ADSORPTION BEHAVIOUR OF P-NITROPHENOL IN AQUEOUS MEDIUM ON NON-TREATED SABA AND LADY FINGER BANANA PEELS

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Abstract. The adsorption behavior of p-nitrophenol (pNP) was studied on two types of banana peels (Saba banana and Lady Finger banana) in aqueous medium. The results showed that, the optimum mass of banana peels to adsorb p-NP were 0.1 g (Saba banana) and 0.02 g (Lady Finger banana). The adsorption exhibits higher uptake of p-NP onto Saba and Lady Finger banana peels in pH 7. The adsorption of p-NP onto both type of banana peels achieved equilibrium at 480 minutes contact time. The adsorptions of p-NP onto banana peel well fitted with Freundlich isotherm (R^2 > 0.9). The FESEM micrographs showed that both peels have an irregular shape and porous texture which contributed to the adsorption behavior of p-NP in aqueous medium onto banana peels.

Keywords: phenolic compound, plant waste, banana peel, langmuir, freundlich, porous.

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Introduction

Banana is one of the most popular tropical fruits of the world. Banana peels constitutes up to 30 % of the ripe fruit and it's protects the inner fruit from sunlight and higher temperature. Since the banana fruits are widely available and the peel could be a potential source of natural adsorbents [1]. Most of the research study about banana peels as an adsorbent does not mention a type of banana [2 - 5]. Several researchers have been done about the potential of Saba banana peels as an adsorbent and there is no reported elsewhere about Ladyfinger banana peels as an adsorbent. Banana peels consist of hemicellulose, cellulose, lignin and pectin [6, 7]. This type of compound is oxygen containing group such as hydroxyl group, carboxyl group, absorbed water and alcohols. Different type of banana peels was given different amount of oxygen containing group. The oxygen containing group can provide more vacant site for adsorption [8]. There have been reported that the existence of different functional groups in banana peel allows the high sorption of different anionic and cationic compounds onto the surface-active sites [9].

Pollution of organic waste discharged from untreated industrial wastewater into rivers is becoming an issue and is gaining worldwide attention as it affects life, microorganisms and humans. The pollutants, such as metals, dyes, phenolic compounds, and anions need specialized treatment to prevent environmental pollutions from becoming worse [8, 10 -11]. For example, phenolic compounds like p-nitrophenols (p-NP) are harmful and toxic [10 -11]. P-NP has been selected as one of the persistent, bio-accumulative and toxic (PBT) chemicals by the US Environmental Protection Agency [8, 10]. Phenol and p-NP were reported to pose serious harm to the organisms at very low concentrations [11].

There are various treatment methods and techniques used to remove phenolic compounds such as p-NP from water namely ion exchange, oxidation, electrolysis, electro-dialysis, reverse osmosis, biodegradation, catalysis and adsorption [8,12]. Adsorption is the best method for removing phenolic compounds in aqueous solutions because it is sensitive to toxic contaminants, has flexibility and simplicity of design and ease of operation and involves lower costs than other treatment methods [5,12]. Activated carbon is the best adsorbent used in adsorption methods for various harmful compounds such a p-NP in water and wastewaters [13]. The surface chemistry and pore structure of porous carbon is the main criteria for selecting activated carbon in adsorption [14]. This activated carbon has been reported to remove various types of pollutants such as heavy metal [15], dyes [6, 16] micropollutants [17], p-nitrophenols [10 - 11], and pesticides [18].

Besides activated carbon, various adsorbents were tested to adsorbed p-NP and other hazardous compounds in aqueous solution in the past years. For example, a nanocomposities [19], organoclays [20], mansonia wood sawdust [21 - 22] and abundance of agriculture waste [23]. Banana peels is one of the abundance agriculture waste that can be used to remove hazardous compounds in an aqueous medium such as gases from motorcycle emissions [4], heavy metal [3, 24 - 25], and dye [5, 26]. Therefore, this study investigated the adsorption behavior of p-NP in aqueous medium on two kinds of banana peels, namely Saba banana (*Musa acuminata* × *balbisiana*) and Ladyfinger banana (*Musa* × *paradisiaca L.*).

Materials and Methods

Materials. Saba banana (*Musa acuminata* × *balbisiana*) and Ladyfinger banana (*Musa* × *paradisiaca L.*) peels were obtained from several banana fritters kiosk in Hulu Bernam, Selangor. P-nitrophenols (98% purity), hydrochloric acid and sodium hydroxide were purchased from Merck and Sigma Aldrich, respectively. Distilled water was utilized in all experiments.

