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Effective use of iron-aluminum rich laterite based soil mixture for treatment of landfill leachate

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ABSTRACT

Landfill leachate poses environmental threats worldwide and causes severe issues on adjacent water bodies and soil by direct discharge. The primary objective of this study is to analyze the efficient use of compost and laterite mixtures (0, 10, 20, 30 and 40 wt% compost/laterite) on leachate treatment and to investigate the associated removal efficiencies under different sorption processes. Therefore, in the experimental design, laterite is used for providing adsorption characteristics, and compost for activating biological properties of the filter. The filtering process is continued until major physical changes occur in the filter at approximately 100 days. The raw leachate used for the experiment shows higher average values for many analyzed parameters. Parameters for the experiment are selected based on their availability in raw leachate in the Sri Lanka. During filtering, removal efficiencies of BOD (>90%), COD (>85%), phosphate (>90%) and nitrate (75–95%) show higher values for all filters. These removals are mainly associated with biodegradation, which is activated by the added compost. Perhaps the removal of nitrate steadily increases with time, which indicates in denitrification by the added excess carbon from the leachate. The removal of total suspended solids (TSS) is moderate to high, but conversely, the electric conductivity (EC) is unsteady, indicating an association between iron exchange and carbonate degradation. A very high removal efficiency is reported in Fe (90–100%), and wide ranges of efficiencies in Mn (30–90%), Cu (45–85%), Ni (30–93%), Cd (37–98%), Zn (15–98%), and Pb (35–98%) involve heterogeneous sorption processes. Furthermore, the normalization of raw leachate by the liquid filtrate has apparent improvements. The differences ($p > .05$) in removal efficiencies between the filters are significant. It can be concluded that the filter with laterite mixed with 20% of compost has the optimum conditions. Further, the Fourier-transform infrared (FT-IR) models for filter media conclude multiple sorptions and reveal evidence on vacant sites. X-ray diffraction (XRD) analyses indicate secondary minerals gibbsite, hematite, goethite and kaolinite as the major minerals that involved on the sorption process.

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1. Introduction

Municipal solid waste (MSW) is a major controversial issue worldwide (Karak et al., 2012; Pelleria et al., 2016). New trends in living standards, population growth and economic developments are the major causes of rapid generation of MSW (Renou et al., 2008; Karak et al., 2012; Pelleria et al., 2016). The solid waste landfill is known as more economical disposal method than incineration and composting (Renou et al., 2008; Pelleria et al., 2016). Most developed countries use sanitary landfills. However, non-

engineered disposal methods are common throughout the world (Trankler et al., 2005; Wijesekara et al., 2014).

Leachate is generated in landfills by their internal chemical and biological processes, rain water percolating through the waste and higher moisture level, as well as and liquid release by the reactions (Rivas et al., 2004; Renou et al., 2008). The heterogeneous compositions in leachates depend on the nature of the landfill (Lema et al., 1988; Rivas et al., 2004), age of waste (Chian and DeWalle, 1976; Kulikowska and Klimiuk, 2008; Abbas et al., 2009), climate (Chen, 1996), degree of compaction (Lema et al., 1988; Abbas et al., 2009), waste type and particle size (Lema et al., 1988; Renou et al., 2008). In addition, subsurface lithology and landfill technology have severe impacts on the composition and mobility of leachate (Christensen et al., 2001; Renou et al., 2008).

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Open landfilling is a popular method for disposing of MSW in developing countries, such as Sri Lanka. These sites are not properly managed, due to technological barriers and over-dumping (Basnayake et al., 2008; Hettiaratchi et al., 2010; Sewwandi et al., 2013; Wijesekara et al., 2014). Untreated leachate poses severe threats, especially in open landfills located in sensitive areas, such as forest margins, roadsides, mangroves, and river catchments (Slack et al., 2005; Hettiaratchi et al., 2010). This leads to several acute and chronic health problems. Eutrophication is also a common phenomenon in surrounding waters (Bandara and Hettiaratchi, 2010; Wijesekara et al., 2014). Apparently, a proper leachate treatment system is a primary need for landfills, particularly in developing countries. Natural clay layers in the weathering profile around landfill sites act as a barrier for leachate mobility (Regadío et al., 2015), which give clues for the use of clay on leachate treatments. Many countries have different treatment systems that are found to be unrealistic, due to the high costs involved (Abbas et al., 2009). Thus, a cost-effective technique has a higher demand.

Filtering of leachate is considered one of the most popular methods (Madhukar et al., 2012). Cost- and time-effective sorption techniques are used for leachate filtering (Chen et al., 2007b). However, complex constituents in leachates have comprehensive chemical and physical properties (Kulikowska and Klimiuk, 2008). Therefore, a single sorption material does not provide optimum conditions. Hence, it is necessary to use a few sorption materials together that have different properties (Svensson et al., 2011; Harlina et al., 2016). For instance, clay minerals adsorb cations to the surface; however, some univalent cations can only absorb in a few types of clay. Conversely, biodegradation should be activated by adding compost to control the redox condition and pH, as well as for the sorption of some anions.

Iron- and aluminum-rich laterite is a common soil type, which is mainly available in tropical countries under wet climate (Oliveira et al., 1992; Thorne et al., 2012). Laterite is considered to have better sorption characteristics of contaminants (Madhukar et al., 2012). Hence, laterite has been used as an effective filter media for contaminated water (Kadam et al., 2009), but it has limited applications in landfill leachate treatments (Syafalni et al., 2012; Harlina et al., 2016). This is mainly because the sorption nature of laterite is inappropriate to filter the complex constituents. To improve the biological properties in laterite, biofiltering should be activated; thus, compost can be used as an enrichment media (Gibert et al., 2003; Rose et al., 2012). The added compost can stimulate microbial activities, thereby improving the filter performance (Jurado et al., 2015). The low-density compost helps to increase the retention time of leachate in the filter media by becoming spongy in nature (Rose et al., 2012). Additionally, the mixed filter materials provide steady conditions for filtering, rather than layer-driven filters, by mitigating the handling and operational efforts (Svensson et al., 2011). Therefore, the scope of this study is to develop laterite and compost mixed materials as a filter medium to remove different constituents produced by landfill leachate. The main objectives are to explore an effective leachate filtering system and to investigate the affiliated sorption processes, biodegradation and removal efficiencies.

2. Materials and method

2.1. Material collection

2.1.1. Laterite and compost sample collection

Laterite soils available in the wet zone of Sri Lanka are extensively developed by in situ weathering of metamorphic rocks (Dahanayake, 1982). Laterite contains large amounts of aluminum,

iron, and manganese, with the depletion of free silica and very little or no alkaline. In this experiment, hard laterites acted as the major filter media. Composite laterite samples were collected by auger drilling to homogenize the material. The required compost for the enrichment material was gathered from a standard compost production plant (Tyrrel et al., 2008). Samples were stored in 4 °C cooling boxes, and physicochemical parameters were immediately analyzed (Jayawardana et al., 2012).

2.1.2. Collection of leachate sample

Leachates were gathered from an open active dumping site in Karadiyana, Colombo, Sri Lanka. The total area of the site is approximately 25 acres. Approximately 575 tons of MSW were received daily from 7 local authorities. The samples were collected into 2 L polypropylene bottles on a weekly basis and stored in 4 °C cooling boxes. The basic physicochemical parameters of leachate were immediately tested (Sewwandi et al., 2013).

2.2. Physico-chemical analyses of laterite and compost

The pH of the soil suspension solutions (1: 2.5 = soil: water) and compost were taken using WTW ProfiLine pH 3110 pH/mV meter with ± 0.005 variance. The oxidation redox potentials (ORP) of the soil and compost samples were also measured, with SenTix® ORP electrode with ± 0.3 mV variance. The ORP and pH values were measured by means of wet sediment analyses method. In the analyses, suspension samples were used to maintain the soil and compost slurry in homogeneity. The conductivity of soil samples was measured using WTW ProfiLine cond3210 conductivity meter with $\pm 0.5\%$ variance of measured value. Moisture content of laterite and compost were measured by direct percentage weight difference method (1):

$$\text{Moisture content} = \frac{W_{\text{wet}} - W_{\text{dry}}}{W_{\text{wet}}} \times 100 \quad (1)$$

where W_{wet} is initial weight of sample and W_{dry} is the dried sample at 105 °C until the samples reached constant weights.

A loss on ignition (LOI) test helped to estimate the organic matter and inorganic carbon contents in laterite (Wang et al., 2011). A split of the sample was ignited at 450 °C for 24 h to determine the organic matter content, and it was further ignited to 1050 °C for 2 h to estimate the carbonate content. The Walkley-Black wet oxidation technique helped to determine the total organic carbon of laterite and compost (Walkley and Black, 1934).

