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RESEARCH ARTICLE

Removal of Three Phenolic Compounds [Polychlorinated biphenyl (PCB 105, PCB 118 & PCB 150)] from Aqueous Solutions by Fiber Palm as a Biosorbent

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Abstract

The aim of this work is to determine the potential of application of palm fiber as a biosorbent for removing of three phenolic compounds[polychlorinated biphenyl (PCB)] (2,3,3',4,4'-Pentachlorobiphenyl PCB 105, 2,3',4,4',5- Pentachlorobiphenyl; PCB 118, and 2,2',3,4',6,6'-Hexachlorobiphenyl; PCB-150 from aqueous solutions. The effect of pH, adsorbent dosage and contact time were investigated. Increase in the pH to neutrality resulted in the increase in the phenolic compounds adsorption capacity. The results showed that the increase in the fiber palm dosage to 0.5g/0.1L significantly increased the phenolic compounds adsorption rates. The adsorption process was fast, and it reached equilibrium in 30 min. contact time. The Freundlich and Langmuir adsorption models were used for mathematical description of the adsorption equilibrium and it was found that experimental data fitted very well to both Freundlich and Langmuir models. These results indicate clearly the efficiency of fiber as a low-cost solution for these compounds in aqueous solutions treatment and give some preliminary elements for the comprehension of the interactions between palm fiber as a bioadsorbent and the very polluting compounds from aqueous solutions.

Keywords: *Freundlich, Langmuir, Palm fiber and phenolic compounds.*

Introduction

A polychlorinated biphenyl (PCB) is an organic chlorine compound with the formula $C_{12}H_{10}{-}xCl_x$. Polychlorinated biphenyls were once widely deployed as dielectric and coolant fluids in electrical apparatus, carbonless copy paper and in heat transfer fluids [1]. Because of their longevity, PCBs are still widely in use, even though their manufacture has declined drastically since the 1960s, when a host of problems were identified [2]. Because of PCBs' environmental toxicity and classification as a persistent organic pollutant, PCB production was banned by the United States Congress in 1979 and by the Stockholm Convention on Persistent Organic Pollutants in 2001 [3].

The International Agency for Research on Cancer (IARC) rendered PCBs as definite carcinogens in humans. According to the U.S. Environmental Protection Agency (EPA), PCBs cause cancer in animals and are probable human carcinogens [4]. Many rivers and buildings including schools, parks, and other sites are contaminated with PCBs, and there have been contaminations of food supplies with the substances. Some PCBs share a structural similarity and toxic mode of action with dioxin [5]. Other toxic effects such as endocrine disruption (notably blocking of thyroid system functioning) and neurotoxicity are known [6]. The maximum allowable contaminant level in drinking water in the United States is set at zero, but because of the limitations of water treatment technologies, a level of 0.5 parts per billion is the de facto level [7].In recent years; interest has been focused on the removal of phenols from aqueous solution.

Traditionally, biological treatment, activated carbon adsorption, solvent extraction, chemical oxidation and electrochemical methods are the most widely used methods for removing phenol and phenolic compounds from wastewaters [8]. Activated carbon is one of the most effective ones, as it has a porous structure and provides a good capacity for the adsorption of organic compounds due to its high surface area [9, 10]. However, activated carbon has a number of disadvantages, such as relatively high cost, expensive cost and considerable loss during chemical or thermal regeneration of spent carbon. This has led many researchers to search for more cost-effective and efficient adsorbents to remove organic contaminants from water and wastewater. Fly ash, [11] rice husk, [12] peat, [13] bentonite, [14] and polymeric adsorbents [15, 16] have been tested for the adsorption of organic pollutants.

The search for new and innovative treatment techniques has focused attention on the adsorption capacities of other adsorbents, such as agricultural by-products [17, 19] which are readily available and do not need to be regenerated due to their low cost. Numerous studies on the sorption of metals and organic pollutants by these alternative adsorbents in batch systems have been reported [17, 20].

Regarding to the sorption of organic pollutants by pine bark reported interesting results for organochlorine pesticides [21]. Adsorption of phenol on formaldehyde-pretreated Pinus pinaster bark: equilibrium and kinetics was studied by Vazquez et al [23]. In Iraq, most historians and researchers that the oldest known of palms were in Babylon on (4000B.C) [24].

