analysis of variance (ANOVA) and Boxplot was performed using MINITAB Release 14.0 version.

Table 2. Performance of Roughing Filter

Reference	Filtration Rates (m/h)	Parameters	Mean Percentage Removed (%)
Pacini (2005)	1.20	Iron & Manganese	85 & 95
Dome (2000)	0.3	Algae & turbidity	95 & 90
Mahyi (2004)	1.5	Turbidity	90
Ochieng and Otieno (2004)	0.75	Turbidity & algae	90 & 95
Dastania (2007)	1.8	Turbidity, TSS &	63.4, 89 & 94
Jayalath(1994)	1.5	Coliforms Colour & Turbidity	50 & 80
Rabindra(2008)	1.0	TSS & Turbidity	95 & 95
Mukhopadhay (2008)	0.75	Turbidity	75

Experiments were conducted using both single and combination of media sizes packed in the column. This provided the opportunity to evaluate simultaneously the filter performance for the combination of media sizes and hydraulic loading rate over a constant filter length.

The three media size ranges (4–8 mm, 8–12 mm, and 12–18 mm) were used to assess the influence of flow rate, pore size and media density on BOD, COD, colour and turbidity removal efficiency. Filter media size were stacked in decreasing size from bottom towards the top for all experiments. The media was washed with 20 liters of dilution water before leachate was passed through the column. Five hydraulic loading rates (100 mL/min, 80 mL/min, 60 mL/min, 40 mL/min and 20 mL/min) were initially assessed in this study to determine the influence of interstitial fluid velocity on removal efficiency of the various parameters. A peristaltic pressure pump was used to generate the pressure for flow of the leachate from the residual tank through the vertical upflow roughing filter. The capacity of the peristaltic pump was 0 to 2000 mL/min. Three collection and monitoring ports at (300 mm, 600 mm and 900 mm) were provided on the filter length. Sampling was done after every 24 h and filtered through a 0.45 µm membrane filter. Analysis for BOD, COD, colour and turbidity removal was then conducted. For each of the flow rates, a new set of media was packed in the filter bed. There were four phases in the experiment to ensure that data obtained were consistent as shown in **Table 3.**

Figure 1 describes the sectional diagram of the filter column used. A detail of the characteristics of the filter column unit is as shown in **Table 4.**

The formula applied to calculate the percentage of COD, BOD, colour and turbidity removal efficiency is as per **Eq. 1**.

$$P = \frac{C_i - C_f}{C_i} * 100 \qquad (1)$$

where, P = percentage removal of impurity (%), C_i= initial concentration of impurity (mg/L or Pt-Co), and C_f = final Concentration of impurity (mg/L or Pt-Co).

Table 3. Experimental Phases and media packing in the vertical test column

Flow Rate (mL/min)	Media Size (mm)	Dilution	Duration (days)	Sampling & Analysis (hour)
	Phase 1			
100	12–18,8–12 & 4–8	No	1	24
80	″	No	1	24
60	″	No	1	24
40	″	No	1	24
20	″	No	1	24
	Phase 2			
	12–18,8–12 & 4–8	50:50 Water: Leachate		24
	Phase 3			
20	12–18,8–12 & 4–8	No	10	24
″	12–18	No	10	24
″	12 –18 & 4–8	No	10	24
″	8–12	No	10	24
″	4–8	No	10	24
	Phase 4			
50	12–18, 8–12 & 4–8	No	10	24

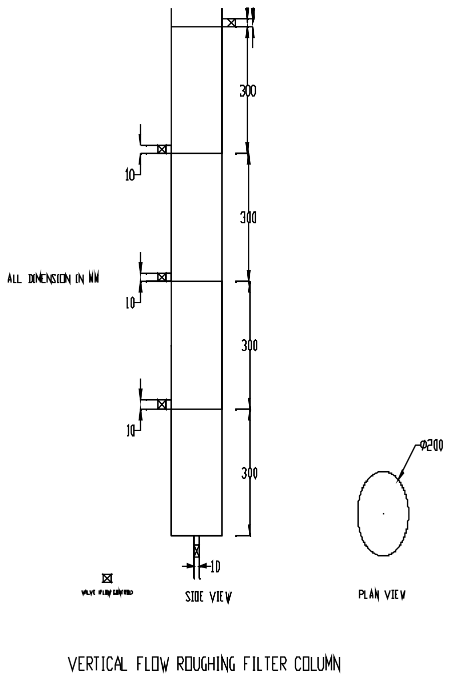

Fig. 1 Sectional Diagram of Vertical Upflow Roughing Filter. Column for the Study (Geometric Similarity; 1:3). Source: Wegelin, 1996.