The laterite and compost samples were analyzed for available major and trace elements by X-ray fluorescence spectrometry, using the Rigaku NEX CG EDXRF analyzer. Splits of each sample were oven-dried for 48 h at 160 °C. Powdered samples ($< 63 \mu\text{m}$) were compressed into briquettes under a force of 200 kN for 60 s. The briquettes were then analyzed for selected major oxides and trace elements. The average error for these elements is less than $\pm 10\%$ relative. The available phosphorus of soil was determined by a spectrophotometer (CECIL CE 1021 series) using sulfomolybdic acid reagents. The total nitrogen content of the compost sample was measured by the Kjeldahl method.

2.3. Filter material preparation and experimental design

The flowchart on the experimental design is presented in Fig. 1. Laterite and compost were air dried for 24 h to remove the moisture and crushed using an agate mortar and pestle. The ground materials were sieved using 0.5 mm and 0.2 mm US standard sieves for compost and laterite, respectively (Kettler et al., 2001; Chen and Wang, 1997). Laterite was uniformly mixed with compost using a mechanical shaker to properly blend the materials while maintaining the compost to laterite weight ratio of 10:90

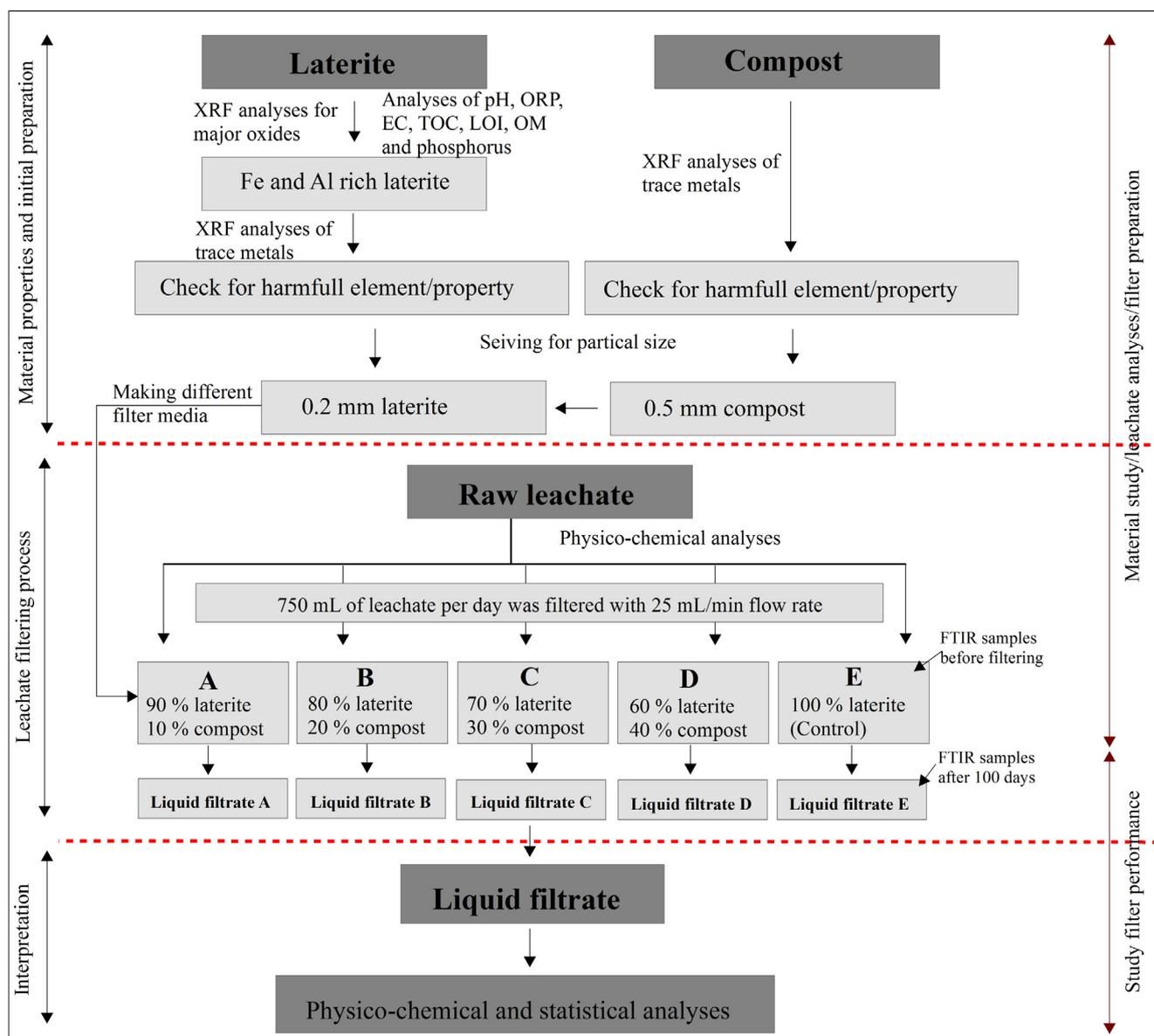


Fig. 1. Flow chart for the research methodology. Red dotted line separates the two major areas of the experimental procedure. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

(filter A), 20:80 (filter B), 30:70 (filter C), and 40:60 (filter D) (Yidong et al., 2012). The raw laterite without mixing compost was used as the control (filter E). Compost was added in 10% intervals because biological filtering is the secondary process; hence, smaller changes in the compost will not impact the overall system.

The experimental setup was prepared in a glass box having five equal columns for filters A, B, C, D, and E (Fig. 2). The prepared filter materials were gently filled into each column separately. Typically, a column is filled with 2 kg of filter materials, and the total volume per column was equal to 0.002 m³ (H, 0.20 m; W, 0.10 m; L, 0.10 m). The top and bottom of each column were covered by gravel (0.002 m) and sand (0.002 m) layers respectively to maintain a consistent flow (Fig. 2; Daud et al., 2015).

The experiment of the filtration process was conducted over 100 days. In general, filter materials after ninety-four days showed a reduced nature and noted physical changes due to rapid mineralization of organic matters. Therefore, filtering was terminated by 100 days of operation. Thirty-one feeding events were included in the filtering process. A two-day gap was maintained between

two feeding events to settle the filter materials for effective use (Fig. 2). Raw leachate was diluted by water in a 1:1 volume ratio before feeding in order to reduce the thickness of leachate for effective filtering (Perera et al., 2014). Leachates were poured under 25 ml/min flow rate on each filter column (Madhukar et al., 2012). Each feeding event included 3 feeding sessions. Approximately 0.75 L was fed to each filter in three separate sessions per day (0.25 L per session). Between the feeding sessions, 1–2 h was given to facilitate trickling (Hongjiang et al., 2009). In general, the overall time required to filter 0.75 L in a single filter column was 6–7 h per day (Kadam et al., 2009).

2.4. Physicochemical analyses of leachate and liquid filtrate

The major physical and chemical properties of each raw leachate and leachate obtained after the filtration (liquid filtrate) were tested after each feeding event. Electrical conductivity (EC), oxidation reduction potential (ORP), and the pH of the leachate and liquid filtrates were directly measured using portable Horiba

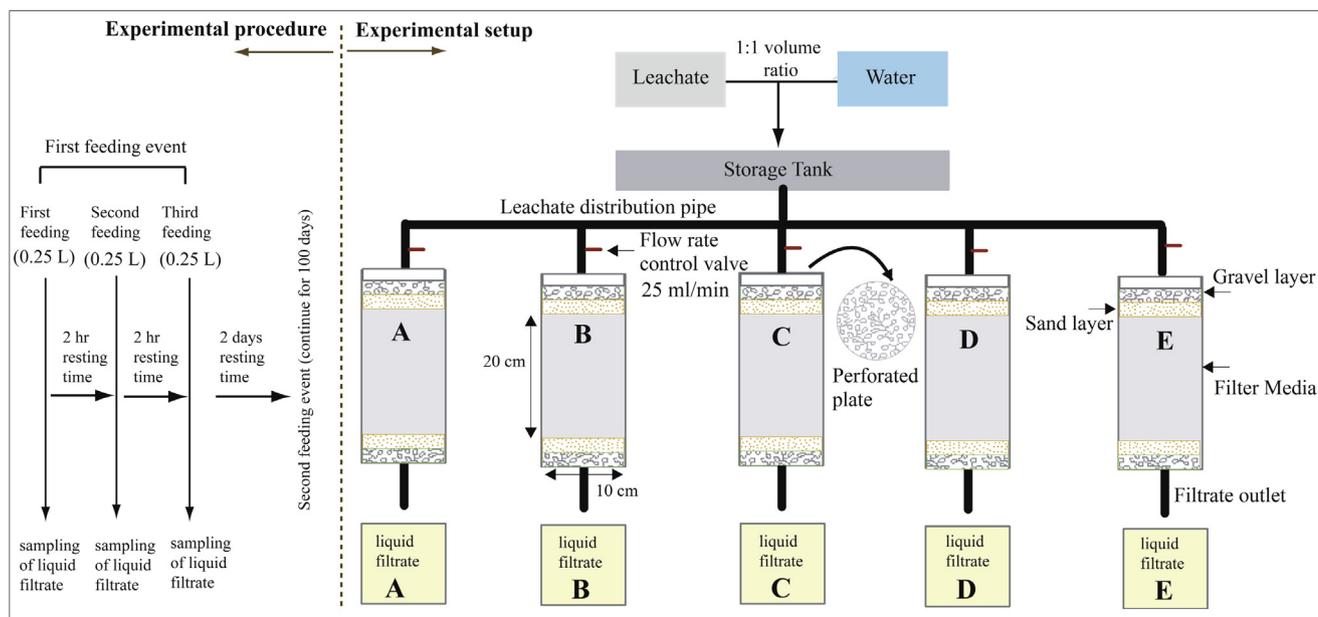


Fig. 2. Experimental design and arrangement of the five different filter systems.