It includes several groups cultivated in a wide range for its economic importance such as date palm, coconut palm, and oil palm .Date palm "the concern of this study" belongs to pal mea family, phoenix class, and Dactylifera type [25]. It is a wooden aged plant of one cleavage [26]. It consists of roots, trunk, Fronds (palm leaves), fiber stem, and leaf base. The chemical structure of these parts is cellulose, hemicellulose, and protein, organic and mineral materials, which are different in their composition in various parts [27].Date palms have feather compound leaves.

The complete palm leave consists of two parts, the hyper part consists of spines as to lower part that includes palm leave base, and it is one of the leave parts which connect the leave with the trunk. Normally it is wide and hard, and its size ranges between (0.25m) to (0.5m) wide by various groups.

The two side edges are tapered ended with fiber sheath stick normally, the leaf base is wide at its connection with trunk and tapered upward [28].The aim of this present work is to investigate the biosorption potentiality of Fiber biosorbent material for the removal of PCB 105, PCB 118 & PCB 150 from aqueous medium. Fiber palm was characterized before and after adsorption of phenolic compounds by Fourier Transform Infrared (FTIR) spectroscopy studies. The effect of various factors, such as pH, biosorbent dosage bent dosage and time of contact on this biosorption process was investigated under batch equilibrium technique.

Materials and Methods

Preparation of Biosorbent

Fiber of palm were purchased from local market of South of Thi Qar, The fiber of date palm tree class Zahady campus and identified and authenticated from Department of Biology-sciences. Fiber was thoroughly washed with distilled water to remove mud and dirt. Then Fiber was soaked in 0.1M Noah to remove lignin based color materials followed by 0.1M H_2SO_4. Finally it was washed with distilled water several times and dried in an oven at 80^oC for 6 h and cooled at room temperature in desiccators.

The chemical content of Fiber is carried out using standard methods [29] and the results are given in Table 1. The dried Fiber was stored in desiccators until used. The dried bark was ground to fine powder and used as biosorbent without any pre-treatment for phenolic compounds adsorption.

Table 1: The chemical analysis of the date palm leaf base

Compound	Wt%
Cellulose	42.59
Heme cellulose	21.30
Lignin	12.20
Ash	7.50
Proteins	1.29
Lipids	0.37
Loss in Ignition	14.63
Total	99.88

Required raw materials, PCB 105, PCB 118 & PCB 150 (BDH Chemicals Co.) were used without further purification. Stock solutions were prepared by dissolving 1.0 g of PCB 105, PCB 118 & PCB 150 individually in one liter of double distilled water. These stock solutions were used to prepare 100, 200, 300,400 and 500 mg/L solutions of PCB 105, PCB 118 & PCB 150. 0.1 M HCl and 0.1 M Noah used to adjust pH. Water used for preparation of solutions and cleaning adsorbents was generated in the laboratory by double distilling the deionized water.

Analysis of Phenolic Compounds

The concentration of PCB 105, PCB 118 & PCB 150, in aqueous medium was determined by measuring absorbance at wavelengths of 270 nm, 274 nm and 280 nm, respectively, Using UV-Spectrophotometer (Shimadzu UV-1601 Spectrophotometer, Japan). In order to reduce measurement errors in all the experiments, UV absorption intensity of each solution sample was measured in triplicates and the average value was used to calculate the equilibrium concentration based on standard calibration curve, whose correlation coefficient square (R^2) was 0.999. The experimental error was observed to be within ±2%.

Batch Studies

Adsorption experiments were conducted by varying pH, adsorbent dosage and contact time. In adsorption equilibrium, experiments were conducted in a set of 100 mL Erlenmeyer flasks, where solutions of phenolic compounds 100 mL (PCB 105, PCB 118 & PCB 150) with different initial concentrations (100-500 mg/L) were added in these flasks.

