

CADMIUM UPTAKE BY COCONUT PALM AND ITS SORPTION ON SOIL IN IONIC MEDIUM

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ABSTRACT

Uptake of cadmium investigated from a solution of Cd(II) in 0.010 M KNO₃ by coconut palm grown in soil, regularly supplied with different phosphate fertilizer sources, namely, triple super phosphate (TSP), imported rock phosphate (IRP), and Eppawala rock phosphate (ERP) for 17 years, indicates that the Cd levels of the soil with different treatments were less, at equilibrium state under laboratory conditions, than the maximum permissible limit of 0.05 mg/kg for agricultural soils. Further, coconut palm is found to uptake Cd from soil and translocate to leaf. The optimum equilibrium condition of soil with ionic solution was achieved after optimizing the parameters, such as shaking time, settling time, initial concentration and solution pH under laboratory conditions. The optimum values obtained are 6.0 h, 1.0 h, 0.70 mgL⁻¹ and 5–7, respectively. Adsorption isotherm analysis depicts that multilayer sorption of Cd(II) on soil is more probable as the results of adsorption equilibrium are better fitted with the Freundlich model than the Langmuir model. This phenomenon highlights the fact that Cd in soil solution easily gets replenished upon sorption of Cd by the coconut palm.

Key words: Cadmium, permissible limit, phosphate fertilizer, soil, sorption

INTRODUCTION

Cadmium is a trace metallic environmental pollutant present in soil, air, water and food (Josthna *et al.*, 2012). As this metal is non-biodegradable, it accumulates in the environment, posing problems on the environment as well as on human health (Salt *et al.*, 1995). Waste streams from industries, such as Zn production, phosphate ore implications, waste from artificial fertilizer production, and bio-industrial manure, cause Cd to enter soil component of the environment (Human, 2013). In addition, Cd enters the soil as depositions from the atmosphere to which Cd is evolved from industrial sources, vehicle exhaust or natural sources, such as volcanic eruptions. Anthropogenic sources add 3-10 times more Cd to the atmosphere than the natural sources do. The average abundance of Cd in earth's crust is suggested to be 0.01-0.05 mg/kg (Soares *et al.*, 2009) and the average concentration of Cd in fresh water ranges from 1 to 10 µgL⁻¹ (Sharmeen *et al.*, 2014). Cadmium added to water bodies as direct industrial effluents and from contaminated soil has a high mobility in the environment and easily absorbed by plants accumulating in tissues (De Maria *et al.*, 2013). Furthermore, it is documented that Cd accumulates in fruits and vegetables at high percentages, and hence, there is a possibility of getting it into human body (Roba *et al.*, 2016).

The scientific focus on toxicity of Cd has commenced during the middle of the last century although its industrial use was started long before. Any detectable biological functions in plants and animals related to Cd have not been identified (De Maria *et al.*, 2013). It is documented that earthworms and other essential organisms, such as soil microorganisms, are extremely sensitive to Cd poisoning as they die at very low Cd concentrations. These implications have environmental consequences on the soil structure as well as on the entire soil ecosystem (Human, 2013). It is considered as a dangerous environmental issue owing to the fact that the element gets into animals and humans through food chains causing dangerous effects as a result of high uptake of Cd from soil and bioaccumulation along the food chains (De Maria *et al.*, 2013). Cadmium is a trace metallic environmental pollutant present in soil, air, water and food (Josthna *et al.*, 2012). As this metal is non-biodegradable, it accumulates in the environment, posing problems on the environment as well as on human health (Salt *et al.*, 1995). Waste streams from industries, such as Zn production, phosphate ore implications, waste from artificial fertilizer production, and bio-industrial manure, cause Cd to enter soil component of the environment (Human, 2013). In addition, Cd enters the soil as depositions from the atmosphere to which Cd is evolved from industrial sources, vehicle exhaust or natural sources, such as volcanic eruptions. Anthropogenic sources add 3-10 times more Cd to the atmosphere than the natural sources do. The average abundance of Cd in earth's crust is suggested to be 0.01-0.05 mg/kg (Soares *et al.*, 2009) and the average concentration of Cd in fresh water ranges from 1 to 10 µgL⁻¹ (Sharmeen *et al.*, 2014). Cadmium