U-23 combined instrument (Rikagaku kenkujo, Japan). The average accuracy for ORP and EC are ± 0.3 mV and $\pm 0.5\%$, respectively. EC is an important factor that reflects the total ions in the leachate and filtrate, which reflects the overall performance of filtering. Further, ORP and pH are coherent variables that control the mobility of redox sensitive elements; hence, they are quite significant for biofiltering activities.

Concentrations of common trace elements in solid waste of Sri Lanka, such as Fe, Mn, Ni, Pb, Zn, Cd, and Cu, in the leachate and liquid filtrate were determined using an Atomic Absorption Spectrometer (AAS, 932 plus series). The flame atomic absorption version of the instrument was used according to standard procedures (Taylor, 1999). The instrument was linearly calibrated with custom single-element standards (Ahmed et al., 2010). The detection limit of the instrument was 1 ppb, and the average accuracy was $\pm 0.5\%$.

BOD₅, COD, TSS, nitrate, and phosphate of the leachate samples were measured according to the standard methods given to water. BOD₅ and COD were measured using the wrinkle method and dichromate method, respectively (Chen et al., 2007a; Wijesekara et al., 2014). The concentration of TSS was tested using a filtration method followed by oven drying at 105 °C. Nitrates and phosphates were analyzed via UV spectrometric method.

2.5. Efficiency and performances of filtering

The most crucial factor in the leachate treatment process is the removal efficiency (2) (Chen et al., 2007a; Yidong et al., 2012; Harlina et al., 2016; Lu et al., 2016):

$$\text{Removal efficiency (x)} = \frac{[\text{RL}]x - [\text{F}]x}{[\text{RL}]x} \times 100 \quad (2)$$

where [RL]x is the raw leachate concentration of parameter 'x' and [F]x is the measured liquid filtrate concentration of the 'x'.

Performance analyses of the liquid filtrates were checked with respect to water quality standards mainly given for drinking water (Table 1), and the parameters were compared to the standards on industrial effluent specified by the Central Environmental Authority of Sri Lanka (GEDSRSL, 2008), Sri Lanka Standards for Potable Water (SLS; SLSI, 2013), drinking water standards proposed by

the United States Environmental Protection Agency (USEPA, 2017), and drinking water standards of the World Health Organization (WHO, 2011).

The first-order Yoon–Nelson model was used to evaluate the kinetic properties of the filter media (Yoon and Nelson, 1992). The model is based on the assumption that the decreasing rate of the probability of adsorption for each adsorbate molecule is proportional to the probability of adsorbate adsorption. The model has been used to evaluate the kinetic properties of different absorption media, including clay (Aksu and Gonen, 2004; Han et al., 2009; Ghribi and Chlendi, 2011). According to the model, different studied variables are evaluated (3):

$$\ln(C_t/C_0 - C_t) = k_{YNt} - \tau k_{YN} \quad (3)$$

where C_t is the concentration of a single variable of time t and C_0 is initial concentration. Correlation curves were plotted between $\ln(C_t/C_0 - C_t)$ vs. time on each variable using Minitab 17 software. The parameters of k_{YN} and τ can be obtained using the nonlinear regressive method. The regression coefficient (r^2) and significance (p) were obtained from the analyses for interpretation.

2.6. FT-IR and XRD analysis for the filter media

FT-IR is a convenient tool to study the behavior of active adsorption or reaction sites in natural materials, such as soil (Cannane et al., 2013). Thus, the FT-IR analyses (Nicolet iS10 spectrometer) were performed on all filter media before and after filtration, using the KBr pellet method (Smidt et al., 2005). The average error for the analyses was less than $\pm 5\%$.

Filter materials were analyzed before and after the experiment using X-ray diffractometer (XRD; Tsao et al., 2013). Rigaku TTRAX III XRD instrument available in University of Sri Jayawardenepura, Sri Lanka was used. The powdered samples were analyzed using Cu K (alpha) radiation and wave length (1.54 Å) over 2 (theta) of 5–70° with a step size of 0.02°, which are relevant parameters for the clay mineral observations.

Table 1

Maximum permissible levels recommended by different water quality standards (GEDSRSL, 2008; WHO, 2011; SLSI, 2013; USEPA, 2017).

Parameter	CEA	SLS	USEPA	WHO
pH	6.0–8.5	6.5–8.5	6.5–8.5	6.5–8.5
BOD (mg/L)	30	–	–	–
COD (mg/L)	250	10	–	–
TSS (mg/L)	50	–	–	–
EC ($\mu\text{s}/\text{cm}$)	–	–	–	–
Phosphate (mg/L)	5	2	–	–
Nitrate (mg/L)	50	–	10	50
Fe (mg/L)	3.000	0.300	–	0.100
Cu (mg/L)	3.000	1.000	1.300	2.000
Ni (mg/L)	3.000	0.020	–	0.070
Cd (mg/L)	0.100	0.003	0.005	0.003
Zn (mg/L)	2.000	3.000	–	5.000
Pb (mg/L)	0.100	0.010	0.015	0.010
Mn (mg/L)	–	0.100	–	0.400

‘–’ denotes no recommended guideline.

2.7. Statistical analysis

Kruskal-Wallis and Mann-Whitney non-parametric analyses were used to study statistical significance using Minitab 14 software ($p > .05$; Simmons et al., 2001; Sakol and Konieczny, 2004). The Kruskal-Wallis test determined the significant differences in removal efficiencies of different variables among the five filters (A, B, C, D, and E). The Mann-Whitney test was performed for the variables that indicated a significant difference for a statistical comparison with the control (E).

3. Results and discussion

The chemical composition of the laterite and compost is mainly inherited from their parental materials (Nesbitt and Young, 1996; Zhang et al., 2017). Major and trace element concentrations of those materials provide useful clues on associated processes in leachate filtering. In addition, unstable physical conditions of the materials during the process play vital roles in the possible leaching of the elements (Carrillo-González et al., 2006). Therefore, with the material analyses, a regular composition study of both leachate and liquid filtrate provide clear evidence on the quality and efficiency of the proposed system.

3.1. Composition of laterite and compost

Table 2 presents the average concentrations of major oxides and trace elements for laterites. The laterites were characterized by water insoluble oxides, such as Fe_2O_3 , Al_2O_3 , SiO_2 , and TiO_2 . The SiO_2 was the most dominant oxide; however, higher values of Fe_2O_3 and Al_2O_3 were common in all samples. Concentrations of labile oxides, such as CaO , MgO , Na_2O , and K_2O , are diminished in laterite, and out of many analyzed trace elements, very few are detected (Table 2). In contrast, the lower concentrations of those elements act as indicators for the terminal chemical weathering property of the soil (Herath, 1973; Dahanayake, 1982).

The physical properties of the laterites are especially important (Table 3). The laterite shows oxidized conditions (average ORP, 98 mV) and acidic pH (average, 4.9; range 4.8–5.0) by the sediment ORP meter. Elution analyses of the laterites indicate lower conductivity due to the lower availability of soluble ions. The results further reveal lower values of TOC and organic matter. In addition, a moderate level of LOI signifies the slight enrichment of carbonates (average 16.23%).

Table 4 presents physicochemical properties of the compost. The compost pH was within the standard level (6.5–8.5), and the chemical composition of the compost denotes free of heavy metals. The carbon to nitrogen ratio varied between 10% and 25%, with a

Table 2

Major and trace elements in the laterite soils selected for the filter media.

	No 1	No 2	No 3	No 4	No 5	No 6	Avg	Min	Max	SD
<i>Mineral oxides (wt%)</i>										
Fe_2O_3	13.0	18.6	10.7	13.4	25.1	20.0	16.8	10.7	25.1	5.4
Al_2O_3	22.9	20.0	23.0	20.1	15.2	20.0	20.2	15.2	23.0	2.8
SiO_2	30.0	35.2	35.5	34.2	13.4	30.1	29.7	13.4	35.5	8.4
TiO_2	1.70	1.20	1.70	1.40	1.30	1.30	1.40	1.20	1.70	0.20
MnO	ND	ND	ND	ND	0.21	0.01	0.11	0.01	0.21	0.14
CaO	ND	0.01	0.01	0.02	0.45	0.06	0.11	0.01	0.40	0.19
MgO	ND	ND	ND	0.30	0.51	ND	0.40	0.30	0.51	0.15
Na_2O	ND									
K_2O	0.22	0.13	0.22	0.52	0.03	0.44	0.26	0.03	0.52	0.19
<i>Trace metals (ppm)</i>										
Zn	0.005	0.005	0.007	0.008	0.450	0.004	0.080	0.004	0.450	0.181
Ni	0.004	0.003	0.005	ND	0.301	0.005	0.064	0.003	0.301	0.133
Cu	0.004	0.004	0.004	0.002	0.008	0.007	0.005	0.002	0.008	0.002
Co	0.014	0.045	0.023	0.017	0.053	0.034	0.031	0.014	0.053	0.016
Cd	ND									
Pb	0.001	0.002	0.001	0.012	0.032	0.003	0.009	0.001	0.032	0.012

ND: not detected, SD: standard deviation.

