PREDICTING USEFUL LIFE OF COCOPEAT IN A FILTER BED TREATING WASTEWATER WITH HEAVY METALS USING HYDRUS-1D

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Abstract

The effectiveness of cocopeat as an adsorbent for the treatment of wastewater containing heavy metals was reported in previous studies. In this study, cocopeat was used as an adsorbent in a filter bed system treating wastewater from a small scale gold mining (SSGM) ball mill facility. A total of 6,000 L of actual SSGM wastewater collected from a ball mill facility in Paracale, Camarines Norte were used in the experimental runs. The filter bed was evaluated by determining its heavy metal removal efficiencies for 50 days at a flow rate of 40 L/hr. After the experimental runs, HYDRUS-1D was used to simulate the transport of lead (Pb) in the filter bed and predict the remaining useful life of cocopeat as a heavy metal adsorbent. Lead was selected for the solute transport modeling in HYDRUS-1D since Pb was the highest concentration in the wastewater and also exceeded the government effluent limit. Measured data from the experimental runs and water flow parameters of cocopeat were used as input values in the simulation with varying cocopeat thickness and initial concentration flux. Results showed that by increasing the thickness of the cocopeat layer, the useful life of cocopeat in the filter bed was extended. By using the actual concentration of Pb (0.0933 mg/L) in the wastewater, HYDRUS-1D was able to simulate Pb transport in the filter and predict that the useful life of cocopeat 50 cm thick treating an actual SSGM wastewater was 2.74 years.

Keywords: Cocopeat, Filter bed, Heavy metals, HYDRUS-1D, SSGM wastewater

Nomenclature

ρ_b	bulk density (g/cm^3)
n	porosity (%)
K_s	saturated hydraulic conductivity (cm/hr)
θ	volumetric water content
h	pressure head (cm)
v_l	volume of liquid (cm^3)
m_l	mass of liquid (g)
ρ_b	bulk density (g/cm^3)
V	total volume of cocopeat (cm^3)
S_e	effective saturation
α, n, and m	fitting parameters
θ_s	saturated volumetric water content
θ_r	residual volumetric water content

Introduction

Small scale gold miners in the Philippines and other developing countries use amalgamation process to recover gold from primary ores with the use of mercury (Hg) [1,2]. This method is commonly used in gold-rush areas by small scale miners due to its simplicity in application and relatively low investment [1,3,4]. However, the use of amalgamation process produces wastewater with elevated amount of Hg and other metals as well as high concentration of suspended solids that exceed the government effluent limits [5]. Other regulated heavy metals such as arsenic (As), cadmium (Cd) and lead (Pb) present in wastewaters from ball mill facilities in the Philippines can be traced from the chemical composition of mined ores and rocks in the mineralized areas [6]. Most of this wastewater is being disposed of to the receiving water bodies with minimal treatment or oftentimes untreated. Mercury in the wastewater, for example, can travel downstream and dispersed to a wide area of bodies of water where it can transform into methylmercury (MeHg) in the sediment of lakes and rivers. Methylmercury can be transferred and concentrated in fish which may be eaten by the people living nearby mining areas. If the workers exposed to the heavy metals in the wastewater, they have risk not only themselves to the contamination, but also their families, neighbors, and the community living near the worksites. Hence, removal of such contaminants is important before the discharge to the receiving surface waters.

In this study, adsorption was used to remove heavy metals from the wastewater because it is economical and promotes recycling and reuse of the adsorbed heavy metals [7,8]. It has frequently been studied and widely applied for the treatment of various heavy metal-contaminated wastewaters [9]. One of the emerging adsorbents used for heavy metal adsorption is cocopeat or coir pith. It is a short spongy fiber and dust that is a by-product in the extraction of coir fiber from coconut husks [10]. This material is cheap and abundant in the Philippines, and its use is limited only to agricultural purposes.

In the previous studies, cocopeat was used as an adsorbent for removing heavy metals from a small scale gold mining (SSGM) wastewater in batch and column experiments as well as in field scale filter bed system [11]. Characterization studies reported that cocopeat consists mainly of lignin, cellulose, hemicellulose, and some pectin and extractives (mainly fat, fatty acids, fatty alcohols, phenols, terpenes, steroids, resin acids, rosin and waxes) [12,13]. Previous adsorption studies suggest that heavy metals were adsorbed mainly to carboxylic (primarily present in pectin and hemicellulose but also extractives and lignin), phenolic (lignin and extractives) and to some extent hydroxylic (cellulose, hemicellulose, lignin, extractives, and pectin) and carbonyl groups (lignin) [12,14]. Laboratory scale adsorption studies were carried out to determine the efficiency of cocopeat in adsorbing zinc (Zn) and Pb [12,15] from ideal aqueous solutions but very few studies performed using actual wastewater samples.