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Coconut Research Institute of Sri Lanka (CRISL) recommends the supply of the major nutrient requirements of nitrogen (N), phosphorous (P), potassium (K) and magnesium (Mg) to the coconut palm through fertilizer sources. Fertilizers, especially phosphorous fertilizers, appear to be one of the Cd sources to introduce Cd to coconut growing soils. It is documented that addition of phosphate fertilizers leads to accumulate Cd in soil causing elevated Cd levels in grazing animals than the permissible limit set in New Zealand, 1 mg/ kg of fresh body weight (Loganathan and Hedley, 1997). In addition, Cd content has been reported to be high in P fertilizers in Khouribga (Morocco) and Central Florida (USA) with average values of 15.06 and 9.13 mg/kg, respectively (Moulina *et al.*, 2006). The maximum permissible Cd content in P fertilizer is less than 3.0 mg/ kg [Sri Lanka Standards (SLS) specification for TSP and rock phosphate, 2014]. Therefore, in this research, accumulation of Cd in the leaves of coconut palm and soil were determined after 17 years of regular application of different phosphorous fertilizer sources in order to test the its uptake by the palm using leaf analysis. Nature of sorption on soil was evaluated in ionic media after optimizing the parameters under laboratory conditions. Finally, the data were fitted to two commonly used isotherm models, Langmuir and Freundlich, to determine the better-fitted model. As such, the objective of the present study was to investigate the uptake of Cd by the coconut palm from soils having less Cd than the permissible limit allowed for agricultural soil, and to investigate the nature of sorption of Cd in coconut growing soil in order to understand the pathway of replenishment of Cd to soil, enabling uptake by the palm.

MATERIALS AND METHODS

Determination of Cd accumulation in soil and coconut palm

Samples of soil and coconut palm leaves were collected from a site in Rathmalagara, in the Intermediate Climatic Zone of Sri Lanka, where the experiment was designed to evaluate the performance of palm with different P sources, namely, ERP, TSP, and IRP, applied along with other fertilizer sources, such as urea, muriate of potash and dolomite, at CRISL recommended rates. The amount of P supplied through different fertilizers were kept constant at the CRISL recommended rates (CRISL Annual Report, 2008). Soil of this site belongs to Andigama series (gravelly in nature) (Somasisiri *et al.*, 1994), where the palm was of tall variety. This experiment was commenced at the seedling stage so that the quantity of P sources supplied to the palms could be changed over time as per the requirement of the growth stage. The different P sources added for a palm during a period of 17 years with the treatment label followed by the dose are as follows: T1 - no P source; T2 - 18.12 kg of TSP; T3 - 25.65 kg of IRP; and T4 - 29.48 kg of ERP. The treatments were laid as a Randomized Block Design with three replicates having six palms per plot. All the samples were collected 17 years after establishment of the experiment during 2011.

Soil and leaf sampling and sample preparation

Twenty-four (24) soil samples and 12 leaf samples were collected from 12 plots. In each plot, a composite sample was made for each type of sample of three randomly selected palms. Soil samples were drawn at two depths, i.e. 0-25 cm and 25-50 cm. In each depth, representative samples were taken by the quartering method after thoroughly mixing three sub-samples (Thennakoon, 2000). Leaf samples were taken from the middle, approximately 10 cm portion of middle leaflets of the 14th frond (Thennakoon, 2000). Each soil sample was air dried, sieved and stored separately in plastic bottles at room temperature for further analysis. Leaf samples were first washed with tap water, and then with distilled water to remove any adhering extraneous material. These were left separately to drain water, oven dried at the temperature of 70 °C for 2-3 days until a constant weight was attained. The samples were ground using a grinder. Dried and ground leaf samples were stored in glass bottles until analysis.

Soil sampling, sample preparation, and soil characterization

Soil samples at the depth of 20-30 cm were collected and prepared as described in the section above. Soil texture and cation exchange capacity (CEC) were estimated according to the standard methods described (Dharmakeerthi *et al.*, 2007); pH and EC of soil samples were measured at 1:10 ratio of soil to deionized water by volume (Thennakoon, 2000), and organic carbon (OC) content was determined as in Walkley and Black (1934).

Analysis of samples for cadmium

Soil Cd was extracted with diethylenetriaminepenta acetic acid (DTPA) solution (Thennakoon, 2000). Leaf samples (0.5000 g) were digested with HNO₃:HClO₄ (4:1) acid mixture. Finally, Cd in all extracts were determined separately by using an atomic absorption spectrophotometer (AAS) (GBC 93.2 Plus).