Preparation and Characterization of Saba and Ladyfinger Banana Peels. The banana peels (1.0 kg) were washed with tap water several times, followed by distilled water to remove dirt and impurities on the peel's surface and cut into small pieces. Then, the peels were dried in an oven at 60 ^oC for 2 days to eliminate the peel's humidity. The dried peel were ground to powder form and stored in a desiccator for further study.

The surface morphology of banana peels was analyzed with Field Emission Scanning Electron Microscope (FESEM) Hitachi 8U 8020 UHR. Fourier Transform Infrared (FTIR) spectroscopy (Thermo Nicolet Nexus) spectra of banana peels were recorded using ATR method and the surface area properties were analysed using a Quantachrome Autosorb automated gas sorption analyser (Autosorb iQ-MP/XR), under nitrogen adsorption–desorption at 77K.

Kinetic and Isotherm Adsorption Studies of p-Nitrophenol (p-NP) onto Saba and Ladyfinger Banana Peels in Aqueous Medium. Kinetic adsorption studies were done with the optimum mass of banana peels, equilibrium contact time and pH. A solution of 50 mg/L p-NP was freshly prepared daily. The study of the optimum mass of adsorbent (banana peels) for the adsorption of p-NP was done by varying the banana peel mass. The mass of banana peels was manipulated in the range of 0.1 - 0.5 g for Saba banana and 0.02 - 0.10 g for Ladyfinger banana peels. The banana peels were added with the p-NP solution into a conical flask and shook on the orbital shaker (Model 720, Protech) at the speed of 120 rpm for 30 minutes. After 30 minutes, 10 mL of each mixture was filtered using a membrane filter (0.45 μm) and the absorbance intensity of the remaining p-NP solution was recorded using an Ultraviolet-Spectrophotometer (Agilent Cary 60) at wavelength 320 nm. These measurements were also applied to the initial concentration of p-NP (before the adsorption process). The value of pH was varied in the range of 2 – 10. The optimum mass of banana peels, p-NP concentration (50 mg/L), agitation speed rate (120 rpm) and contact time (30 minutes) were kept constant. The value of analyte pH was maintained by adding hydrochloric acid, HCl (0.1 M) and sodium hydroxide, NaOH (0.1 M). Experiments on equilibrium contact time was carried out at various time intervals ranging from 10 to 1440 minutes. All solution mixtures were filtered (0.45 μm membrane filter) and the absorbance intensity was measured using the same method as in the optimum mass experiment. The adsorption uptake was calculated using Equation 1 [12, 27] as follows;

$$q_e = \frac{(C_o - C_e)\ x\ V}{m} \tag{1}$$

q_e (mg/g) is the uptake of p-NP onto banana peels, C_o (mg/L) is the initial concentration of p-NP, C_e (mg/L) is the remaining concentration of p-NP and V is the experimental volume (L). The adsorption of p-NP onto both banana peels was investigated using pseudo first and pseudo second kinetic model. The pseudo first and second were used Equation 2 and 3 [27], respectively.

$$ln\ ln\left(q_e - q_t\right) = ln\ q_e - kt \tag{2}$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \tag{3}$$

where q_t (mg/g) and q_e (mg/g) are the amount of p-NP adsorbed by banana peels at time t (min) and equilibrium, respectively. k_1 (min^{-1}) is the equilibrium rate constant of pseudo-first-order adsorption [12, 27]. q_e and k_1 are determined from the intercept and slope of the plot of ln ($q_e - q_t$) versus t, respectively. The slope and intercept of the plot t/q_t versus t yielded k_2 and q_e, respectively.

The isotherm studies were carried out over an 8 hours equilibrium contact time, with the mass of banana peels (0.1 g (Saba banana and 0.02 g Ladyfinger) remaining constant. The concentrations of p-NP were varied in the range of 10 to 250 mg/L. After 8 hours, the mixtures were filtered using 0.45 μm membrane filter into bottle samples. The absorbance intensity and uptake were measured using the same method as in the kinetic studies experiment. For the adsorption of p-NP onto banana peels, two isotherms were tested: Langmuir and Freundlich. The monolayer adsorption of p-NP onto saba and Ladyfinger banana peels was described using the Langmuir isotherm. Equation 4 was used to plot the liner Langmuir isotherm [28 - 29]. The slope and intercept of the plot C_e/q_e against C_e can be used to determine maximum uptake, q_m, and the Langmuir constant k_L.