Table 3
Physico-chemical properties of the laterite soils selected for the filter media.

Parameter	No 1	No 2	No 3	No 4	No 5	No 6	Avg	Min	Max	SD
Moisture Content (%)	19.3	29.4	18.7	18.2	14.9	16.8	19.6	14.9	29.4	5.1
pH	4.8	4.9	4.9	5.1	5.0	5.0	4.9	4.8	5.1	0.1
ORP (mV)	97	100	102	95	105	91	98	91	105	5
EC ($\mu\text{S}/\text{cm}$)	31.4	31.5	31.4	31.3	31.4	30.5	31.3	30.5	31.5	0.4
TOC (%)	9.3	8.5	7.5	9.6	8.9	8.8	8.8	7.5	9.6	0.7
OM (%)	0.83	0.76	0.81	0.71	0.75	0.74	0.77	0.71	0.83	0.05
LOI (%)	16.95	15.98	15.08	16.78	16.33	16.24	16.23	15.08	16.95	0.67

ORP: oxidation reduction potential, EC: electrical conductivity, TOC: total organic carbon, OM: organic matter, LOI: loss on ignition, moisture content: $(W_{\text{wet}} - W_{\text{dry}})/W_{\text{wet}} \times 100$.

Table 4
Average main composition characteristics of the compost samples used for the filter media.

Parameter	Concentration
Moisture content (%)	12.07
Organic Carbon (%)	12.30
Nitrogen	1.20
Phosphorus as P_2O_5	0.96
Potassium as K_2O	1.07
pH	8.45
C/N ratio	10.25
Sand (%)	11.93
Heavy metals	ND

ND: not detected. Trace Metals: Fe, Mn, Cu, Ni, Cd, Zn, and Pb.

high level (average 12.3%) of organic carbon content. This shows a greater level of humus, which provides more surface area on the filter media (Gibert et al., 2003). The composition of compost used in this experiment complied with the compost production standards given by Central Environmental Authority in Sri Lanka (CEA).

3.2. Physicochemical characteristics of raw leachate

Table 5 presents the statistical summary for physical and chemical properties of raw leachates. The landfill area where leachate was collected received over 2500 mm of mean annual rainfall. However, the results indicate a slightly basic pH value for leachate (range: 7.4–8.8), which reveals an initial methanogenic phase (Regadío et al., 2015). Major landfill sites under the same climatic conditions in Sri Lanka show methanogenic conditions (Wijesekara et al., 2014); however, there is a slight acidic pH trend that reflects an acetogenic nature in some studies (Perera et al., 2014). Most of the parameters, such as BOD, COD, phosphate, nitrate, TSS, EC, and

Table 5
Statistical summary of the raw leachate samples (before dilution) collected during the period of the experiment (Number of samples = 31).

Element	Avg	Min	Max	SD
pH	8.0	7.4	8.8	0.4
BOD (mg/L)	1491	840	2200	416
COD (mg/L)	2434	1344	4000	737
EC ($\mu\text{S}/\text{cm}$)	13.59	10.59	16.5	1.41
TSS (mg/L)	6510	1020	24,750	7097
Phosphate (mg/L)	203	92	290	73
Nitrate (mg/L)	111	79	182	31
Fe (mg/L)	11.88	7.58	32.12	6.88
Cu (mg/L)	1.32	0.28	2.20	0.79
Ni (mg/L)	0.94	0.50	1.20	0.18
Cd (mg/L)	0.26	0.11	0.58	0.18
Zn (mg/L)	0.79	0.47	1.47	0.37
Pb (mg/L)	1.11	0.73	1.60	0.32
Mn (mg/L)	5.51	0.74	12.99	3.49

Fe, reveal very high average values (Table 5). The standard deviations for BOD, COD, and TSS are quite high, probably due to the influence of local climatic variation, waste type and particle size, degree of compaction and age of the waste during leachate sampling (Chen, 1996; Kjeldsen et al., 2002). It has been observed that most of the landfill leachates of Sri Lanka have very high BOD, COD and Fe values (Wijesekara et al., 2014; Perera et al., 2014). The BOD/COD ratio of the leachate is approximately 0.5, which indicates that leachate is readily biodegradable (Banar et al., 2009; Regadío et al., 2015). Selected trace metals, such as Cu, Ni, Cd, Zn, Pb, and Mn, are at slightly higher concentrations in raw leachate (Table 5).

3.3. Physicochemical characteristics of the liquid filtrate

Table 6 depicts physical and chemical properties of the liquid filtrates. With the progress of the process, the pH value for all liquid filtrates was within the recommended level (range: 6.0–8.4), which may be buffered by the soluble carbonates in the soil. Most chemical parameters (Fe, Cu, Ni, Cd, Zn, Pb, Mn, nitrate, and phosphate) show a remarkable improvement and indicate lower values than raw leachate. The average values for TSS display a higher range in the liquid filtrate (range: 140–4320 mg/L). However, this indicates pronounced improvements concerning raw leachates. The average EC in the liquid filtrate does not delineate the pronounced improvement.

3.4. Comparison with water quality standard

The average concentrations in the liquid filtrates were compared with the standard water quality guidelines to ascertain the performances of five filters (Table 1). Values of pH in the filtrates comply with the recommended standards. The slightly acidic nature of the leachate has been improved during filtering, which may

Table 6

Statistical summary of the liquid filtrate samples collected during the period experiment (Number of samples = 155).

Parameter		Avg	Min	Max	SD	Parameter		Avg	Min	Max	SD
pH	A	7.2	6.4	8.0	0.5	Fe (mg/L)	A	0.31	0.02	0.66	0.13
	B	7.1	6.1	8.0	0.5		B	0.29	0.03	0.64	0.12
	C	7.3	6.8	8.1	0.4		C	0.28	0.02	0.47	0.09
	D	6.9	6.0	8.4	0.6		D	0.26	0.03	0.47	0.09
	E	7.3	6.2	8.4	0.6		E	0.36	0.04	1.83	0.34
BOD (mg/L)	A	213	38	950	260	Cu (mg/L)	A	0.31	0.10	0.56	0.16
	B	167	8	978	260		B	0.31	0.11	0.57	0.16
	C	162	22	994	252		C	0.32	0.11	0.53	0.15
	D	136	15	824	214		D	0.32	0.11	0.53	0.14
	E	239	15	882	244		E	0.30	0.11	0.50	0.15
COD (mg/L)	A	703	122	3116	589	Ni (mg/L)	A	0.29	0.13	0.39	0.07
	B	567	24	3325	886		B	0.29	0.08	0.39	0.07
	C	545	70	3380	861		C	0.32	0.12	0.43	0.07
	D	461	47	2835	740		D	0.34	0.13	0.47	0.08
	E	788	50	2911	805		E	0.24	0.14	0.47	0.08
EC (μ S/cm)	A	11.00	2.43	13.34	2.73	Cd (mg/L)	A	0.05	0.00	0.14	0.05
	B	11.01	2.38	13.48	2.77		B	0.05	0.00	0.15	0.05
	C	11.34	3.00	14.06	2.66		C	0.06	0.00	0.15	0.05
	D	11.53	3.14	14.21	2.63		D	0.06	0.01	0.14	0.04
	E	1.96	2.34	13.51	2.72		E	0.04	0.00	0.13	0.04
TSS (mg/L)	A	1543	140	4320	1063	Zn (mg/L)	A	0.19	0.02	0.48	0.11
	B	1573	264	3100	931		B	0.18	0.02	0.46	0.11
	C	1540	170	2875	870		C	0.17	0.02	0.49	0.11
	D	1615	348	3140	967		D	0.12	0.02	0.21	0.04
	E	513	194	3740	944		E	0.15	0.02	0.35	0.07
Phosphate (mg/L)	A	3.0	0.7	17.9	3.0	Pb (mg/L)	A	0.39	0.12	0.61	0.11
	B	2.6	0.3	15.9	2.8		B	0.41	0.13	0.71	0.14
	C	2.1	0.4	4.5	0.8		C	0.46	0.12	0.72	0.16
	D	1.8	0.4	3.0	0.7		D	0.48	0.16	0.74	0.15
	E	4.5	0.6	45.4	8.8		E	0.34	0.09	0.58	0.12
Nitrate (mg/L)	A	8.9	5.8	31.0	4.4	Mn (mg/L)	A	1.04	0.05	1.75	0.56
	B	8.6	4.2	21.0	2.9		B	0.96	0.05	1.64	0.51
	C	7.7	4.1	9.9	1.5		C	0.93	0.05	1.61	0.53
	D	5.8	1.8	11.6	2.0		D	0.68	0.05	1.36	0.40
	E	12.4	2.2	40.4	5.9		E	1.22	0.05	2.08	0.60

be due to pH buffering by soil carbonates (Regadío et al., 2013; Villen-Guzman, et al., 2015). In addition, BOD, COD, and phosphate levels in the filtrates followed the CEA and SLS standards, especially after 25 days. Nitrate was within the CEA and USEPA guidelines. The average Cd and Cu in the liquid filtrates fit with all standards. The Fe and Zn of the liquid filtrates were compatible up to the drinking water guidelines. The liquid filtrates did not comply with the CEA guidelines, due to very high values of TSS in leachate. This can be improved by the increasing thickness of given sand layers. The average level of Ni is only comparable to the CEA standard, whereas Pb and Mn in the liquid filtrates ignored the standard guidelines. This can be improved by adding a raw laterite layer to the filters.