Nature of Cd sorption in soil

Cadmium in soil usually reaches equilibrium between the soil surface and the soil solution. This condition was achieved while optimizing experimental parameters under laboratory conditions to identify the nature of sorption of Cd on soil. The data taken after the equilibrium was reached were fitted to isotherm models to determine a suitable model to describe Cd sorption on soil.

Optimizing the effects of parameters in soil

Effects of contact time, initial concentration and pH were optimized using batch experiments for a set of 24 samples having three replicates by changing experimental parameters. Each set consisted of 20.00 mL of Cd at different concentration in 0.010 mol/L KNO₃ and 1.00 g of soil in polypropylene bottles. Thereafter, each equilibrated solution was filtered leaving a pre-determined period of time to settle the contents. Finally, the concentration of Cd in solution was determined by AAS. The sorption percentage of Cd on soil in each parameter optimized was calculated using following equation (Navaratne *et al.*, 2013).

$$\text{Sorption percentage} = \frac{(C_i - C_o)}{C_o} \times 100 \%$$

Where, C_i and C_o are the initial and equilibrium metal ion concentrations, respectively.

The contact time is composed of shaking and settling times. The samples were shaken over 2 to 36 h and left to settle down over 20 min to 1.5 h. The initial concentrations were changed from 0.3 mgL⁻¹ to 10.0 mgL⁻¹. The pH range considered was 3.0 to 10.0. The samples were shaken using an end over end shaker at a rate of 22 rpm.

Isotherm analysis

The amount of Cd (μg/g) of soil sorbed was plotted against the equilibrium concentration of Cd to determine the standard type of isotherm. The sorption data was fitted to the two isotherm models, Langmuir and Freundlich, of which the standard equations are given in Table 1.

Table 1: Standard equations of isotherm models.

Isotherm Model	Standard Equation
LANGMUIR	$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{q_m \cdot K_L} \cdot \frac{1}{C_e}$
FREUNDLICH	$\ln q_e = \frac{1}{n} \ln C_e + \ln k_F$

where, q_e , q_m , C_e , K_L , K_F and n are described as equilibrium adsorption capacity, maximum adsorption capacity, equilibrium concentration of metal ion, Langmuir constant, Freundlich constants related to sorption capacity and sorption intensity respectively (State *et al.*, 2012).

Statistical analysis of data

Analysis of variance was performed using the SAS statistical package (1998), and standard deviations were calculated using Excel software.

RESULTS AND DISCUSSION

Cd Accumulation in Soil and Coconut Palm Uptake

The pH and EC of soils of respective treatments ranged as follows and there was no significant difference ($p < 0.05$) between the treatments: pH at 0 - 25 cm: 6.10 to 6.13; pH at 25 - 50 cm: 5.06 to 5.67; EC at 0 - 25 cm: 78.57 to 151.60 μS/cm; and EC at 25 - 50 cm: 79.80 to 98.47 μS/cm. According to the results obtained, soil pH of the experimental site is slightly acidic which would increase the Cd(II) availability in soil (Gray *et al.*, 1999).

The availability of Cd in soil and Cd sorption by palms between the different treatments are shown in Table 2. The data clearly reveal the availability of Cd in soil and Cd absorption by the coconut palm. However, there was no significant difference of Cd in the soil or in plant samples between the different phosphorus fertilizer treatments.

Table 2: Cd Contents in soil and palm leaf with different phosphorous fertilizer treatments

Treatment	Cd Content		
	Soil ($\mu\text{g}/\text{kg}$)		Leaf (mg/kg)
	0 - 25 cm	25 - 50 cm	
T1-NO P SOURCE	0.62	0.05	0.02
T2-TSP	0.80	1.00	0.19
T3-IRP	0.40	0.10	0.09
T4-ERP	2.10	0.90	0.04
SIGNIFICANCE	ns	ns	ns

ns: not significant

The soil samples treated with ERP showed higher Cd levels as compared to all other treatments. This is probably due to the dissolution of different types of phosphorous-containing substances in soil. It is known that both TSP and IRP fertilizers are much soluble in soil than the ERP is (Amarawansa and Indraratne 2010). Hence, there is a possibility in releasing Cd(II) to soil and thereby allowing continued plant absorption. Therefore, the higher concentration of Cd in ERP treated top soil could be the result of availability of Cd(II) after a long period of ERP application. Although ERP-treated soil shows higher values, there is no significant difference among the treatments.