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m k_L} \tag{4}$$

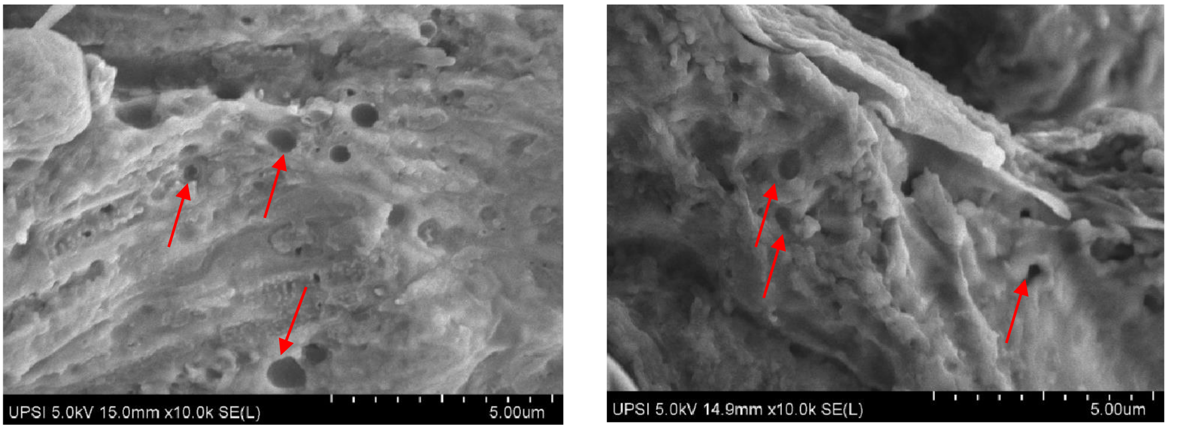
The adsorption onto heterogeneous surfaces was modelled with Freundlich isotherm and the linear isotherm can be defined according to Equation 5. Freundlich constants, k_F represents adsorption capacity, and n represents adsorption strength. The value of k_F and 1/n can be obtained from the intercept and slope of the linear plot of ln q_e versus ln C_e. The magnitude of 1/n demonstrate the adsorption favorability and heterogeneity degree of banana peels surface [27].

$$ln\, ln\, q_e = \frac{1}{n} ln\, ln\, C_e + ln\, ln\, k_F \tag{5}$$

Results and Discussion

Characterization of Saba and Ladyfinger Banana Peels. The surface morphology of Saba banana and Ladyfinger banana peel was studied using FESEM microscope (Figure 1). The FESEM micrograph showed that both peels have an irregular shape and porous texture which is similar as previously reported by others researchers [30 - 31]. The porous texture affects the surface area and further contributes to the adsorption rate of p-NP.

Figure 2 shows the adsorption and desorption of nitrogen gas onto both banana peels under nitrogen-desorption at 77K. The curve shows that the isotherm for Saba and Ladyfinger banana peel are type III implying weak interaction of absorbate-adsorbent [32]. The BET surface area of the Saba banana peel is higher than the Ladyfinger banana peel with a larger pore volume and diameter (Table 1). The average pore diameter of Saba banana consistent with the higher surface area and pore classification as macropores (> 50 nm) whereas, Ladyfinger banana has mesopores with average pore diameter of 36.61 nm. The wider range of pores of 2–50 nm, which are of particular significance in the context of capillary condensation, are known as mesopores [32].



(a) (b)

Figure 1. FESEM micrographs at 10.0k for Saba (a) and Ladyfinger (b) banana peels