3.5. Characteristics of the filtering process

Natural clay materials, such as laterite, have very low permeability despite a high porosity. Their high specific surface areas allow for strong physical and chemical interactions (Michael et al., 2002). Even highly permeable sandy earth materials, such as sandstone and arkosic materials, can be used as liners for landfills by adding clay to them (Ruiz et al., 2012). Cuevas et al. (2012) observed that the clay mineral rich layer around the landfill has an effective filtering ability of leachate. This controversial property of clay was highly present in laterite soils, due to the availability of terminal clay minerals from the chemical weathering (Madhukar et al., 2012).

BOD, COD, phosphate, and nitrate filtering from the leachate are highly effective. Several actions are involved with the removal of

those parameters, i.e., ion exchange, surface adsorption, chemical precipitation, and biological processes (Gálvez et al., 2005; Abbas et al., 2009; Chen et al., 2007b). It was observed that the removal efficiencies of BOD, COD, phosphate, and nitrate were unsteady during the first 20 days (Fig. 3). Subsequently, biodegradation has increased the stability of the efficiencies. The reason may be the alterations in the microenvironment of the filter media caused by the microbial population. They required an initial time period to adapt to new conditions (Rose et al., 2012). Removal efficiencies after 20 days did not show a significant difference ($p > .05$) among the filters, whereas the control produced the lowest value (Fig. 3). After stabilization of the filters (A, B, C, and D), the removals reached higher levels for BOD (>90%), COD (>85%), and phosphate (>90%). Further, nitrate removal was gradually increased (75–95%), which may be due to denitrification by excess carbon added to the filter media (Pattnaik et al., 2007). Removal efficiencies for TSS show wide ranges of fluctuation (Fig. 3e; 40–90%). Conversely, EC displayed a diverse efficiency range during the experiment period (Fig. 3f; 0–80%). This may be due to the leaching of labile ions, such as Ca and Mg ions, into the liquid filtrate by the ion exchange process and pH changes in laterite (Regadío et al., 2015; Jun et al., 2007).

Metal filtering efficiencies tend to increase at higher pH (Benedetti et al., 1995). The time series plot for Fe, Mn, Cu, Ni, Cd, Zn, and Pb is provided in Fig. 4, which shows greater removal efficiency for Fe during the 100 days (90–100%). This may be due to the formation of Fe-oxides and Fe-sulfides within the system and adsorption on to the clay (Madhukar et al., 2012). The Eh-pH relationship of Fe shows appropriate conditions for the formation

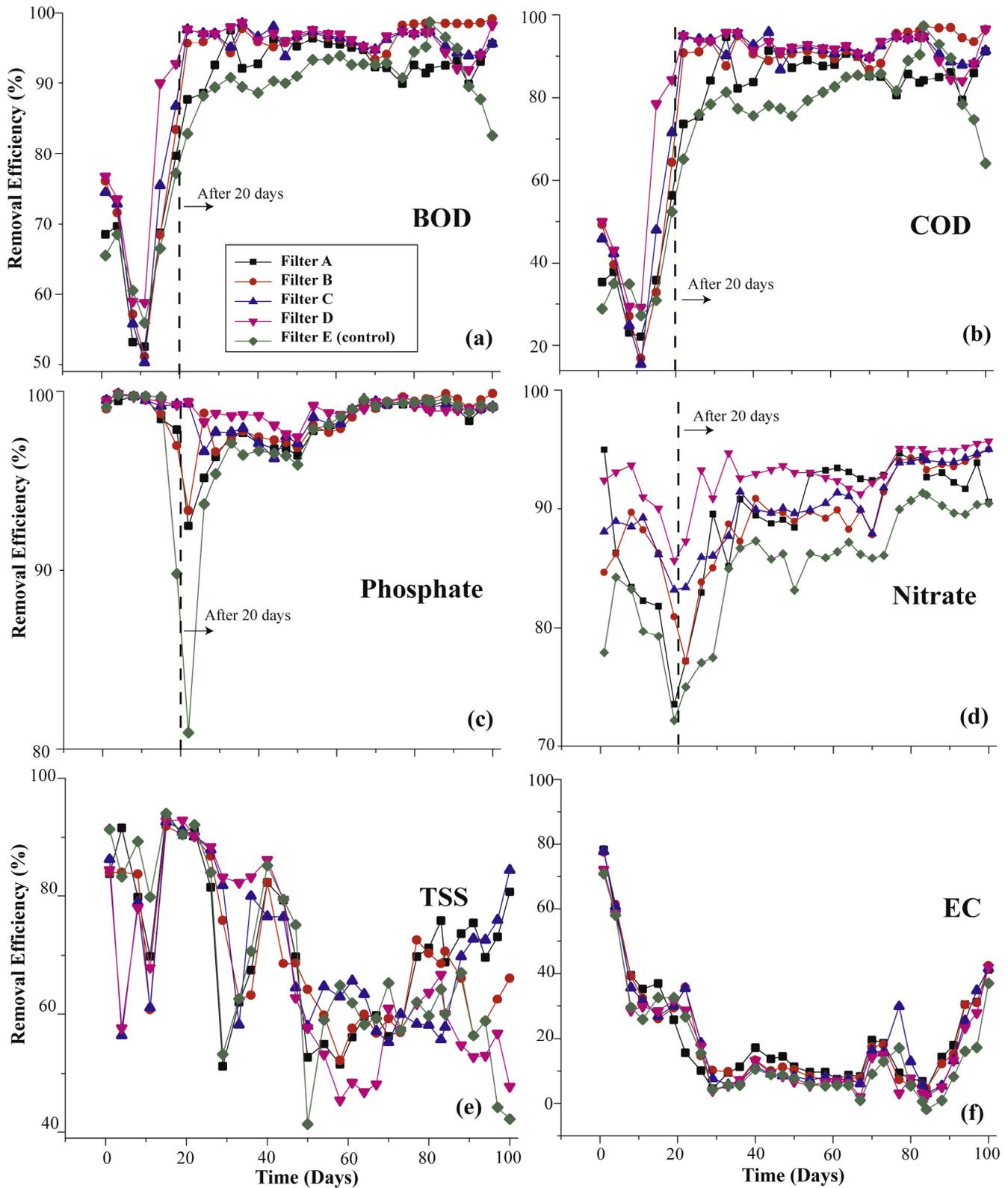


Fig. 3. Removal efficiencies of BOD, COD, phosphate, nitrate, TSS, and EC with reference to the original concentration.

of oxides and sulfide, and the conditions are developed with the usage. According to phase relationships, Fe ions only form under very acidic pH and a wider range of ORP (Takeno, 2005; Regadío et al., 2013). However, with time, a basic condition develops in the filter system, which supports effective filtering by precipitating oxides and sulfide.