Table 2 further reveals that the concentration of Cd in soil is comparatively lower than that in the leaf. This is an indication of bioaccumulation of Cd by the coconut palms as in some other plants, such as rice, fruits and vegetables (Hale *et al.*, 2012; Herath *et al.*, 2014; Hilber *et al.*, 2012). The Cd content in soils of none of the treatments exceeded the usual abundance range of 0.01 to 0.05 mg/kg (Soares *et al.*, 2009).

The Cd levels of leaf with no P treatment (T1) were within the normal range of 0.05 - 0.2 mg/kg in plant tissues (Cieśliński *et al.*, 1996). However, 17-year application of TSP would have led to increase the leaf Cd levels closer to the upper limit of the normal range; hence, it is strongly recommended to monitor these values for a longer period. The data also indicated a possibility of Cd accumulation by the coconut palm. Cadmium content of 0.38 - 1.98 mg/kg has been shown in both spinach and lettuce plants in the presence of less than 0.33 mg/kg available soil Cd (Page *et al.*, 1987). The higher Cd levels in these plant tissues could be the result of higher levels of Cd in soil (Table 2).

Soil pH, contents of clay minerals, manganese oxides and organic matter in the soil are the main factors affecting the movement of Cd from soil to plants (Cieśliński *et al.*, 1996). It is documented that 50% of root-absorbed Cd is retained in the root system, and the Soil pH, contents of clay minerals, manganese oxides and organic matter in the soil are the main factors affecting the movement of Cd from soil to plants (Cieśliński *et al.*, 1996). It is documented that 50% of root-absorbed Cd is retained in the root system, and the rest is translocated to different parts of the plant (Cieśliński *et al.*, 1996). Therefore, determination of the nature of Cd(II) sorption on soil of the same type in ionic medium, representing the normal soil solution, would be very important in coconut palm industry.