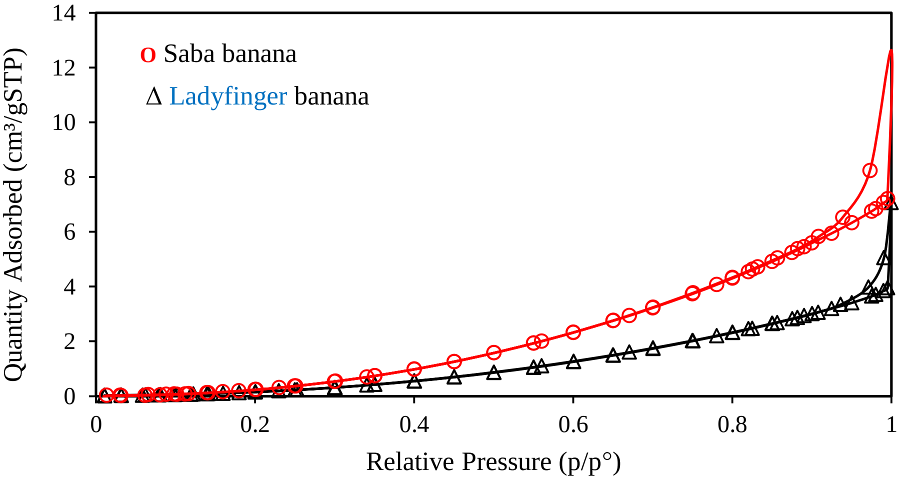


Figure 2. Adsorption-desorption of nitrogen gas onto Saba and Ladyfinger Banana Peels.

Table 1. Parameters of BET surface area analysis of banana peels

Banana peels	BET surface are (m^2/g)	Total pore volumes (cm^3/g)	Average pore diameter (nm)
Saba Banana	0.6820	0.008796	51.59
Ladyfinger Banana	0.5083	0.004653	36.61

The FTIR spectrums (Figure 3) of banana peels in range of 4000 - 500 cm^{-1} were taken to identify the functional groups of banana peels. The adsorption capacity and removal mechanism of adsorbates are influenced by the functional group in the banana peel. There are O-H stretching vibrations (3237.9 and 3207.1 cm^{-1}) from the spectrum of Saba and Ladyfinger banana peels. There is also aliphatic C-H at 2896.6 and 2902.3 cm^{-1}, C=C stretching at 1614.1, 1295.9 cm^{-1} and 1585.2, 1334.5 cm^{-1} and C-O stretching at 977.7 and 981.6 cm^{-1}. The O-H stretching vibrations correspond to cellulose, pectin, absorbed water, hemicellulose and lignin, whereas the C=C stretching that can be attributed to the presence of aromatic or benzene rings in lignin and C-O correspond to carboxylic acid and alcohols [33]. The functional group absorption peak of Saba banana peels more intense as compared to Ladyfinger banana peel demonstrate higher content of O-H, C=C, aliphatic C-H and C-O in Saba banana peels. The intensity of functional group peak decreased if fewer of the group present [7].

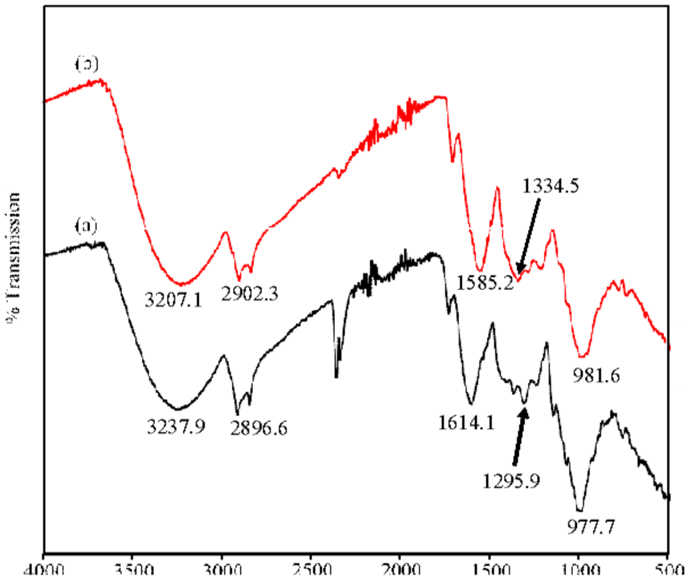


Figure 3. The FTIR Spectrum of Saba (a) and Ladyfinger (b) banana peels.

Kinetic and Isotherm Studies of p-Nitrophenol onto Saba and Ladyfinger Banana Peels in Aqueous Medium. The number of available adsorption sites increase with adsorbent mass, thus increasing the amount of phenolic compounds adsorbed [7]. Figure 4 shows that p-NP (mg/g) uptake onto banana peels decreases as the adsorbent mass increases. This phenomenon is believed to occur due to a limited number of p-NP associated with unchanged of adsorption rate and the active site in banana peels become saturated therefore reducing material’s adsorption capacity. The same phenomena occurs in previous studies using biochar to remove p-NP [12, 34].