Table 4. Characteristics and operational parameters of the column experiment

Parameter	Data	Unit
Flow rate	20	mL/min
Column Height	1500	mm
Internal Diameter		
Surface area of Column	200	mm
Column material	314	cm^2
	Perpex	
Total bed Vol. *net	plastic	
Particle size	28260	cm^3
	12-18, 8-12	
Limestone density	& 4-8	mm
Retention time		
Contact time	2554	kg/m^3
Filtration rate	31.4	hours
Mode of flow	24	hours
	0.2	m/hr
	upflow	

RESULTS AND DISCUSSION

Quality of raw leachate

Initial analysis of the raw leachate sample collected is shown in **Table 5**. The characteristics are representative of the methane fermentation phase. This phase is usually characterized by microorganisms, which converts the acetic acid and hydrogen gas formed by the acid formers in the acid phase to methane (CH_4) and CO_2, which become more predominant. Both methane and acid fermentation proceed simultaneously, but the rate of acid fermentation is considerably reduced compared to the former. Because the acids and the hydrogen gas produced by the acid formers have been converted to CH_4 and CO_2, the pH within the landfill will rise to more neutral values in the range of 6.8 to 8. In turn, the pH of the leachate will rise, and the concentration of BOD_5, COD and conductivity value of the leachate will be reduced.

With higher pH values, fewer inorganic constituents are solubilized; as a result, the concentration of heavy metals present in the leachate will also be reduced (Tchobanoglous & Kreith, 2003).

Table 5. Raw Leachate Quality

Parameter	**Unit**	**Reading**	**Aziz *et. al* (2006)**
COD	mg/L	2100-2530	1533-3600
BOD	mg/L	271-370	48- 1120
pH		7.94-8.12	7.8-9.4
Turbidity	NTU	226-274	50-450
Colour	Pt-Co	3310-3920	2430-8180

Weekly observation for three months period.

Performance of various media sizes

The removal efficiency of the limestone media (12–18 mm, 8–12 mm & 4–8 mm) in combination during the trial runs is shown in **Fig. 2**. Each flow rate was experimented for 24 h using different set of limestone each day. The best removal was observed at 20 mL/min. It shows that BOD, COD, colour and turbidity were removed 23%, 22%, 38% and 30% respectively. Hence, it was used for the subsequent experiments performed in phase three.

The removal efficiencies when 50 mL/min flow rate was applied to media 12–18 mm, 8–12 mm & 4–8 mm size is as shown in **Fig. 3**. It indicates removal in the range of 7% to 18%, 32% to 40%, 20% to 63% and 28% to 40% for BOD, COD, turbidity and colour respectively. The limestone media experienced a breakthrough except for turbidity.

The BOD,COD, turbidity and colour removal by the 12–18 mm 8–12 mm and 4–8 mm media combination is illustrated in **Fig. 4**. It indicates BOD removal of 22% to 81% , COD removal of 22% to 75%, turbidity removal of was 32% to 86% and colour removal of 36% to 62%. Except for turbidity, a decline in removal of the other parameters was observed after the sixth day owing to the optimum adsorption reached.

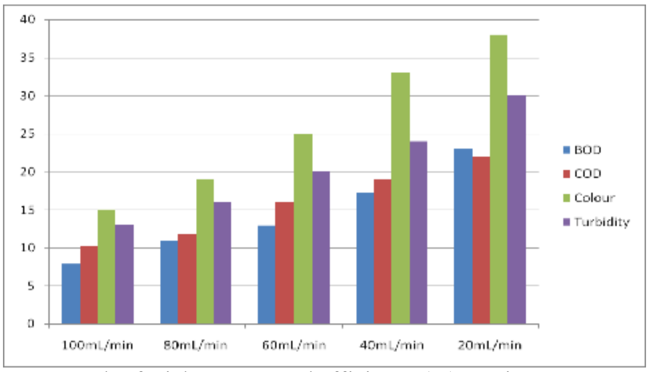

Fig. 2 Graph of trial run removal efficiency (%) various parameters versus flow rates.