The removal efficiency of Mn shows a diverse variation (30–90%). It may be due to the complex behavior of Mn. Mn has several oxidation states that involve several physical, chemical and biological reactions (Bradl, 2004). Cation exchange capacity, soil organic matter and amorphous iron oxides in the soil and compost affect Mn adsorption (Harlina et al., 2016). In contrast, Fe and Ca are sub-

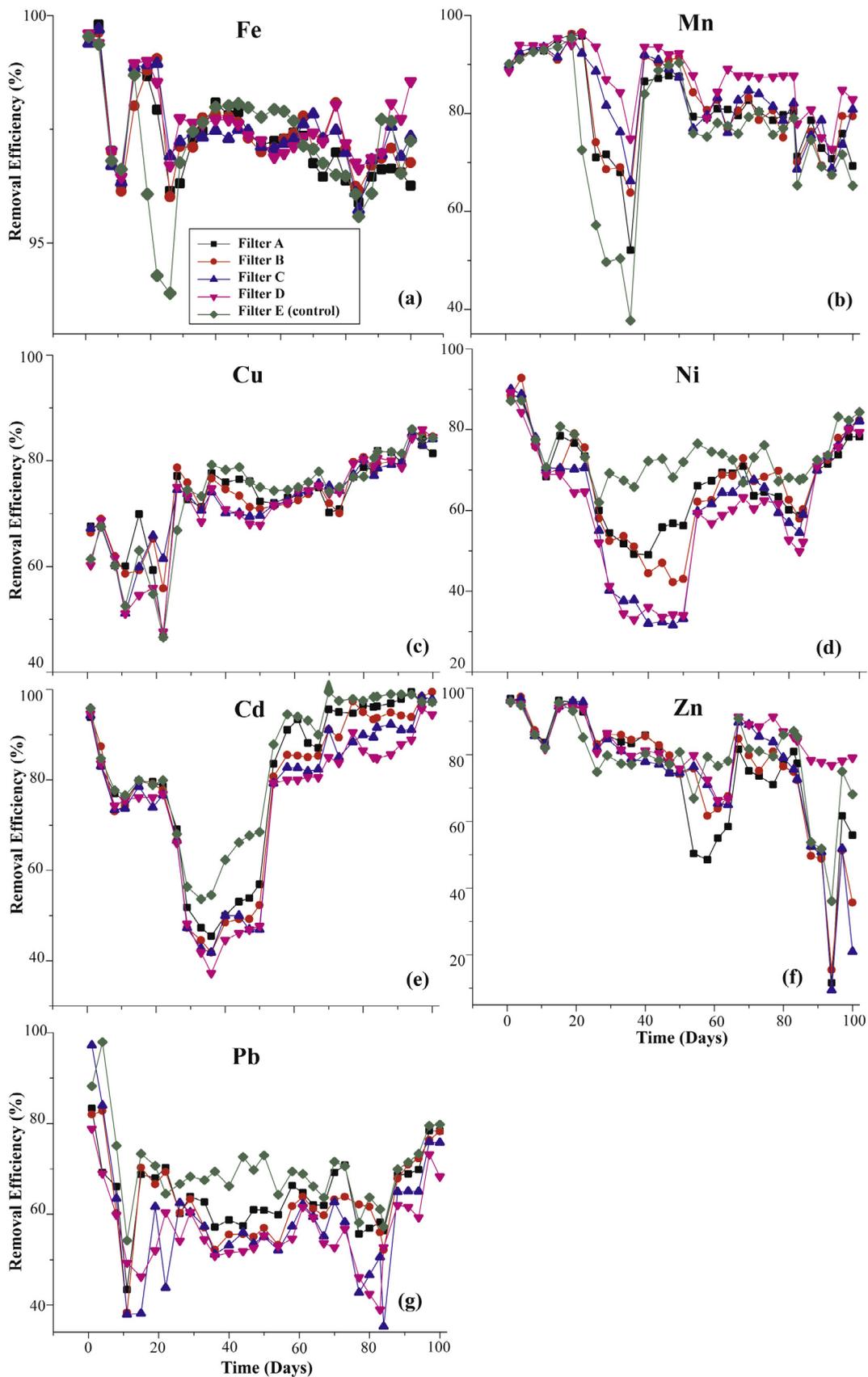


Fig. 4. Removal efficiencies of the selected heavy metals (Fe, Mn, Cu, Ni, Cd, Zn, and Pb) with respect to the original concentrations.

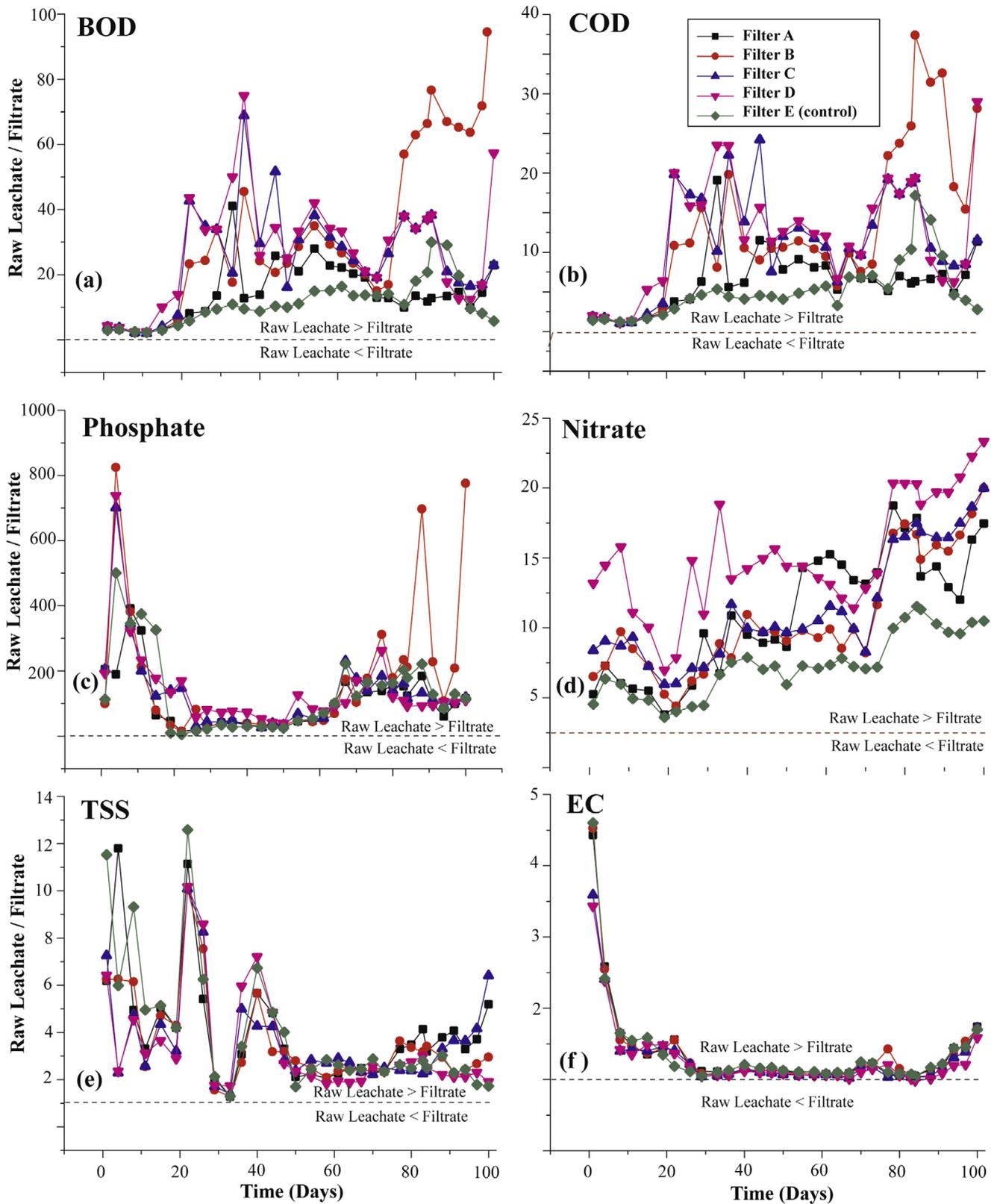


Fig. 5. Normalization of the leachate concentrations on BOD, COD, phosphate, nitrate, TSS, and EC against liquid filtrate concentrations.

stitutes for Mn, and they may influence the removal of Mn (Bradl, 2004).

Cu indicates an increasing trend of removal efficiency during the 100 days (45–85%). There is a high affinity of soil organic matter (OM) for Cu (McGrath et al., 1988). The most important substi-

tutes for Cu removal are Fe, MnO, sulfides, OM, and carbonates (Couillard, 1994; Bradl, 2004).