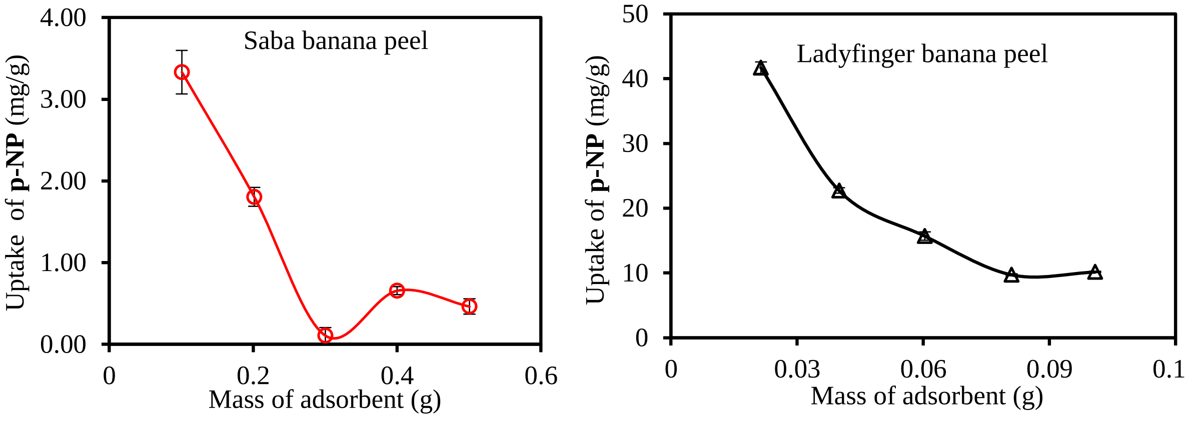


Figure 4. Effect of adsorbent mass against uptake of p-NP onto Saba and Ladyfinger banana peels.

The effect of pH on the adsorption of p-NP onto banana peels in aqueous medium is shown in Figure 5. The amount of adsorption uptake increases with pH and reach a maximum at pH 7 for both banana peels. Ladyfinger banana peel have greater uptake than Saba banana peel. At lower pH (< pH 4), H^+ ion is excess in an aqueous solution and may compete with p-

NP for adsorption sites on the banana peel. This phenomenon reduces the uptake of p-NP at lower pH. There is a reduction in adsorption rate at lower pH due to the high concentration and mobility of H^+ ion [33]. As the pH increase (pH 4 – 7), the number of H^+ ion in the aqueous solution decreased. This reduces the number of H^+ ion competing with the positive charge p-NP for the vacant sites. A higher pH (acidic medium) would enhance the p-NP adsorption due to the electrostatic forces of p-NP with the negatively charged surface of the adsorbent [35]. Hence, there is an increase in adsorption when the pH exceeds 2. As a result of the interaction of the oxygen containing functional groups from the banana peel with the ring and O-H group of p-NP via hydrogen bonding/electron donor-acceptor/lewis asid-base interaction, the adsorption of p-NP in the aqueous solution at lower pH was increased [27].

The uptake of p-NP onto Saba banana and Ladyfinger peels is significantly reduced above pH 7. The surface functional group is fully or partially deprotonated at higher pH, resulting in an increase in the negative charge on the surface. Furthermore, at pH lower than pKa (pKa =7.08), p-NP is present in the molecular form [21]. Because p-NP molecules become phenolates ions above pKa (alkali medium), no adsorption of p-NP molecules onto banana peels was observed [36]. The negative charge surface attracts electrostatic repulsion phenolates ions, and the presence of an excess hydroxyl group (pH>7), which competes with p-NP molecules, reduces adsorption uptake.