Nature of Cd sorption on soil

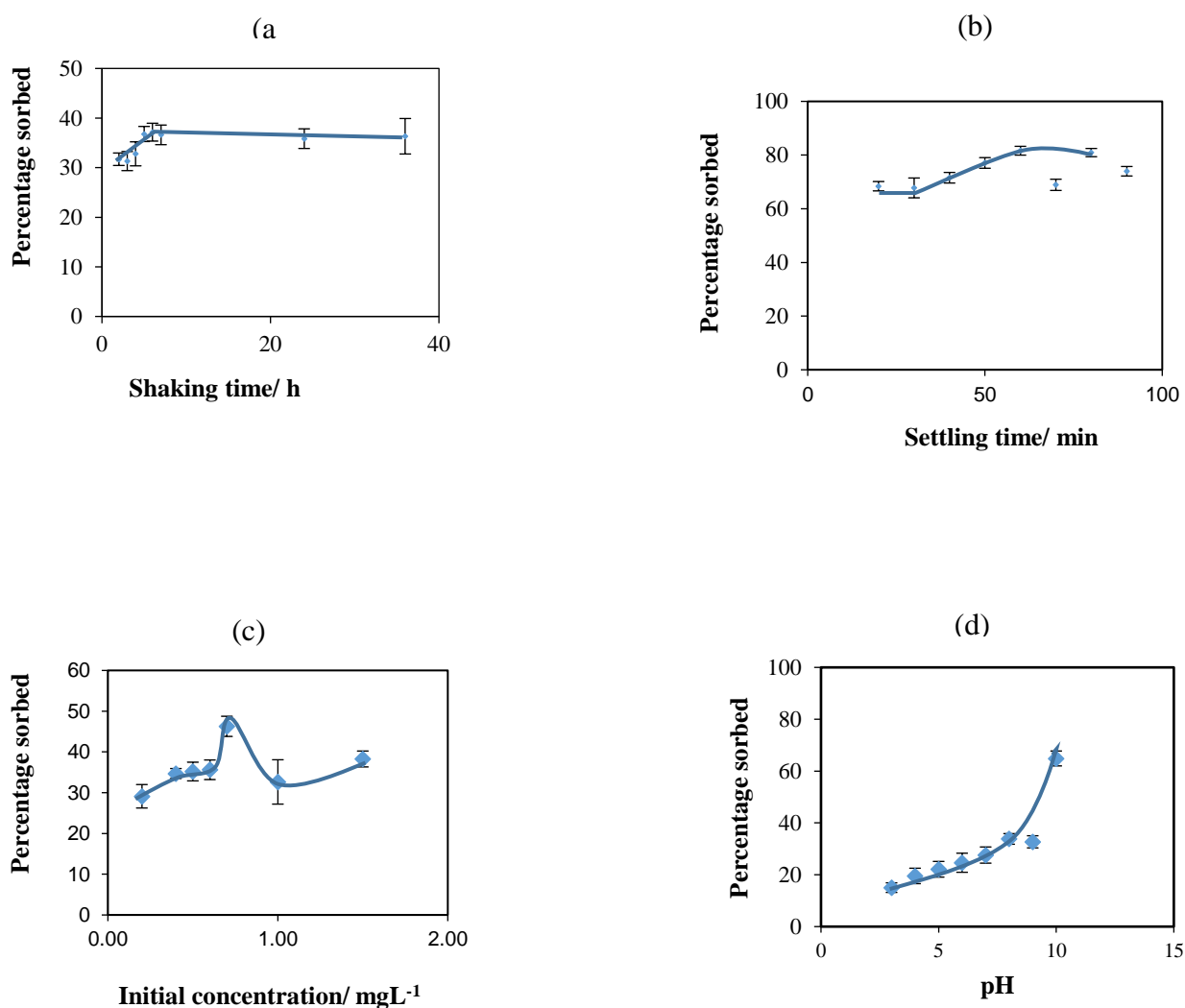
The nature of Cd sorption on soil was observed after optimizing each experimental parameter i.e., shaking time, settling time, initial concentration and pH through batch experiments under laboratory conditions. In addition, important soil characteristics were also determined (Table 3). The optimum parameters with respect to the soil characterized under laboratory conditions was obtained by studying the variation of the extent of sorption of Cd against shaking time, settling time, initial concentration and pH (Figure 1a and 1b). The optimum parameters with respect to the soil characterized under laboratory conditions was obtained by studying the variation of the extent of sorption of Cd against shaking time, settling time, initial concentration and pH (Figure 1a, 1b, 1d and 1c).

Table 3: Characteristics of the soil

Parameter	Value
CLAY CONTENT (%)	22
SILT CONTENT (%)	21
SAND CONTENT (%)	57
pH	4.84
EC ($\mu\text{s}/\text{cm}$)	24.7
CEC ($\text{cmol}^+/\text{100g}$)	19.55
OC CONTENT (%)	1.22

Values given in Table 3 indicates that the soil is sandy clay loam textured, acidic with comparatively high CEC, and low OC in comparison to a similar type of chemical fertilizer amended soil (Annual Report of CRISL, 2008).

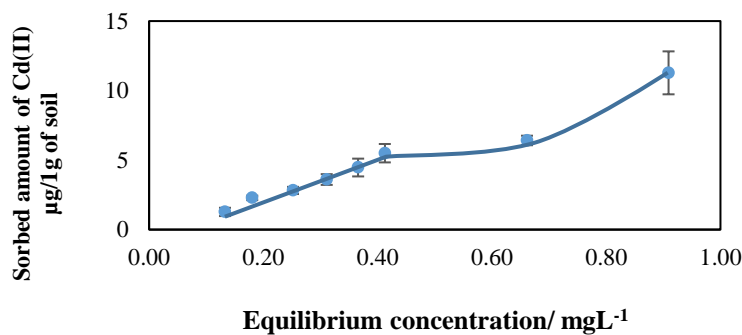
Figure: 1a. Variation in the extent of sorption of Cd(II) with (a) shaking time (with 1.00 g soil, 10 mgL⁻¹ Cd(II) solution, and 50-min settling time), (1b) settling time with 1.00 g soil. **1b.** Variation in the extent of sorption of Cd(II) with (1c) initial concentration. (with 1.00 g soil, 6-h shaking time, and 30-min settling time), and (1d) pH. (with 1.00 g soil, 0.70 mgL⁻¹ initial concentration, 6-h shaking time, and 30-min settling time).