In the current study, a field scale filter bed system, composed of sedimentation tank and filter bed with cocopeat as adsorbent, was developed and tested to remove heavy metals from the actual SSGM wastewater. After the experimental runs, the transport of heavy metal in the adsorbent column is simulated by numerical computer model HYDRUS-1D - a public domain Windows-based modeling environment for analysis of water flow and one-dimensional solute transport in variably saturated porous media and was first introduced in 1998 by Simunek, et.al [16]. The purpose of the study is to simulate the heavy metal transport in the cocopeat column of the filter bed, to predict adsorption capability of cocopeat over time and to determine the saturation point where the maximum adsorption of heavy metal onto the surface of cocopeat will be achieved, using HYDRUS-1D.

Materials and Methodology

SSGM Wastewater

A total of 6,000 L of wastewater sample was gathered from the sedimentation tank of an active ball mill facility in SSGM area in Paracale, Camarines Norte, 320 km south of Manila. The water sample was transported to Quezon City where the pilot filter bed system was installed. Physico-chemical characteristics, such as pH and turbidity, were measured on site using Horiba Multi Water Quality Checker U-5000G (Japan) while total suspended solids (TSS) were measured in the laboratory using the gravimetric method after drying the sample at 105°C. Heavy metal concentrations were analyzed using hydride generation-AAS for As; flame AAS for Ba, Cd and Pb; and cold vapor AAS for Hg following the method suggested by APHA-AWWA Standard Method for the Examination of Water and Wastewater [17]. The characteristics of SSGM wastewater used in the experiment are presented in Table 1.

Table 1. Average Heavy Metal Concentrations and Physico-Chemical Characteristics of Wastewater from The Source and After Sedimentation (*n*=50)

Parameter	Unit	Wastewater from the source	Wastewater after sedimentation	DAO 2016-08 Limits (Class C waters)
pH	unit	6.88	6.59	6.0 – 9.5
Turbidity	NTU	$\geq$800	76.16	--
TSS	mg/L	3,816.72	24.77	100
As	mg/L	0.155	0.0045	0.04
Ba	mg/L	0.154	0.0930	6.00
Cd	mg/L	0.008	0.0020	0.01
Hg	mg/L	0.143	0.0043	0.004
Pb	mg/L	7.938	0.0933	0.10

Filter Bed Materials

Figure 1 shows the filter bed system used in the experiment. It was composed of a sedimentation tank and filter bed, both of which were made of concrete. The filter bed box was packed with gravel, cocopeat and sand layers. Gravel and sand were sieved, and the particle sizes ranging from 5–20 mm and 1–2 mm, respectively, were collected and washed with deionized water before it was stacked inside the filter bed box.

Cocopeat was sourced from the stockpiles of coconut husk decorticating plant in Sariaya, Quezon Province. The same cocopeat samples were used as an adsorbent in the previous studies on batch and column tests conducted in University of the Philippines-Diliman [11] for removing heavy metals from aqueous solutions. Long fibers and impurities were removed from cocopeat samples and were subjected to physical characterization. Parameters such as bulk density (ρ_b), particle density, porosity (n), saturated hydraulic conductivity (*K_s*) of cocopeat were analyzed because these were needed as input parameters in HYDRUS-1D modeling. In the particle size distribution analysis, the cocopeat particle size ranged between 0.075 mm and 2.0 mm, and it was characterized as sand according to the USDA textural soil classification [18].