Equal masses of 0.4g/0.1L of Fiber were added to phenolic compounds solutions and each sample was kept in a shaking Water Bath Temperature controller of 250 rpm at 25±1^{0}C for 1h to reach equilibrium of the solid-solution mixture. Samples were taken out from flasks and the solutions were separated from the adsorbent by filtered through Whatman No.50 filter paper to eliminate any fine particles. Then the concentration of phenolic compounds was determined by measuring absorbance using UV spectrophotometer at 270 nm, 274 nm and 280 nm for PCB 105, PCB 118 & PCB 150, respectively. The amount of adsorption at equilibrium, qe (mg/g) and the percent adsorption (%) was computed as follows:

$$q_e = \frac{(C_0 - C_{eq})V}{m} \qquad (1)$$

$$\text{Percent adsorption} \quad (\%) = \frac{C_0 - C}{C_0} \times 100 \qquad (2)$$

Where C_0 and C_{eq} (mg/L) are the liquid-phase concentrations of phenolic compounds at initial and equilibrium, respectively. V (L) is the volume of the solution, m is the mass (g) of the powder and C the phenolic compounds concentrations at the end of adsorption.

Results and Discussion

Characterization of Palm fiber Powder Fourier Transform Infrared (FTIR) Studies

Characterization of the biosorbent was carried out by Fourier Transform Infrared Spectroscopy to determine the type of functional groups on the biosorbent. FTIR spectra in the range of 4000-400 cm^{-1} for the pure fiber powder and the Fiber loaded with PCB 105, PCB 118 & PCB 150 .

The spectra display a number of absorption peaks, indicating the complex nature of the biosorbent material examined.

The FTIR spectra had a broad band absorption peaks at 3426 cm^{-1} corresponding to the overlapping of -OH and -NH peaks. On the other hand, the two peaks appear at 2918-2850 cm^{-1} which represent the asymmetrical and symmetrical stretching vibration of methylene ($-CH_2$) group, respectively. The C=O stretching of Palm fiber powder as at 1613.8 cm^{-1}. The presence of -C-O and -C-N linkages are confirmed from the peaks at 1321.7 and 1033.2 cm^{-1}.,

A significant difference can be seen in the FTIR spectra of biosorbent before and after adsorption. The FTIR band of C=O stretching shifted to higher frequency due to involvement of carboxyl (-C=O) group in the adsorption process of phenolic compounds with pure bark powder. This shift and/or broadening of some of the FTIR spectral peaks of the biosorbent in the presence of the phenolic compounds studied provides a clear indication that the functional groups like- NH_2, -OH and -C=O present on the Fiber palm surface are involved in PCB 105, PCB 118 & PCB 150 adsorption.

Effect of PH

The most important parameter influencing the adsorption capacity is the pH of adsorption medium [30]. The final pH of an adsorption medium affects the adsorption mechanisms on the adsorbent surface and influences the nature of the physico-chemical interactions of the species in solution and the adsorptive sites of adsorbents (31). Phenolic compounds biosorption onto Fiber as a function of pH ranging from 3.5 to 11.0 are given in Fig. 2. The removal of phenolic compounds by Fiber was highly dependent on pH.

In order to optimize the pH for maximum removal efficiency, experiments were conducted in the pH range from 3.5 to 11.0 using 0.5 g of Fiber with 100 ml of 100 mg/L initial phenolic compounds concentrations at 25±1°C. In the alkaline range, the pH was varied using aqueous 0.1 M NaOH, where as in the acidic range, pH was varied using 0.1M HCl. The FTIR spectroscopic analysis showed that the Fiber has a variety of functional groups, such as carboxyl, hydroxyl and amine and these groups are involved in almost all potential binding mechanisms.

Moreover, depending on the pH values of the aqueous solutions these functional groups participate in phenolic compounds bindings. The maximum adsorption capacities increase with increase in pH up to pH 7 and decreases there on. It was observed that the relative amount of phenolic compounds adsorbed was significantly affected by pH. The maximum adsorption occurs at neutral pH. Significant decline in removal efficiency was observed for further increase in pH, which may be attributed to formation of phenolate anions. PH 7.0 was selected as an optimum pH value.