Adsorption is the most common method to remove Ni, Cd, Zn, and Pb from leachate (Fig. 4). The adsorption process is controlled by cation exchange capacity, OM content, clay content, and iron

oxide (Bradl, 2004; Mellis et al., 2004). Greater fluctuations of removal efficiencies are common in Ni (30–93%), Cd (37–98%), Zn (15–98%), and Pb (35–98%). This may depend on the concentration

variation of those elements in raw leachate, particularly a higher pH influence on the electrostatic interaction between clay and Cd ions. Conversely, Cd dissolves under lower pH and negative redox

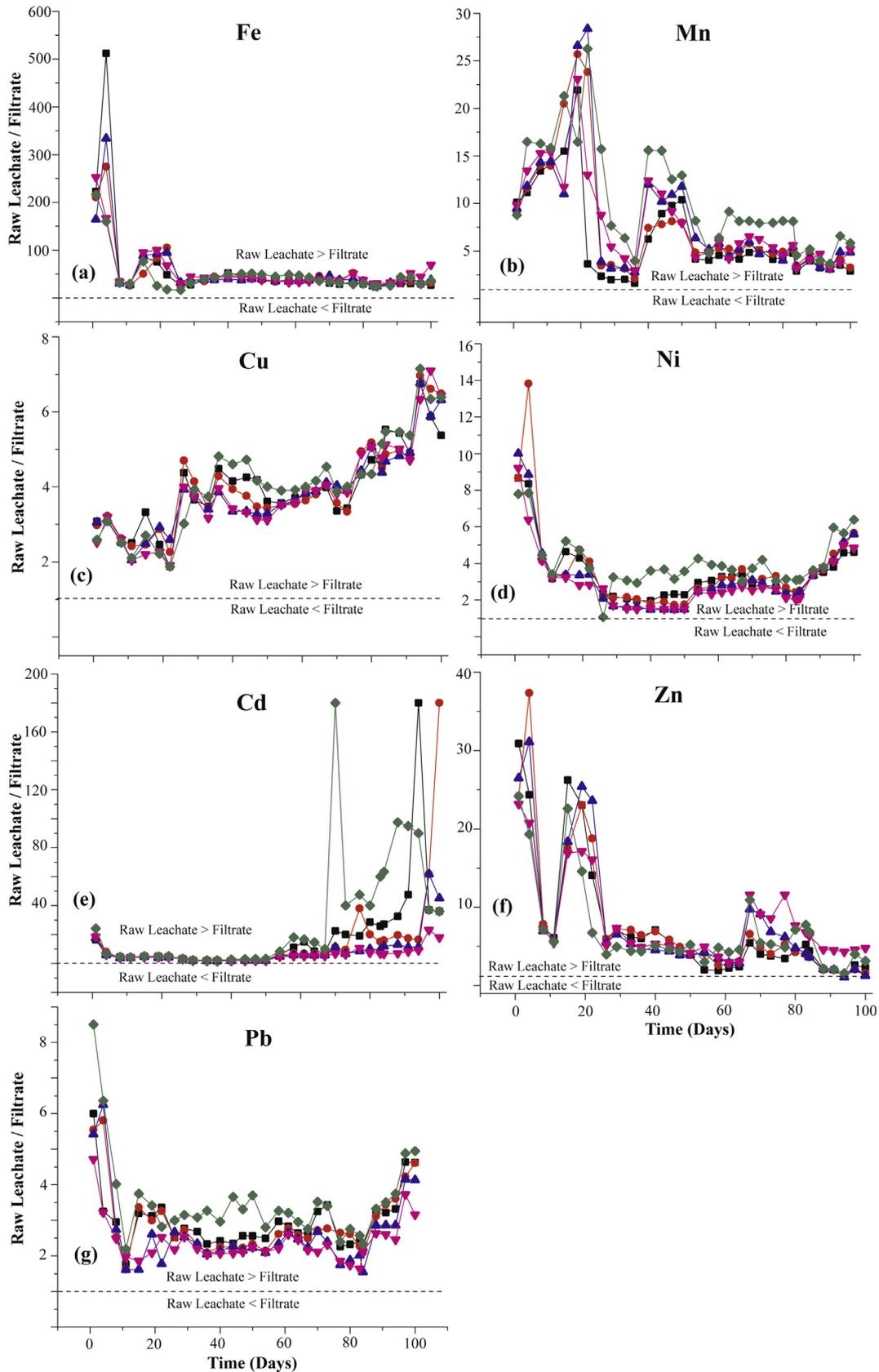


Fig. 6. Normalization of the leachate concentrations on selected heavy metals (Fe, Mn, Cu, Ni, Cd, Zn, and Pb) against liquid filtrate concentrations.

potential (Elliott et al., 1986). Zinc (Zn) forms complexes with Cl^- , PO_4^{3-} , NO_3^- , and SO_4^{2-} to filter from raw leachate. Pb is removed by forming stable chelates or complexes and precipitation, and the presence of Mn and Fe oxides may play a predominant role on Pb adsorption (Bradl, 2004).

3.6. Quality of liquid filtrate against raw leachate

Normalization ($[\text{raw leachate}]/[\text{liquid filtrate}]$) of all those parameters clearly demarcate a greater level of removals (Fig. 5). The concentration depletion on BOD (1–98 times) and COD (1–38 times) shows strong removal against raw leachate (Fig. 5a and b). The tendency of phosphate removal is 810 times greater than the maximum (Fig. 5c). Nitrate shows an increasing trend on removal performance, with a range of 2–24 times with respect to the raw leachate (Fig. 5d). However, normalization of TSS (1–5 times) and EC (1–13 times) indicate relatively lower levels of the raw leachate (Fig. 5e and f).

Fig. 6 presents the normalization plots for Fe, Mn, Cu, Ni, Cd, Zn, and Pb. After approximately 20 days, the normalization of Fe was stabilized (25–55 times improvements), and Fe removal continued under a higher rate, which may be caused by adsorption, complexation, ion exchange, and precipitation (Bradl, 2004). The normalization patterns on Mn (2–28 times), Ni (2–14 times), and Zn (2–35 times) reveal almost similar fluctuation ranges. A wide range of normalization values is expressed for Cd (2–180 times), but very low values are indicated in the 25–45-day period. Cu (2–7 times) and Pb (2–8 times) show the lowest normalization values due to lower leachate concentrations.

In general, trace metal contaminations (Cu, Ni, Cd, Zn, and Pb) of surface water around the dumping site are moderate (Mandakini et al., 2016). However, compared to surface water, groundwater can enrich them, due to prevailing anoxic conditions and acidic pH (Takeno, 2005). Conversely, BOD and COD in surface water significantly exceeded the tolerance limits (Mandakini et al., 2016). Therefore, possible implementation of the proposed filter provides a significant advantage to the surrounding water environment.

3.7. Performance of the filter media

The FT-IR analysis is worthwhile to investigate possible changes in filter materials. The results proved that more vacant sites of

sorption are present even after hundred days (Fig. 7). This indicates the filter media can be further used for filtering. A higher transmission in the $3625\text{--}3695\text{ cm}^{-1}$ region of FT-IR spectra on filters A, B, C, D, and E is dominant. This may reveal the sorption on cations, which are bonded to hydroxyl groups, such as phenol, alcohol, and organic acid, in the filter materials (Dupuy and Douay, 2001; Scholtzová et al., 2003; Johnston et al., 2008).

Peaks at approximately 1635 cm^{-1} related to the variation of the O–H bond of adsorbed water suggest the formation of PbOH^+ (Fonseca et al., 2009; Vithanage et al., 2012). The wave numbers $1508\text{--}1560\text{ cm}^{-1}$ and $1408\text{--}1479\text{ cm}^{-1}$ mark the carbonate region, which totally or partially disappeared (Fig. 7), likely caused by the reaction of metals with the carbonates in laterite (Tinti et al., 2015; Lim and Lee, 2015). Peaks from 1020 to 1055 cm^{-1} are related to the breakdown of Si–O–Si bonds that play a significant role in heavy metals sorption, such as that of lead (Fonseca et al., 2009; Lim and Lee, 2015). The peaks of wavelengths between 777 cm^{-1} and 794 cm^{-1} are related to the Fe–OH bonds, and peaks at 912 cm^{-1} are related to the Al–OH bonds (Mozgawa et al., 2009), due to the absorbance of water into Fe and Al in the laterite. Furthermore, the peak of 529 cm^{-1} shows empirical evidence of the deformation of clay minerals by filtration (Alvarez-Puebla et al., 2004; Tyagi et al., 2006).

XRD peak patterns for the filter materials are given in Fig. 8. Secondary minerals gibbsite, hematite, goethite, kaolinite and primary mineral quartz are the main sources present in the mixed filter media (Fig. 8a). It has been recognized that the available minerals in the filter media are identical to the bauxite (Neumann et al., 2014). Trace metal sorptions from those minerals are commonly developed with the availability of compost (Meiera et al., 1999; Bradl, 2004). In contrast, after the filtering process XRD peaks show significant variation compared to the initial peaks (Fig. 8). This may indicate sorption during the leachate filtering has changed the conditions of available minerals. Primary mineral quartz is not useful for the sorption process. However iron coated quartz is available within the laterite and XRD results reveal those quartz has involved significantly for the sorption (Fig. 8b; Benjamin et al., 1996).