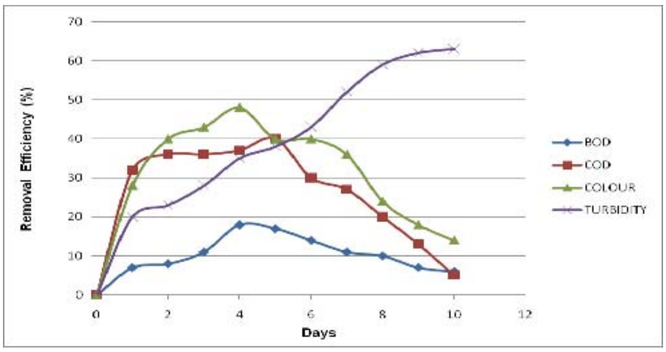

Fig. 3 Graph of (12–18 mm, 8–12 & 4–8 mm) removal efficiency (%) of various parameters versus days at 50 mL/min.

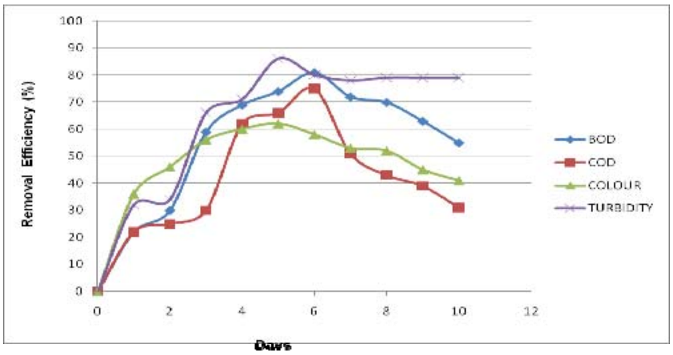

Fig. 4 Graph of (12–18 mm, 8–12 mm & 4–8 mm) removal efficiency (%) of various parameters versus days at 20 mL/min.

The limestone media 12–18 mm, 8–12mm & 4–8 mm sizes was experimented using a 50:50 (leachate:water) dilution. It indicated removal of BOD , COD, turbidity and colour of 46% to 68% , 27% to 46%, 58% to 82% and 60% to 70% respectively. Adsorption is the major process for removal of soluble organics such as COD from leachate especially when using columns in batch processes (Christensen *et al.*, 2001). This process involves adsorption of the contaminants by the microporous calcium carbonate (limestone) used as filtration media into their sites. In a study, analysis of the limestone media after filtration indicated that adsorption and absorption processes were among the mechanisms involved in removal of organics (Christensen *et al.*, 2001). The low removal of COD was probably due to reduction in the concentration of the adsorbate molecules of the leachate onto the limestone media sites (i.e. the molecules being accumulated on the surface and sites of the adsorbent).

Figure 5 illustrates limestone media 12–18 mm size removal efficiency. BOD removal was 22% to 41%, COD removal was 9% to 27%, turbidity removal was 62% to 66% and colour removal was 33% to 56%. Low removal in BOD and COD can be attributed to large media size which resulted in reduced specific surface area of the media adsorption.

Figure 6 shows the pattern of removal that occured in the 12–18 mm & 4–8 mm, media. In the experiment, BOD removal was 34% to 60% , COD removal was 19% to 54%, turbidity removal was 32% to 86% and colour removal was 35% to 52%. After the seventh day, the media was fully adsorbed and hence showed reduction in its removal capacity except for turbidity.

The percentage removal for 8–12 mm size media was 14% to 61% , 32% to 62%, 17% to 83% and 34% to 61% for BOD,COD,turbidity and colour respectively. Breakthrough was experienced in all the parameters

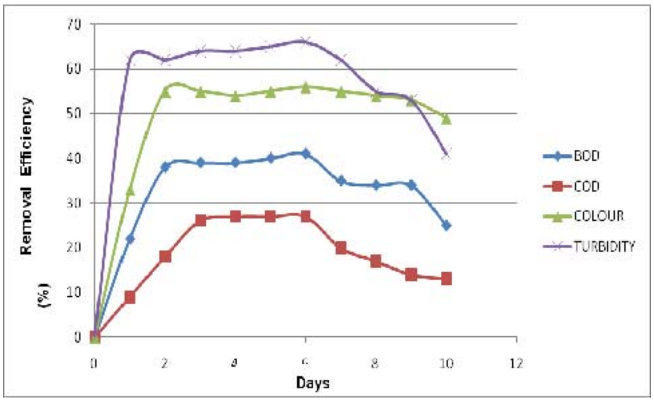

Fig. 5 Graph of (12–18 mm) removal efficiency (%) of various parameters versus days at 20 mL/min.