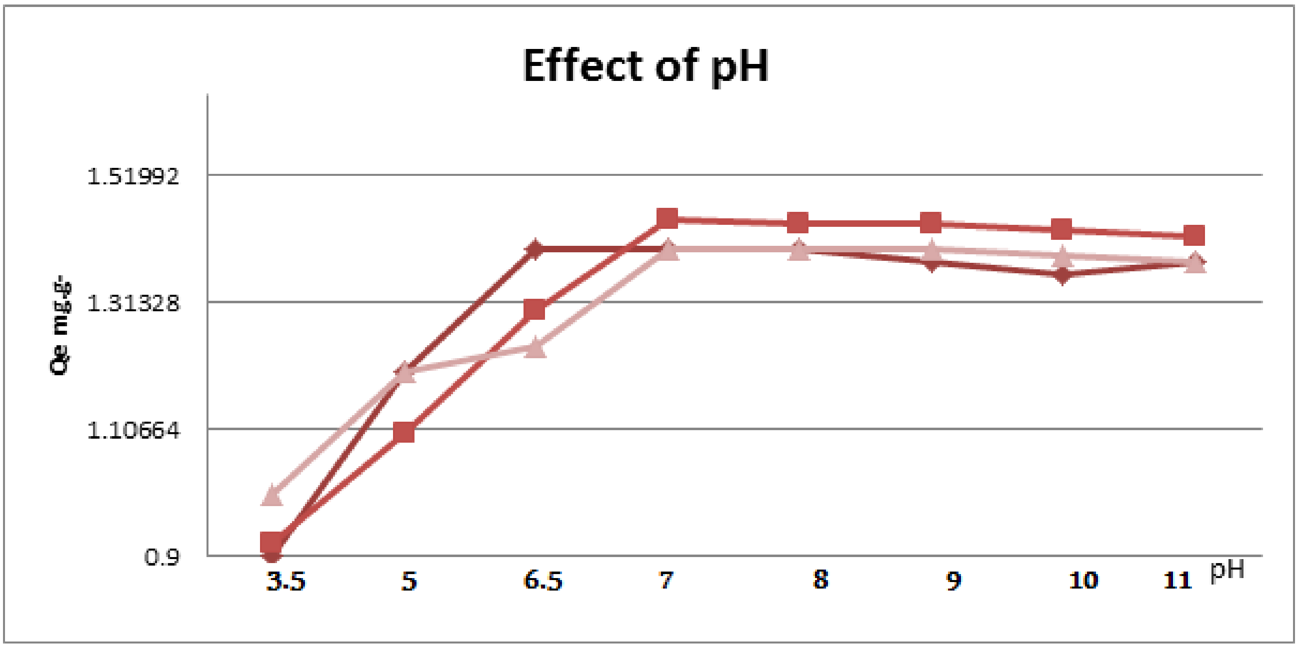

Fig 2: Effect of PH on the biosorption of (◊)PCB 105,(□) PCB 118 and (Δ) PCB 150onto fiber powder .Experimental conditions: for phenolic compounds: intiatial conc.=100 mg\l ,contact tim=30 min, biosorbent dosage =0.5 g\0.1l,Temp.=25 ◦C ±1

Effect of Biosorbent Dosage

The adsorptions of PCB 105, PCB 118 & PCB 150 on Fiber were carried out at different adsorbent dose by keeping other parameters constants. The relationship between adsorbent dose and substrate removal for same initial concentrations of phenolic compounds is presented in Fig. 3. It can be seen from Figure that percentage removal of PCB 105, PCB 118 & PCB 150 increased with the increase in adsorbent dose while loading capacity, q_e (mg/g), (amount of PCB 105, PCB 118 & PCB 150 loaded per unit weight of adsorbent) gradually decreased for the same. This increase in loading capacity is due to the availability of higher number of solutes (PCB 105, PCB 118 & PCB 150) per unit mass of adsorbent, i.e. higher solute/adsorbent ratio. These experiments were performed with initial concentration of 100 mg/L of solutes and neutral pH of the solution. It can also be seen from these Figures that the uptake of solute markedly increased up to adsorbent dose of 0.5g/0.1L and thereafter no significant increase was observed.

It can be concluded that the rate of phenolic compounds binding with adsorbent increases more rapidly in the initial stages and after some point the adsorption is marginal and becomes almost constant. Therefore, the optimum biosorbent dosage was taken as 0.5g/0.1L for further experiments.