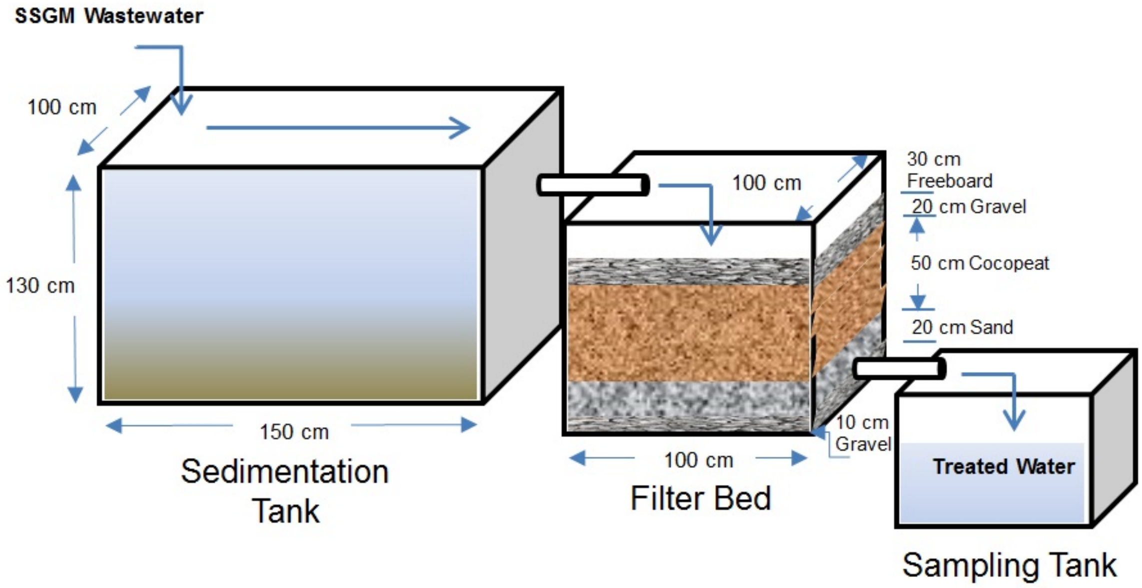

Figure 1. Sedimentation tank and cocopeat filter bed system used in the study

Table 2. Characteristics of Cocopeat Used in the Study

Parameter	**Unit**	**Value**
Bulk density (ρ_b)	g/cm^3	0.101
Particle density	g/cm^3	1.56
Moisture content	%	38.3
Porosity (n)	%	93.5
Particle size (mesh)	Mm	0.075 – 2.00
Saturated hydraulic conductivity (K_s)	cm/hr	70.0
Arsenic (As)	mg/kg	N.D.
Barium (Ba)	mg/kg	N.D.
Cadmium (Cd)	mg/kg	N.D.
Mercury (Hg)	mg/kg	0.208
Lead (Pb)	mg/kg	N.D.

Detection limits for As, Ba, Cd, Hg and Pb are 0.001, 0.2, 0.003, 0.0001 and 0.01 mg/L, respectively.

Complete physico-chemical characteristics of cocopeat used in this study and heavy metal concentrations are presented in Table 2. All heavy metals, except from Hg with 0.208 mg/kg, were not detected. The presence of Hg in the cocopeat sample used in the experiment can be traced from the environmental condition of stockpiles where the cocopeat is placed and stocked in an open area, and the sample is prone to sorb heavy metals including organic and inorganic Hg from different environmental media such as water, soil, vegetation and others. A total of 50.50 kg of cocopeat was stacked in the filter bed with the dimension of 100 x 100 x 50 cm (l x w x h).

Operation of Filter Bed System

SSGM wastewater samples from the site were introduced to the filter bed system for 50 days with a volume of 120 L per day. Wastewater was poured to the sedimentation tank with a flow rate of 40 L/hr to undergo settling and the overflow was collected through a distribution pipe connected to the filter bed. Overflow from the sedimentation tank was introduced as influent, to the filter bed and distributed evenly with the aid of gravel layered on the top of cocopeat. The effluent flowed downward through the packed layers inside the filter bed. Treated water in the bottom was collected by drainage pipes connected to the storage tank for sample collection. After 50 days of experimental runs, cocopeat samples from 5 layers (10, 20, 30, 40 and 50 cm) along the depth of cocopeat were collected and analyzed for heavy metal concentrations to determine the distributions of the adsorbed heavy metals in the cocopeat filter bed.

HYDRUS-1D Modeling

Soil Water Retention Curve

In this study, cocopeat was categorized as sand in HYDRUS-1D computer modeling [19]. A tension plate in contact with a hanging water column was used to determine the soil water retention curve [20, 21]. The volumetric water content (θ) of cocopeat at different pressure heads (h) was determined using the hanging water column technique, where θ was computed using,

$$\theta = v_l / V \tag{1}$$

where v_l is the volume of liquid, which was computed by the term $v_l = m_l / \rho_l$, where m_l is the mass of the liquid, ρ_l is the density of water, and V is the total volume of the cocopeat sample used. Pressure head and volumetric water content observed data by the tension plate experiment are presented in Table 3.