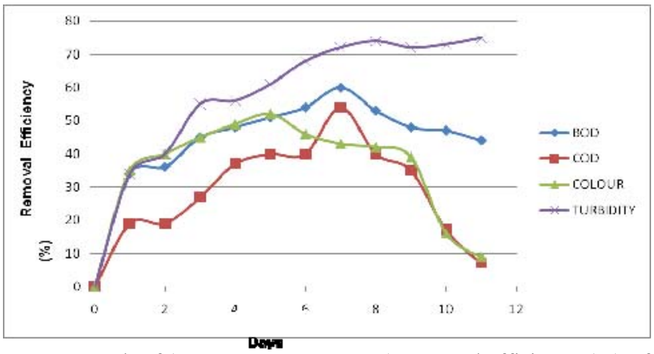

Fig. 6 Graph of (12–18 mm & 4–8 mm) removal efficiency (%) of various parameters versus days at 20 mL/min.

except in turbidity which was however not very pronounced on the tenth day of the experiment.

The 8–12 mm media size (graph not shown) had BOD removal of 6% to 23%, COD removal of 24% to 42%, turbidity removal of 18% to 37% and colour was 39% to 45%. Flow continued until media pores were blocked at end of the fifth day causing experiments to terminate.

One-way ANOVA analysis performed on the experimental data is shown in **Table 6**. The P column indicates the significance level: a P-value less than 0.05 indicate that the variable (factor) is significant to a level of 95.0%. The p- value of the media size was p = 0.317 > 0.05. This affirms that media 8–12 mm size was not significant for the pre-treatment of the leachate. In several other figures not included in this work, box plots were also performed to indicate which observations, might be considered outliers in the distribution efficiency.

TURBIDITY REMOVAL

Effect of filter media size on turbidity removal

The most important factor enhancing turbidity removal with respect to filter media size is the reduced pore spaces between the grains of the media in the filter bed.

Table 6. One-Way ANOVA summary for phase 2, 3 & 4 experiments

Media size (mm)		Flow rate (mL/min)	P value	Phase
12–18,8–12 & 4–8		50	0.016	4
12–18,8–12 & 4–8		20	0.011	3
12–18,8–12 & 4–8	@50% dilution	20	0.090	2
12–18		20	0.043	3
12–18 & 4–8		20	0.082	3
8–12		20	0.317	3
4–8		20	0.094	3

Smaller grain sizes have larger adsorption area and perform better in treatment processes (Tamar, 2008; Wegelin, 1996). In this experiment, the specific surface area and particle size arrangement for limestone media 12–18 mm, 8–12 mm & 4–8 mm were sufficient to reduce turbidity of the leachate.

Effect of flow rate on turbidity removal

The higher pilot flow rates strongly affected the resultant effluent turbidity values because the main mechanism for removing turbidity is filtration. The higher the flow rate, the less time a particle has to travel the settling distance and stick onto the media's surface and layers or be adsorbed. Higher flow rates would have been desirable if they produced greater quantities of treated leachate with better removal efficiency.

Results obtained from the roughing filtration for treatment of water, (Nkwonta & Ochieng, 2009) and Muhammad *et al.* (1996) show that flow rates when lower will remove turbidity more effectively. However, Wegelin (1996) upholds that pressure drop will be a setback for such filtration unit. With this in mind, operational flow rate for a desired turbidity to be achieved was considered and implemented.

COD AND BOD REMOVAL

Effect of filter media size on COD and BOD removal

Biological "ripening" of filter media may improve particle removal efficiency in roughing filters due to increased stickiness of filter media (Collins *et al.*, 1994). Exposure of the media to sunlight was avoided to minimize potential errors which can be introduced by biological activity into the column. To evaluate the potential in removal of BOD and COD, the experiment was non-recycle and upon final use of the media it was observed to be very sticky by means of touching. In addition, media 4–8 mm, 8–12 mm and 12–18 mm size had a higher removal as against 8–12 mm, 4–8 mm alone and other combinations. This could be as a result of increased surface area and hence adsorption sites. This could be similar to Wegelin (1996) slow sand filtration experiments where removal of pathogens was by the sedimentation or deposition and effective settling of particles on the grains of the media. Thus, COD and BOD_5 are possible to be reduced from leachate by limestone media.