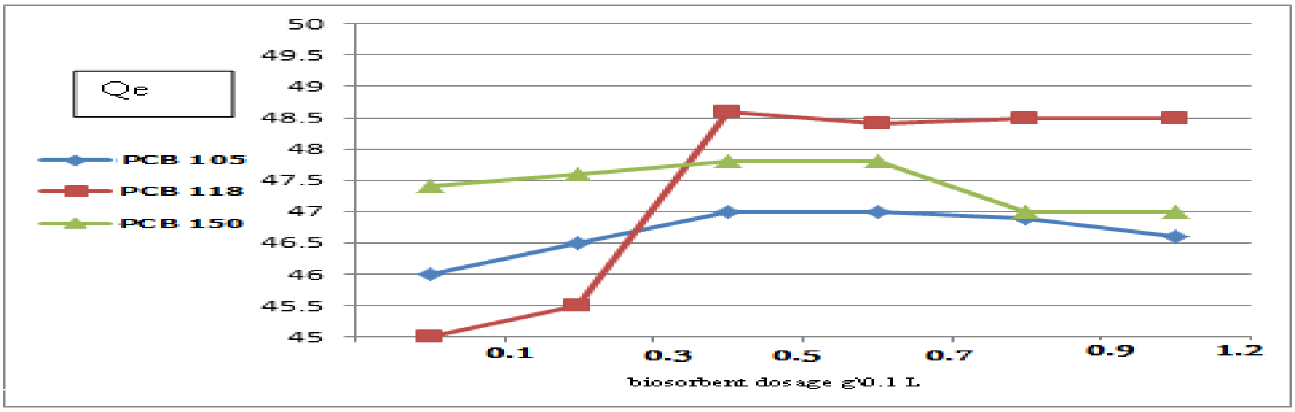

Fig 3: Effect of biosorbent dosage on the biosorption of (◊) PCB 105 (□) PCB 118 and (Δ) PCB 150onto fiber powder .Experimental conditions: for phenolic compounds: intiatial conc. =100 mg\l, contact tim=30 min, pH =7.0, Temp. =25 ◦C ±1

Effect of Contact Time and Initial Concentration

The effect of agitation time on the extent of adsorption of phenolic compounds at different concentrations is shown in Figure 4 for PCB 105, PCB 118 & PCB 150, respectively. Effect of shaking time on biosorption of phenolic compounds onto Fiber was studied over a shaking time of 0-120 min, using 0.5g/0.1L of Fiber, 100-500 mg/L of phenolic compounds concentration at pH 7.0, 25±1^{0}C and 250 rpm shaking speed. The saturation curves rise sharply in the initial stages, indicating that there are plenty of readily accessible sites. It can be seen from Figure that the contact time needed for phenol solutions with initial concentration of 100-500 mg/L to reach equilibrium was 30 min. Almost; no remarkable improvement was observed after longer contact time. After this equilibrium period, the amount of solute adsorbed did not change significantly with time, indicating that this time is sufficient to attain equilibrium for the maximum removal of phenolic compounds from aqueous solutions by Fiber. So, the optimum contact time was selected as 4h for further experiments. However, for adsorbate solutions with higher initial concentrations, lower equilibrium times were required. It was also seen that an increase in initial adsorbates solution concentrations resulted in increased phenolic compounds uptake. The removal curves are single, smooth and continuous, indicating the formation of monolayer coverage of the phenol molecules onto the outer surface of the adsorbent.