The collected data from hanging water column method (Table 3) were used as initial retention curve data to RETC Version 6.02 program [22] to fit a smooth curve to the observed data for HYDRUS-1D modeling. To give smooth functions of soil properties in HYDRUS-1D, the soil water retention curve equation, developed by van Genuchten in 1980 [23], was used in this study as followed;

$$S_e(h) = \frac{1}{\left[1 + (-\alpha h)^n\right]^m} \tag{2}$$

where $S_e(h)$ is the effective soil water saturation and α, n, and m are fitting parameters. Effective saturation (S_e) is expressed as;

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} \tag{3}$$

where θ_s is the saturated volumetric water content and θ_r is the residual water content. By using the two equations, and solving for $\theta(h)$;

$$\theta(h) = \frac{\theta_s - \theta_r}{\left[1 + (-\alpha h)^n\right]^m} \tag{4}$$

where the relationship between m and n is assumed to be $m = 1 - 1/n$.

Cocopeat was considered as sand in the soil catalog and provided by the van Genuchten water flow parameters as θ_r (0.045%), θ_s (0.430%), α (0.145), and n (2.680) except for K_s (1,680 cm/day) that was determined in the laboratory.

Table 3. Pressure Head vs Volumetric Water Content by Tension Plate Experiments

Pressure Head (*h*)	**Volumetric Water Content (θ)**
5	0.7235
10	0.6720
20	0.5670
30	0.5010
40	0.4643
50	0.4532
60	0.4410
70	0.4361
80	0.4357
90	0.4351
100	0.4342

HYDRUS-1D Model Parameters

Figure 2 shows the schematic diagram of the HYDRUS-1D modeling used in the present study. The main process of the model is to simulate a general solute transport of nonlinear cation adsorption. The soil type simulated for adsorption was the cocopeat in the filter bed with a column thickness of 50 cm. The fitted water flow parameters from RETC program were θ_r, θ_s, α, and n while K_s value was 1,680 cm/day. For the water flow boundary conditions; constant pressure head in the upper boundary and free drainage for lower boundary were set. The transport parameter fraction of the adsorption site of cocopeat ρ_b (0.101 g/cm^3) was set at 1 while longitudinal dispersivity was set at 10.

For the solute transport boundary conditions, the upper boundary was a concentration flux with known constant heavy metal concentration whereas the lower boundary was zero concentration gradient. In this study, Pb was the heavy metal under investigation since it had the highest concentration among the measured heavy metals from the SSGM wastewater (Table 1). Initial concentration of Pb was the average measured influent concentration in the experimental runs ($9.33\text{x}10^{-5}$ mg/cm^3). Observation nodes were set at 0, 10, 20, 30, 40 and 50 cm. The adsorption of Pb onto cocopeat with respect to depth was presented with time until the entire column was filled with Pb concentration equal to its initial concentration.