Effect of flow rate on COD and BOD removal

The flow rate which removed optimum COD and BOD was observed to be 20ml/L. This was obvious when higher flow rates indicated lower removal of COD and BOD. However, a more critical factor that can be attributed to the low removal was the media saturation point that must have been reached. Lower flow rate allows an increased pathogen removal, which is especially important in colder climates where biological activity is more time dependent (Huisman & Wood, 1974; Wegelin, 1996; Nkwonta & Ochieng, 2009). Recent research carried out by Jenkins *et al.* (2009) on intermittently-operated sand filters has highlighted the importance of sand size and hydraulic loading both of which directly affect microbiological removal. However, it is possible to increase the filtration rate if effective pretreatment has been given and if an effective disinfection stage follows after filtration (Ellis, 1987).

COLOUR REMOVAL

Effect of flow rate on colour removal

A report observed that colour removal was poor when higher filtration rates were applied in continually-operated sand filters, although the filtrate quality remained reasonably good (Muhammad *et al.*, 1996). In this study, it was justified and reasonable to apply lower flow rates for optimum removal to be obtained. The limestone media was not capable of removing the colour from the leachate. It would therefore require a chemical or biological treatment for effective removal of colour.

Effect of filter media size on colour removal

The removal of dissolved colour or true colour by varying filter media size in roughing filtration has not been properly documented. Since, colour is related to humic substances, it is expected that true colour exists in relatively stable suspension and it is more difficult to remove. Collins *et al.* (1994) reported that removal of true colour in roughing filters compares favourably to that achieved by slow sand filtration. Wegelin (1996) observed true colour removal in the range of 20 to 50 %. There has been numerous reports of removal of apparent colour, which is the colour attributed to undissolved particulate matter. Wolters *et al.* (1989) and Barret (1989) both found removal of apparent colour to be 45 to 80 %. In this study, the optimum true colour removal was 62% using the media 12–18 mm, 8–12 mm & 4–8 mm size combination.

Length of run time on removal efficiencies

The length of run for each set of media combination study was ten days. The limestone media experienced an average breakthrough in a period of six days as found from the laboratory experiments. The length of run was basically dependent on the performance of the media in terms of its removal and adsorption capacity. In each study, when breakthrough was experienced the experiment was terminated.

pH Change during the roughing filtration process

During the roughing filtration treatment the pH showed a little variation. The raw leachate sample was between pH 7.94 and pH 8.12 but increased to pH 8.42 after filtration. This is due to the presence of CO_3 in limestone (Aziz *et al.*, 2004) an unstable oxide of carbon and declined as removal efficiency reduced. The dissolution of calcium carbonate can increase the concentration of alkalinity.

Headloss during length of run time

The headloss was also considered in this study. Although, the main essence of conducting the research was to ascertain the effectiveness of the limestone media in pre-treatment of leachate, a documentary of the headloss during each run time for every media combination was randomly done. To affirm the postulation by Boller & Kavanaugh (1995) which demonstrated that the rate of headloss build-up in a granular media filter, for a constant mass of solids being removed, is strongly dependent on the size of the particulates in suspension and the size of the granular media. In addition, the principal cause of the rapid increase in headloss observed for smaller particles in many filtration studies is due to the nature of the aggregation or deposition process inside the pore spaces of the porous media. Observations during this experiment indicated that maximum headloss was 0.5 cm at 20 mL/min flow rate. According to Amin & Aziz (2002), negative heads do not occur in the upflow vertical roughing filter bed, however a crack occurs at the bottom layers of filter bed, when headloss reaches maximum. It is noteworthy that, during the filtration treatment process for media 4–8 mm size, there was an entire blockage and flow stopped abruptly.

CONCLUSION

- The optimum removal efficiency of BOD, COD, colour and turbidity using the limestone media in the vertical upflow roughing filter stacked with 12–18 mm, 8–12 mm and 4–8 mm size from bottom towards the top of the column at 20 mL/min and with no dilution of leachate was 81%, 75%, 62% and 86%, respectively. During the 50:50 water-leachate dilutions, colour removal was 70 %.

- Increasing filtration rate and reducing contact time simultaneously reduced adsorption and hence removal efficiency.

- The vertical upflow roughing filter can be used for pre-treatment or polishing of leachate before further biological treatment.

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