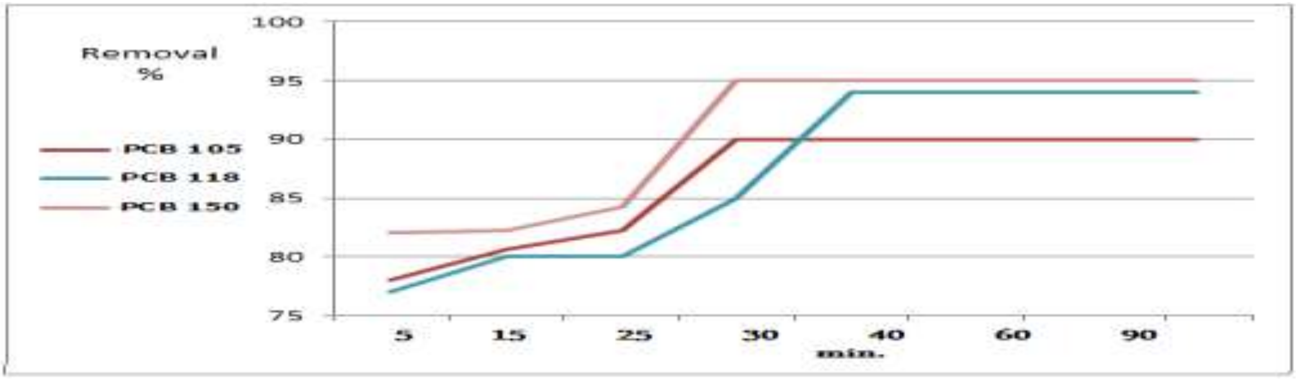

Fig 4: Effect of contact time on the removal % of (◊) phenol, (□) 20Cph and (Δ) 4-Cph onto fiber powder .Experimental conditions: for phenolic compounds: intiatial conc. =100 mg\l, biosorbent dosage =0.5 g\0.1l, pH =7.0, Temp. =25 ◦C ±1

Adsorption Isotherms

Equilibrium relationships between sorbent and sorbate are described by sorption isotherms, usually the ration between the quantity sorbed and that remaining in the solution at a fixed temperature at equilibrium. In order to optimize the design of a sorption system to remove phenolic compounds from aqueous solution, it is important to establish the most appropriate correlation for the equilibrium curve. Many theories which described adsorption equilibrium were applied. Several isotherm equations are available, and two important isotherms are selected for this study: the Langmuir and Freundlich isotherms [32, 33]. The linearised Langmuir equation is represented as follows:

$$C_e/q_e = 1/bQ_m + C_e/Q_m \quad (3)$$

Where q_e (mg/g) and C_e (g/L) are the amount of adsorbed phenolic compounds per unit weight of adsorbent and the unadsorbed phenolic compounds concentration in solution at equilibrium. b is the equilibrium constant or Langmuir constant related to the affinity of binding sites (L/g) and Q_m represents a particle limiting adsorption capacity when the surface is fully covered with phenolic compounds and assists in the comparison of adsorption performance.

The essential characteristic of the Langmuir isotherm can be expressed by the dimensionless constant called the equilibrium parameter, R_L, defined as:

$$R_L = 1 / (1 + b\, C_0) \quad (4)$$

Where b is the Langmuir constant, C_0 is the initial phenolic compounds concentration (g/L) and RL values indicate the type of isotherm to be irreversible (RL =0), favourable ($0< R_L <1$), linear ($R_L =1$), or unfavourable ($R_L >1$) (34). Our results show that the adsorption for phenolic compounds on the palm fiber is favourable and has an R_L value between 0 and 1 (table 2). Also, data were studied with the Freundlich isotherm, which can be expressed by logarithmic form as:

$$\ln q_e = \ln K_F + 1/n \ln C_e \quad (5)$$

Where K_F is a Freundlich constant that shows adsorption capacity of adsorbent, n is a constant which shows greatness of relationship between adsorbate and adsorbent.

The Freundlich describes reversible adsorption and is not restricted to the formation of monolayer. It has been found that the adsorption for phenolic compounds on the palm fiber is favourable and has an n value between 1 and 3 (Table 2). The q_m, b, R_L, R_1^2 (correlation coefficient for Langmuir isotherm), K_F, n, and R_2^2 (correlation coefficient for Freundlich isotherm) are given in Table 2 and the curves are shown in Fig 5a and Fig 5b.