According to the results of the Yoon–Nelson regression-based kinetic model, different significances for the analyzed parameters can be seen Fe, Zn, Mn and nitrate have positive regressions with the time for all the filters (Table 7), which indicate $\ln(C_t/C_0 - C_t)$

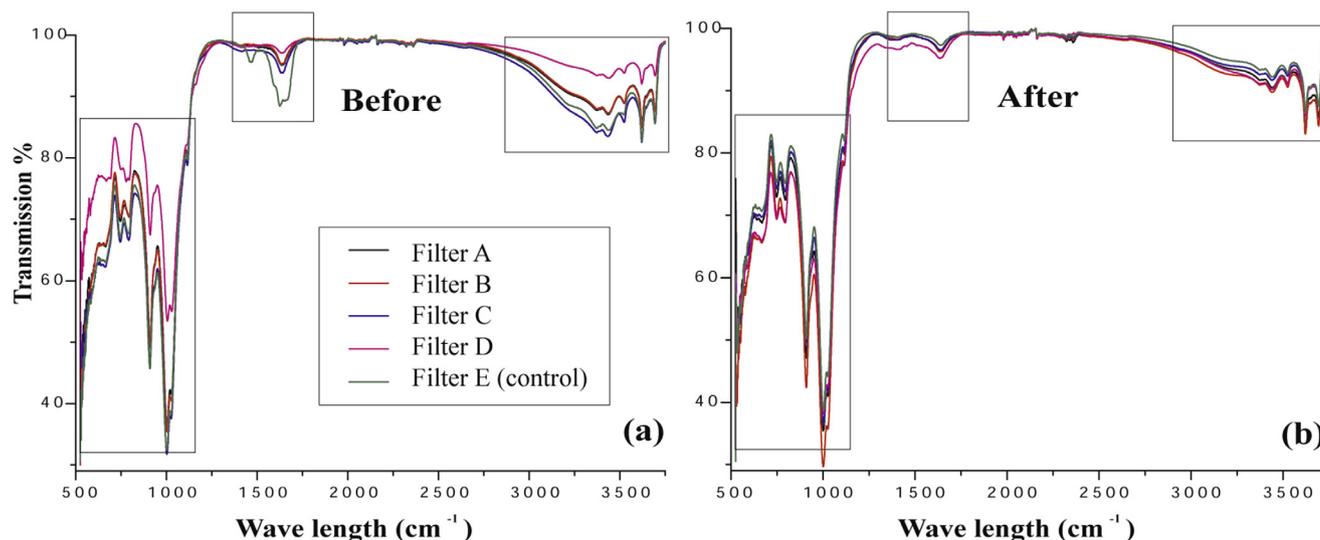


Fig. 7. FT-IR spectra of the filter materials (a) prior to commencement of the experiment; (b) after 100 days of filtering. Black color rectangles denote the major wavelengths used for sorption processes.

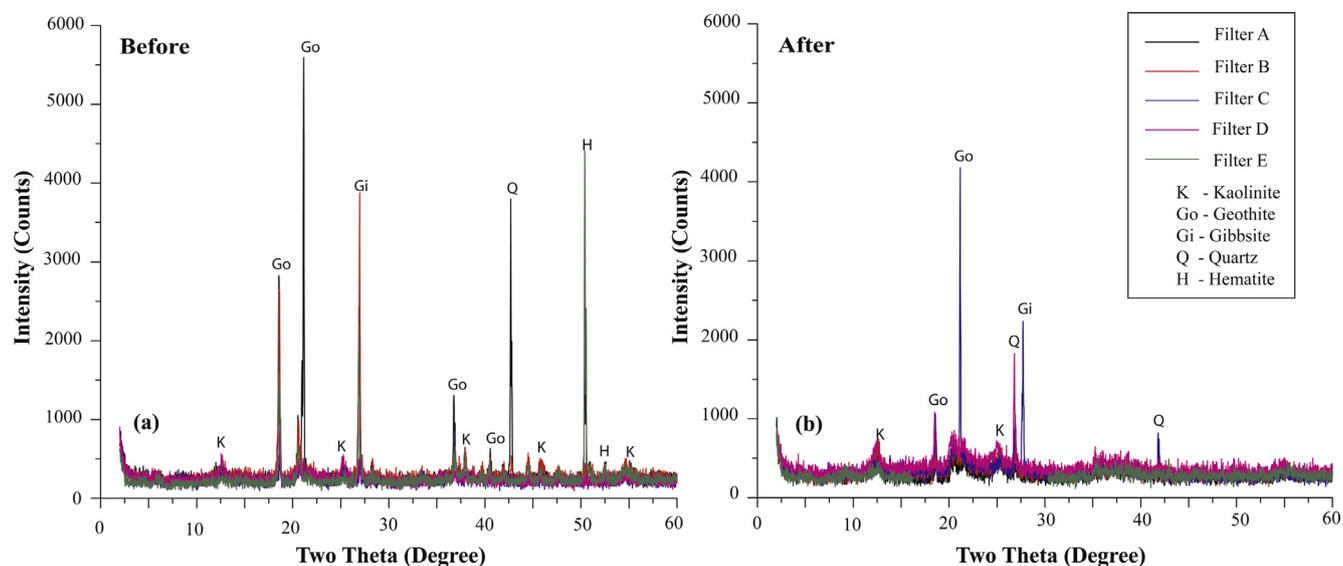


Fig. 8. XRD study of the filter materials (a) prior to commencement of the experiment; (b) after 100 days of the filtering.

Table 7

Regression coefficient values for the Yoon–Nelson kinetic model.

	A	B	C	D	E
Fe	0.60	0.56	0.56	0.47	0.36
Zn	0.78	0.83	0.75	0.48	0.62
Mn	0.56	0.56	0.70	0.67	0.40
Cu	−0.76	−0.80	−0.87	−0.86	−0.83
Cd	−0.70	−0.63	−0.58	−0.46	−0.74
BOD	−0.62	−0.83	−0.54	−0.48	−0.74
COD	−0.66	−0.83	−0.58	−0.58	−0.75
Nitrate	0.45	0.62	0.79	0.80	0.63

r^2 : regression coefficients, BOD: biological oxygen demand, COD: chemical oxygen demand.

are increased with the filtering time. This reveals that the sorption kinetics of the filter media decreases with the increasing time period of the filtering (Han et al., 2009; Ghribi and Chlendi, 2011). Conversely, Cu, Cd, BOD and COD have negative correlations and reveal that the adsorption abilities of filter media were increased with the filter usage (Table 7). The kinetic model did not prove a clear relationship between the impact of level of compost and the sorption process for all of the parameters.

3.8. Statistical significance of the filtering system

The Kruskal–Wallis test was performed on the analyzed variables to compare removal ability from laterite soil with the increasing level of compost (Kang et al., 2017). The results show that the removal efficiencies of EC ($p = .207$), TSS ($p = .674$), phosphate ($p = .564$), Fe ($p = .411$), Cu ($p = .780$), and Zn ($p = .345$) have no significant differences among the filters ($p > .05$). This showed that there is less of an impact on filtering efficiencies by adding compost to those variables. This may be due to the increase in pH and OM mineralization under a lower level of phosphate-amended compost, which is responsible for the reduction in exchangeable and bioavailable Cu and Zn (Lu et al., 2014). In addition, Fe complexation is taking place in Al-bearing substrates, such as soil; hence, there is less impact from the level of OM on Fe removal efficiency (Starcher et al., 2017).

Conversely, the Kruskal–Wallis test showed significant differences in removal efficiencies ($p > .05$) of BOD ($p = .000$), COD ($p = .000$), nitrate ($p = .000$), Ni ($p = .044$), Cd ($p = .044$), Pb ($p = .000$), and Mn ($p = .003$). Compost contributed to altering the filtering

ability of laterite, which may be due to a strong sorption capacity of organic matter (Kwiatkowska-Malina, 2017). Therefore, the Mann–Whitney test was performed on those variables to identify the composition of compost required to obtain a significant change of removal efficiencies (Yue and Wang, 2002). The results indicated that a minimum of 20% of compost was needed for a significant development of removal efficiencies on BOD and COD. Furthermore, 10% compost caused a significant increase in nitrate filtering, and the additional amount shows an added advantage. However, Ni and Cd removal efficiencies were depleted only after 30% of compost was added. However, Pb and Mn show a depletion of filtering efficiencies to a certain extent by the addition of 10% compost.

Results of the statistical analysis were used to select the best filter composition. Selection is mainly based on positive significant values given for the studied parameters. The filter medium with 20% compost with 80% laterite by weight ratio indicates positive significant differences for the most of the parameters with respect to the other filters. Therefore, it is evident that 20% of compost by weight is the most suitable condition to achieve higher filtering efficiencies.

4. Conclusions

In this experiment, laterite, as a major sorption and compost material for biological processes, was successfully used for leachate filtering. The leachate uses for the filter reflect the methanogenic stage with strong biodegradation. The higher removal efficiencies of BOD, COD, phosphate and nitrate showed that the

biodegradation was activated by the added compost. The removal of nitrate steadily increased with time, which may show that denitrification occurred due to the excess carbon enriched from the leachate. Moderate to high removal efficiencies of TSS may indicate disaggregation of filter materials by biological and physical processes. The unsteady nature of EC would conclude iron exchange and dissolution of soil carbonate. The pH values show buffering by soluble carbonates in the soil. An extremely high removal efficiency was reported for Fe, and wide ranges of efficiency trends were reported for Mn, Cu, Ni, Cd, Zn, and Pb, showing different metal sorption processes. The normalization values further showed marked improvements during filtering. Statistically, it can be concluded that 20% compost by weight is the optimum condition for the filter. The FT-IR models on filter materials showed multiple sorptions during the filtering.

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