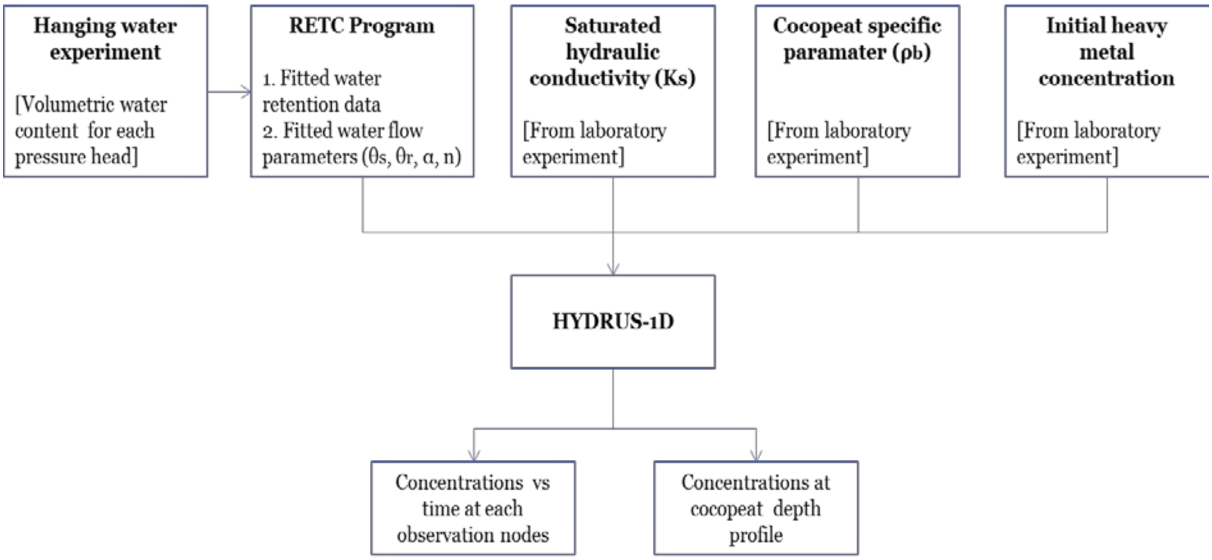

Figure 2. Schematic diagram of modeling using HYDRUS-1D

Water Flow and Solute Transport Model

HYDRUS-1D was used to simulate water flow and solute transport in variably saturated media. Transient water flow in the adsorbent domain was governed by Richards equation [20]. Solute transport equation in an unsaturated soil was obtained by combining the conservation equation, storage equation, vapor flux equation, reaction term and solute flux equation [21] In this model, water vapor phase was negligible; so the simplified solute transport equation was;

$$\frac{\partial}{\partial t}\left(\rho_b C_a + \theta C_l\right) = \frac{\partial}{\partial z}\left(D_e \frac{\partial C_l}{\partial z}\right) - \frac{\partial}{\partial z}\left(J_w C_l\right) - \left(\rho_b r_a + \theta r_l\right) \tag{5}$$

where ρ_b is the bulk density [M L^{-3}], C_a is the adsorbed chemical concentration [$\mathrm{M_{sorbent}}$ M^{-1}dry soil], θ is the water content, C_l is the dissolved chemical concentration [M L^{-3}], z is distance [L], D_e is the effective dispersion coefficient [L^2 T^{-1}], J_w is the volumetric flux density [L T^{-1}], r_a and r_l is the sorbed [M^{-1} soil] and dissolved [L^{-3}] reactions, respectively.

Inverse Solution Modeling

Inverse solution modeling was performed to come up with the fitted values of K_d and beta (or *1/n*) of Pb since these values were not established from batch and column adsorption experiments in the previous studies.

Direct Modeling

The fitted values K_d and *beta* (or *1/n*) of Pb gathered from inverse solution modeling were used as input parameters for the direct modeling to determine the useful life of cocopeat. For the printing times, 50, 1,000, 2,000 and 8,000 days were set while observation nodes at the adsorbent depths were set at 0, 10, 20, 30, 40 and 50 cm depths. The process of modeling solute transport with nonlinear cation adsorption was followed in the direct modeling. The final time, when the entire column was filled with the Pb concentration equal to its initial concentration, was considered as the time in which the filter bed was saturated and replacement of fresh cocopeat adsorbent was required.

Results and Discussion

Heavy Metals Adsorbed onto Cocopeat Column

After 50 days of operation and consuming a total of 6,000 L of raw SSGM wastewater, the measured total heavy metals adsorbed onto cocopeat were 13.20 259.20, 2.40, 25.20, and 462.00 mg for As, Ba, Cd, Hg and Pb, respectively. Most of the heavy metals (As, Cd, Hg) were adsorbed onto cocopeat at the top 20 cm of the column while Ba and Pb were adsorbed at the top 30 cm (Figure 3a-e). The adsorption in the upper part of the column was enhanced with the presence of organic matter in cocopeat that contains polar functional groups, which dissociate and take part in metal uptake through surface complexation and exchange of metal cations [24]. Substantial concentrations of Pb and Hg observed in the lower part of the cocopeat suggest that Pb and Hg leaching occurred during the experiment. High organic carbon in cocopeat caused the possibility of Hg leaching from the top to bottom of the cocopeat column. The variation of concentration of Pb in the lower part of the cocopeat column (Figure 3e) can be attributed to the high amount of Pb entering the filter bed and the high affinity of Pb to lignin caused Pb ions to go deeper, as described by Guo et al. [25].