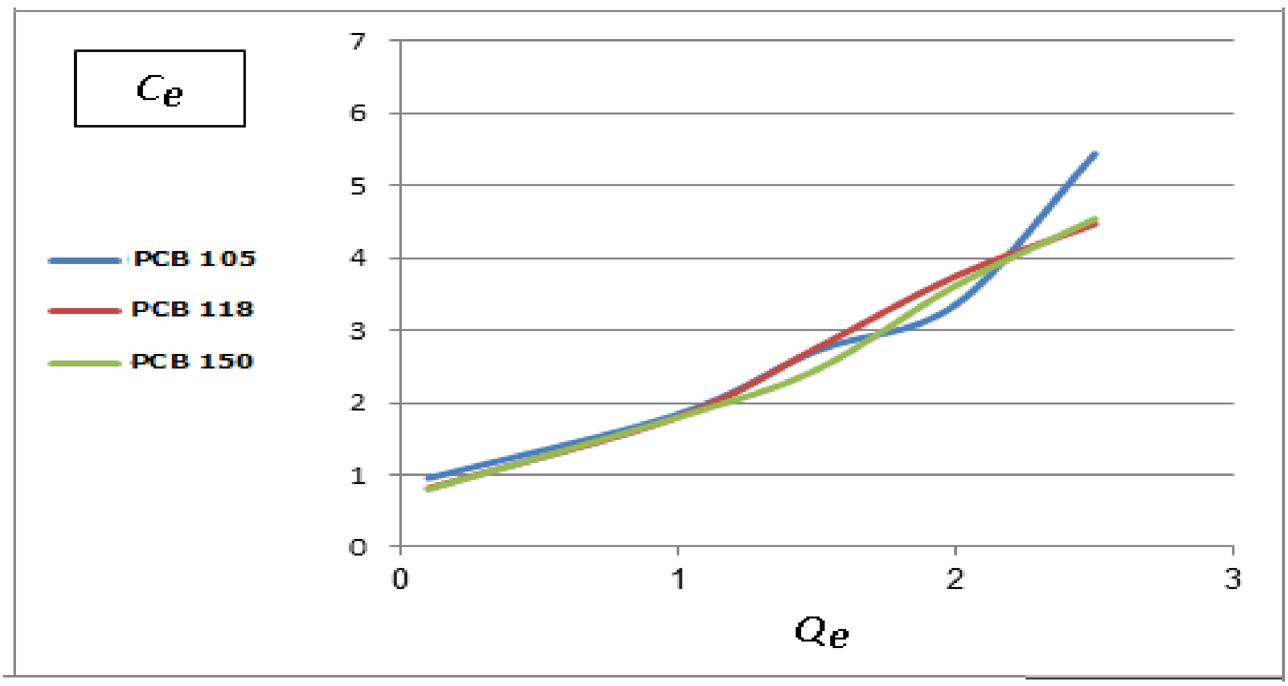

Fig 5a: Isotherm constants and the correlation coefficients of Langmuir

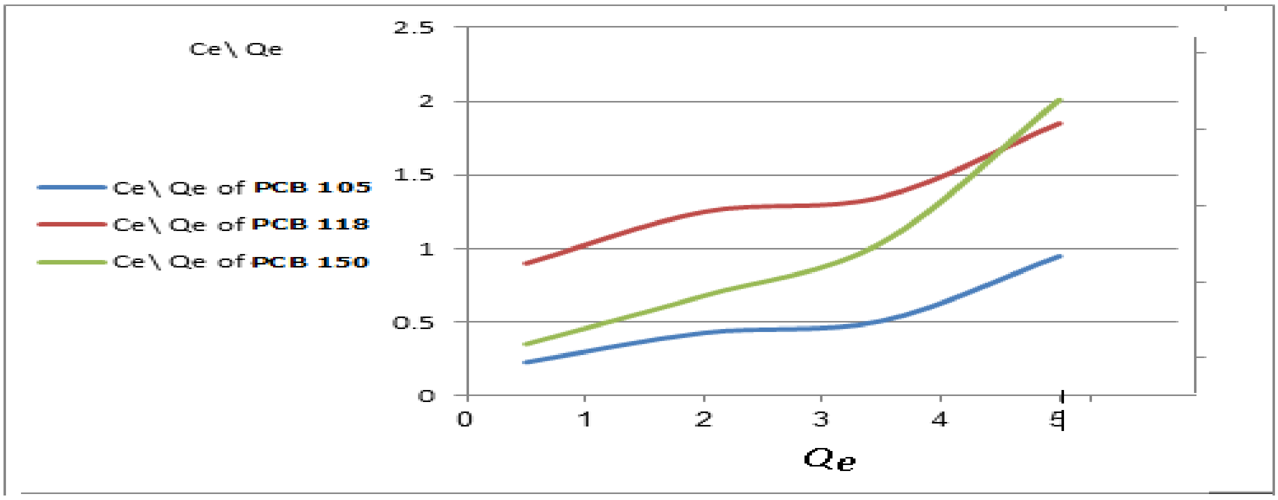

Fig 5b: Isotherm constants and the correlation coefficients of Freundlich isotherms

Table 2: Freundlich and Langmuir isotherm constants for phenolic compounds uptake by adsorbent

Phenolic compound	Langmuir equation			Freundlich equation		
	R_L	q_m	r^2	K_F	n	r^2
PCB 105	0.059	1.202	0.9907	0.947	1.940	0.8675
PCB 118	0.044	1.453	0.9953	0.597	2.147	0.9733
PCB 150	0.835	1.824	0.9974	-	-	0.4389

The $R_1$2and $R_2$2 values shown in Table 2 are evidence that the phenolic compounds adsorption in this study is well fitted to both Langmuir and Freundlich models; a possibility of mono and heterolayer phenolic compounds formation on the adsorbent surface. This observation is not rare as similar findings have been reported before [23, 35, 36]. This phenomenon can be further explained by understanding the surface chemistry of palm fiber used in this study. The presence of active functional groups with different intensity and non-uniform distribution may cause differences in the energy level of the active sites available on the palm fiber surface thus affecting its adsorption power. Active sites with higher energy level tend to form heterolayer phenolic compounds coverage with robust support from strong chemical bonding whilst active sites with lower energy level will induce monolayer coverage due to electrostatic forces. Table 3. Shown the comparison between Maximum Adsorption capacities Qo (mg\g) for the adsorption of PCB 105, PCB 118 & PCB 150 compounds by Various Adsorbents and the present study.

Table 3:Maximum Adsorption capacities Q_o (mg\g) for the adsorption of PCB 105, PCB 118 & PCB 150 compounds by Various Adsorbents

Sorbent	Adsorbates Qo (mg\g)			References