Figure 6 shows the effect of contact time on p-NP adsorption by Saba and Ladyfinger banana peels. For the first 20 minutes, the adsorption of p-NP onto banana peels increases rapidly (region I). This is due to many accessible vacant sites for adsorption, which increased the amount of p-NP accumulated on the surface of both banana peels. The best explanation is that the strong, attractive forces between p-NP and the adsorbent allow rapid diffusion into the interparticle matrix, leading to rapid equilibrium [37]. Both banana peels showed a decrease in uptake between 20 to 400 minutes (region II). This phenomenon was widely assumed to occur solely on the external surface, with the adsorbate quickly escaping to the solution. Saba and Ladyfinger banana peel equilibrium contact time was chosen to be within 480 minutes (8 hours). After equilibrium is achieved, there are no significant changes in p-NP uptake. This occurred due to the repulsive force between the positively charged p-NP adsorbed and the p-NP in the bulk phase. The repulsive forces between phenolic compounds on the solid surface and the bulk phase cause the remaining vacant sites on the surface difficult to occupy [38]. This situation inhibit further adsorption of p-NP onto the adsorbent (region III). Furthermore, due to steric hindrance, p-NP took longer to diffuse into small pores [13].

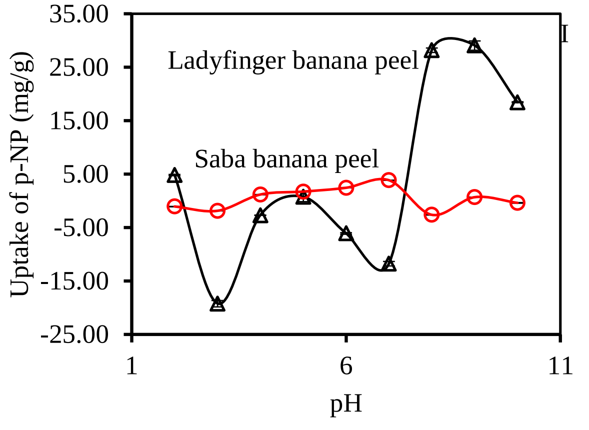


Figure 5. Effect of pH on adsorption of p-NP onto Saba and Ladyfinger banana peels.

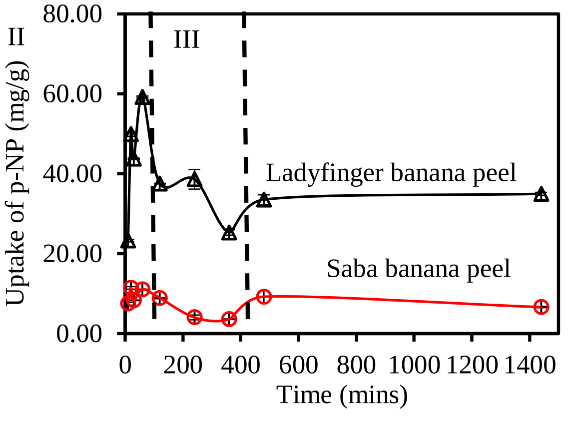


Figure 6. Equilibrium contact time on adsorption of p-NP Saba and Ladyfinger peels in aqueous medium.

Figure 7 shows the pseudo-first and second-order kinetic model plot. The scattered point ($r^2 < 0.1$) showed that Saba banana peel does not fit with pseudo first order kinetic model compared to Ladyfinger banana peel. As a result, the adsorption of p-NP on banana peels in aqueous solution did not obey the first pseudo kinetic model. The pseudo-second-order kinetic model is the best to represents the adsorption of p-NP onto both banana peels with $r^2 > 0.9$ (Table 2).