HYDRUS-1D Modeling

The flow through the soil column (50 cm) was calculated using fitted and measured data. For solute transport and reaction parameters, K_d and *1/n* values were the fitted values from inverse modeling. The initial concentration for Pb (9.326×10^{-5} mg/cm^3) used as a boundary condition was the average influent concentration entering the filter bed reactor during the experiment.

Results of Inverse Modeling for Pb

After executing the HYDRUS-1D program in an Inverse Solution mode, the resultant fitted values of K_d and beta (*1/n*) were 3.125×10^{-4} and 0.948, respectively. The fitted values K_d and *1/n* for Pb adsorption to cocopeat in this study, together with values found in literature are presented in Table 4. Freundlich constant, K_d fitted in HYDRUS-1D in this study was lower than the values found in literature, which has a range of 0.57-15.03 L/g. The lower value of K_d in this study may be attributed to the low initial concentration of Pb in an actual SSGM wastewater used in the experiment as an input in the modeling, compared to the spiked concentration aqueous solution used in the laboratory experiments. The value of beta (0.95) was within the suggested empirical value of $0<\beta<1$ [26]. In addition, the fitted beta value was within the values of other studies (0.3845-1.00) on Pb sorption onto cocopeat that suggest a favorable adsorption.

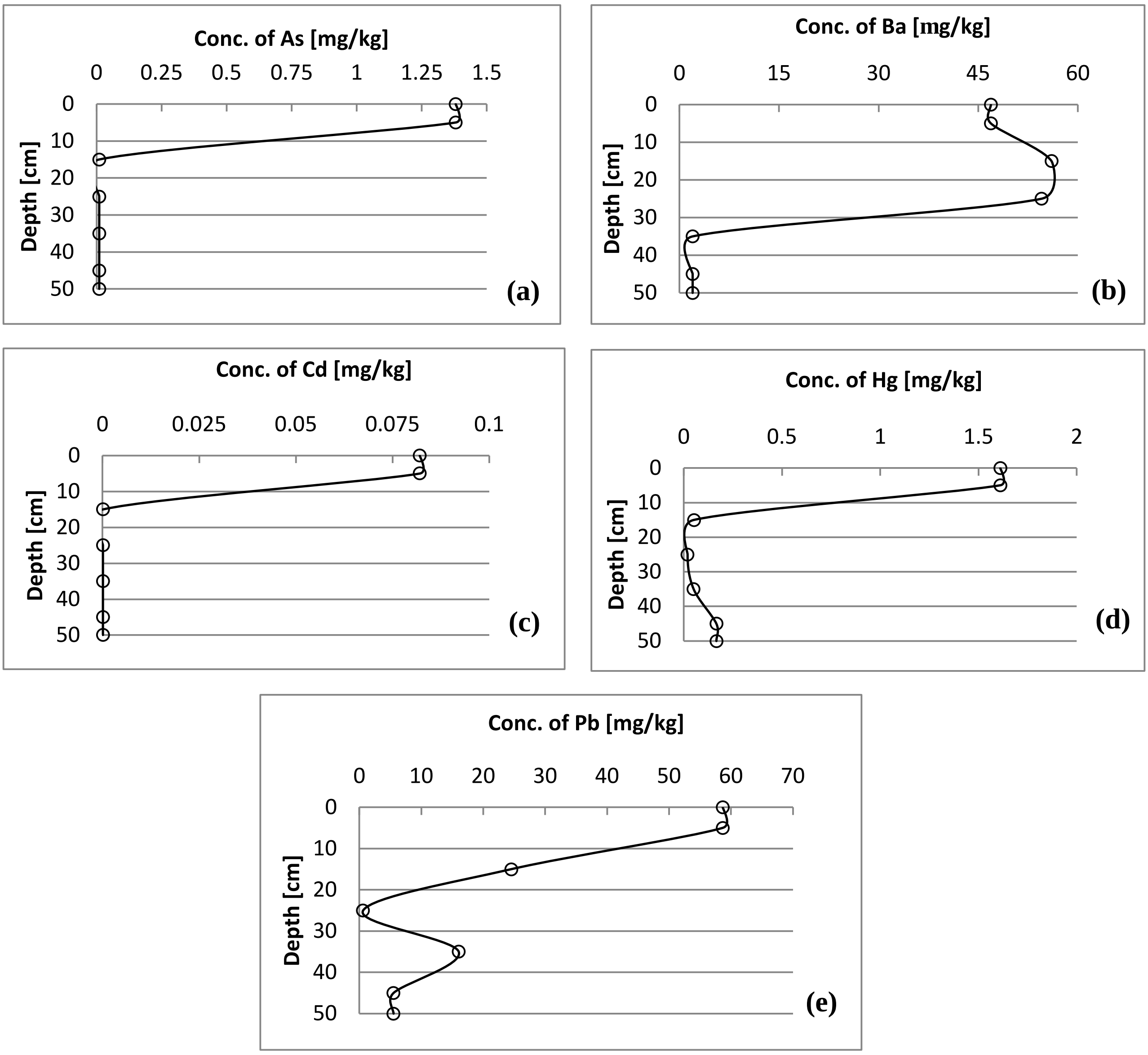

Figure 3. (a) As, (b) Ba, (c) Cd, (d) Hg, and (e) Pb concentration profiles with cocopeat depth after 50 days of operation

Results of Direct Modeling for Pb