	PCB 105	PCB 118	PCB 150	
Organomodified bentonite	333	-	-	14
Bentonite &perlite	-	-	10.63 &5.4	32
CS\Ab blended beads	156	204	278	37
Sugarcane bagasse fly ash	23.83	-	-	38
Manganese nodule leached residue	28.5	-	-	39
Brown alga sargssum muticum	4.6	79	251	40
A.Leucocephala bark	94.33	147.05	181.81	41
Rice husk	7.91	-	36.23	42
Fly ash	3.85	-	-	43
Date seed carbon	138.88	-	-	44
Activated sewage sludge	29.46	-	-	45
Commercial activated carbon	49.72	-	-	46
Fiber of palm	98	89.6	120.2	Present study

Conclusions

Palm fiber has proven to be a promising material for the removal of contaminants from his aqueous solution. Not only palm fiber is an abundant cheap adsorbent, but it is also highly efficient for removing phenolic compounds from aqueous solution. The main characteristics of the adsorption process of the natural phenolic from aqueous solution on palm fiber can be summarized as follows:

- The palm fiber showed a high adsorption capacity of phenolic compounds (100 mg/l), revealing that palm fiber could be employed as a promising adsorbent for phenolic compounds adsorption.
- The adsorption process was very fast, and it reached equilibrium in 30 min. of contact. The equilibrium of the solid-phase concentration of phenols (q_e) decreased with increasing adsorbent concentration. This is mainly attributed to the unsaturation of the adsorption sites through the adsorption process.
- The pH played an obvious effect on the phenolic compounds adsorption capacity onto fiber. An increase in the solution pH leads to a significant increase in the adsorption capacities of phenolic compounds on the palm fiber, maximum adsorption capacity occurred at neutral pH.
- Both Langmuir and Freundlich isotherms provide good correlations for the adsorption of phenolic compounds onto palm fiber.

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