Figures 1a and 1b reveal that the sorption of Cd has reached the maximum with shaking time and settling time at 6 h and at 60 min, respectively. Therefore, the optimum shaking time and settling time under the laboratory conditions were to be 6.0 h and 1.0 h, respectively. Change in the extent of sorption of Cd on soil with initial concentration of Cd(II) and pH are shown in Figures 1c and 1d, respectively. Figure 1c reveals that gradual increase of the extent of sorption of Cd reaches the maximum limit at 0.70 mg/L, followed by a decline and fluctuation with the increase of initial Cd(II) concentration. The same pattern has been reported to support the findings of this research (Mesquita, 1998). This is probably due to the competition of trace and major ions for the limited ion exchange sites in the ionic medium after initial sorption on the specific sites, comprising of various functional groups. Figure 1d exhibits a gradual increase in the extent of Cd sorbed upon increase in pH, followed by a rapid increase from pH 9.0 to 10.0. In the amount of Cd sorbed vs. pH relationship, the pH range between 5 and 7 can be considered as the optimum for sorption experiments under laboratory conditions. As the pH at ambient temperature of Cd(II) solution in 0.01 M KNO₃ was around 5.50, no pH adjustment was thus considered necessary. Sharp increase in Cd sorption in basic pH values is probably due to precipitation of Cd as its hydroxide.

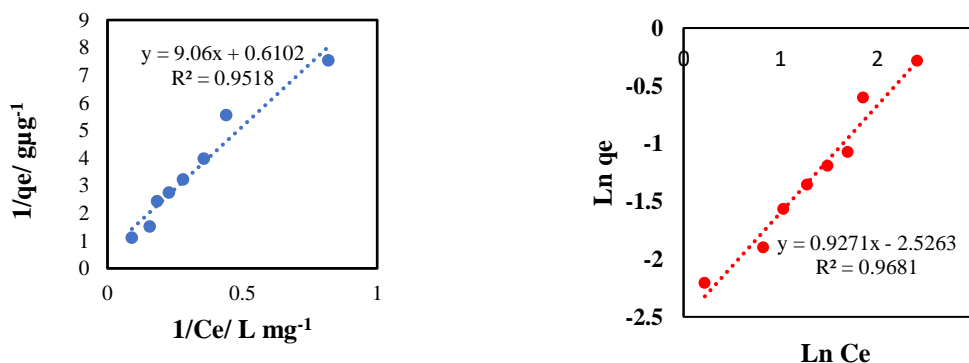
The nature of sorption of Cd on soil was investigated by studying variation of the amount of Cd sorbed with equilibrium concentration under optimized laboratory conditions (Figure 2).

Figure 2: Change in amount of Cd(II) sorbed (µg) on 1.00 g of soil with equilibrium concentration.



According to Figure 2, the amount of Cd(II) sorbed on soil increases with the increase in equilibrium concentration, then levels off to a constant value with a final continuous increase. This type of behaviour indicates multilayer sorption of Cd(II) on soil (State, et.al., 2012). Further, observation carried out using model-derived data indicates that regression coefficients (R²) of the Langmuir isotherm which describes monolayer sorption on homogeneous surface and Freundlich isotherm which illustrates multilayer sorption on heterogeneous surface models are 0.9518 and 0.9681, respectively (Figure 3). Higher R² value of the Freundlich isotherm model and soil being heterogeneous, the amount of Cd sorbed by soil is more than what is required for monolayer coverage. Adsorption of Cd onto the soil surface with concomitant transfer to micro and macro pores of the soil, and intra- and inter-particle diffusion would be possible modes of mass transfer of Cd.

Figure 3: Fitted isotherm data to (a) Langmuir model, and (b) Freundlich model



Note: qe and Ce amount of Cd(II) sorbed on 1 g of soil and equilibrium concentration respectively.

CONCLUSION

Coconut palm is able to absorb Cd from soil and translocate to leaf. The Freundlich adsorption isotherm model describes the multilayer sorption of Cd on sandy clay loam textured soil under equilibrium condition. Multilayers of sorption of Cd(II) on soil facilitates the replenishment of Cd(II) to soil solution when palm uptakes Cd(II) from soil solution. As the sorption of Cd by coconut palm is a complex process with many different modes of mass transfer, further research during a longer sorption period is needed for better understanding in an attempt to formulate a suitable model.

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