The results of Pb concentration in the column at different times are shown in Figure 4 while the concentrations in different observation points set along the column are presented in Figure 5. Regression analysis of the results between measured and predicted concentrations in the upper 20 cm of the column after 50 days showed high positive correlation of $R^2 = 0.86$ (Figure 6).

By examining the observation points set along the cocopeat column (Figure 5), after 2,000, 4,000 and 6,000 days, Pb equilibrium concentrations at the bottom (50 cm depth) were $2.5\text{x}10^{-5}$, $7.0\text{x}10^{-5}$ and $8.8\text{x}10^{-5}$ mg/cm^3, respectively. At 8,000 days, the Pb concentration in the entire column was $9.326\text{x}10^{-5}$ mg/cm^3. This means that the cocopeat is saturated and no more adsorption occurs in the filter bed. At this period, replacement of cocopeat is recommended. When the input data measured from the experiment with daily wastewater application rate of 120 L and volumetric flow of 40 L/hr were used, the useful life of cocopeat that adsorbed Pb onto cocopeat filtered bed with a depth of 50 cm was calculated at 2.74 years.

Table 4. Values of K_d and Beta ($1/n$) Constants for Pb Sorption onto Cocopeat

References	K_d (L/g)	**Beta ($1/n$)**
HYDRUS-1D Fitted (This study)	3.125×10^{-4}	0.95
Hazeri, et al., 2012 [15]	2.11	0.65
Conrad and Bruun Hansen, 2007 [12]	2.90	1.00
Amarasinghe, 2011 [27]	15.03	0.3845
Ong, 2010 [28]	0.57	--

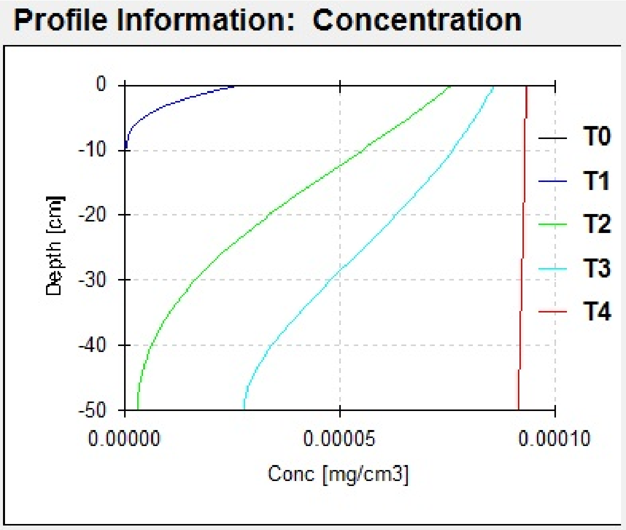

Figure 4. Profile showing Pb concentration along depth at t_0=0 day, t_1=50 days, t_2=1,000 days, t_3=2,000 days, and t_4=8,000 days.

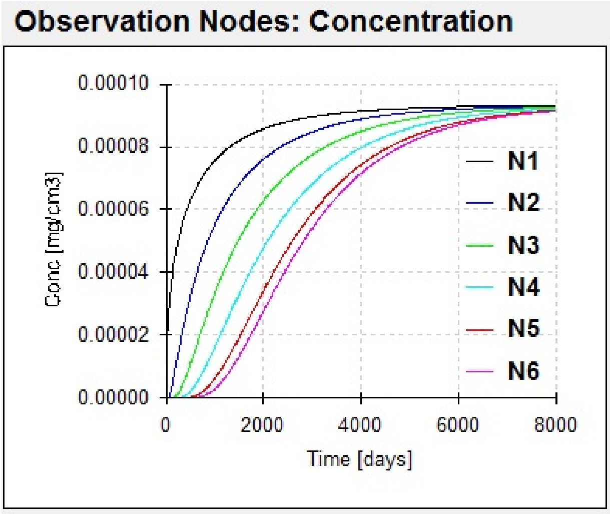

Figure 5. Observation nodes showing Pb concentration at n_1=0cm, n_2=10 cm, n_3=20 cm, n_4=30 cm, n_5=40 cm, n_6=50 cm.

Based on the results given by HYDRUS-1D using the parameters in this study, the life span of cocopeat can be managed by decreasing or increasing the amount of cocopeat to be put in the filter bed. Given the depth of cocopeat, its useful life in years can be estimated using the trendline in Figure 7 or using the generated equation, $y=0.055x$, where y is the useful life, and x is the depth of the cocopeat. This assumption was based on a 1 m x 1 m filter bed area and same depth of substrates used in this study. Depth of the filter bed reactor increases depending on the thickness of cocopeat stacked.

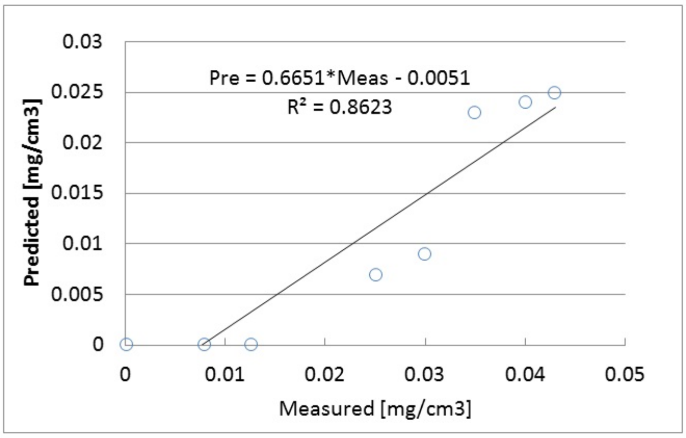

Figure 6. Predicted vs measured concentrations along cocopeat column after 50 days of operation

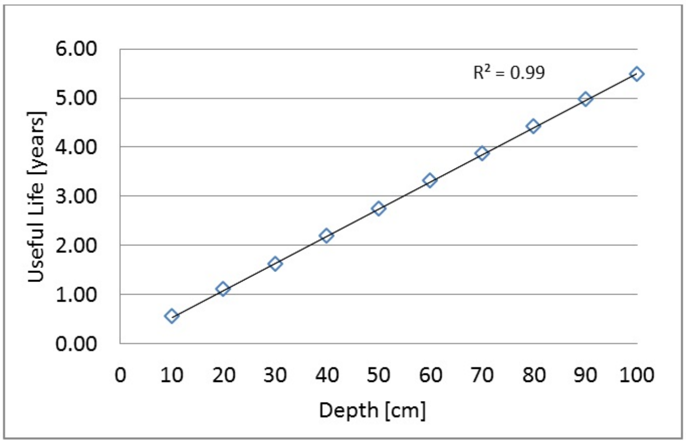

Figure 7. Thickness of cocopeat (cm) and its corresponding useful life (years)

Conclusions

In this study, a filter bed system composed of sedimentation and cocopeat filter bed was found suitable treatment system to remove heavy metals (As, Ba, Cd, Hg, and Pb) from an actual SSGM wastewater. With the application of hydrologic and flow parameters, depth of cocopeat, and reaction parameters from previous studies, HYDRUS-1D was able to simulate and predict that the useful life of cocopeat in a 1 m x 1 m x 0.50 m filter bed was 2.74 years for Pb. Measured and predicted concentrations in the upper 20 cm of the column after 50 days showed high positive correlation ($R^2 = 0.86$). Using the design parameters developed in this study, HYDRUS-1D was able to determine the extended useful life of cocopeat by increasing the thickness of the cocopeat at a rate of $y=0.055x$, where y is the useful life and x is the depth of the cocopeat.

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