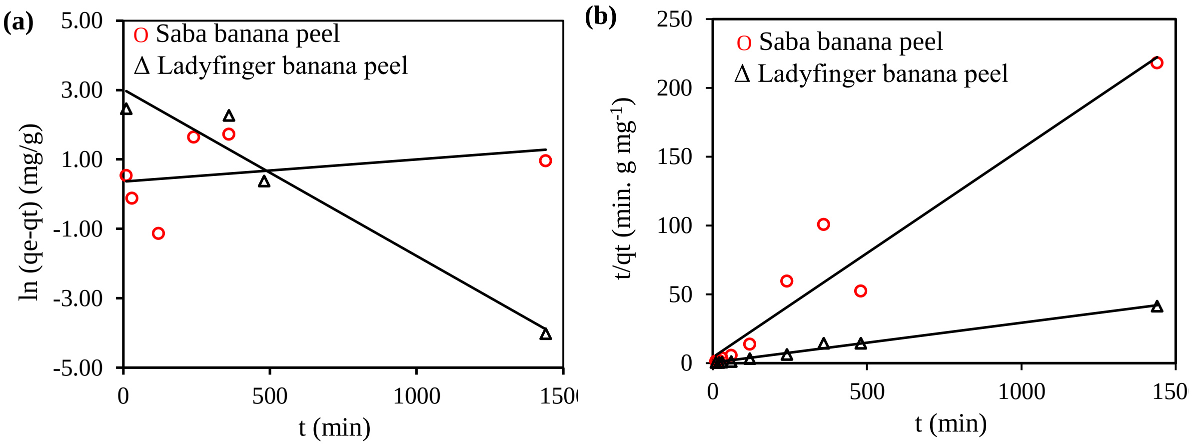


Figure 7. Kinetic model pseudo-first (a) and pseudo-second (b) order for the adsorption of p-NP onto banana peels.

Table 2. Pseudo first and second order parameter for the adsorption of p-NP onto Banana Peels

Banana peels	Pseudo first order			Pseudo second order		
	q_e (mg/g)	k_1	r^2	q_e (mg/g)	k_2	r^2
Saba banana	1.44	6.0×10^{-3}	0.099	6.605	1.51×10^{-1}	0.928
Ladyfinger banana	20.48	4.80×10^{-3}	0.950	34.482	4.43×10^{-3}	0.988

The adsorption isotherm study of Langmuir and Freundlich plot is shown in Figure 8, and the parameters for both isotherms are tabulated in Table 3. As seen in the graph, the adsorption data for both banana peels are well fitted with Freundlich ($r^2 > 0.90$) as compared to Langmuir isotherm ($r^2 < 0.9$). Ladyfinger has a higher k_F value than Saba banana peel, indicating that p-NP has a higher adsorption tendency for Ladyfinger than Saba banana peel. The k_F is related primarily to the capacity of the adsorbent for a given molecule [24]. The lower value of the heterogeneity factor (1/n) for Ladyfinger, which is close to 1.0, indicates that p-NP adsorption on Ladyfinger is better than Saba banana. The magnitudes of k_F and 1/n suggest that p-NP is easily removed from the aqueous solution and that adsorption is favourable. Despite the fact that Saba banana peels have a larger surface area, the uptake is not as great as predicted. This phenomenon suggests that the surface of the Saba banana peel is more negative than the Ladyfinger banana peel, which raises the p-NP repulsion barrier and reduces adsorption uptake.

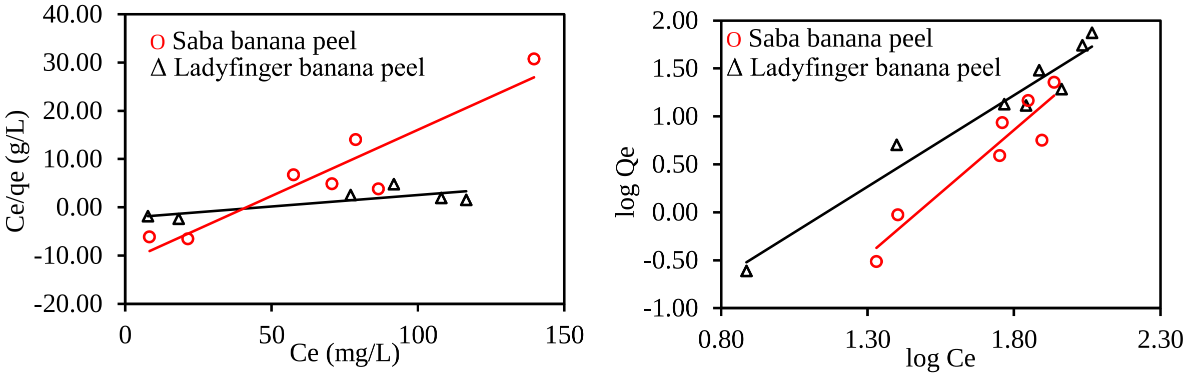


Figure 8. Langmuir (a) and Freundlich isotherm for the adsorption of p-NP onto Saba and Ladyfinger banana peels

Table 3. Langmuir and Freundlich parameters for adsorption of p-NP onto banana peels

Banana Peels	**Langmuir**		**Freundlich**	
	q_m (mg/g)	b (L/mg)	k_F (L/mg)	1/n
Saba banana	3.65	-0.0241	0.0001	2.62
Ladyfinger banana	21.01	-0.0214	0.0061	1.91

Conclusion

The adsorption of p-NP in an aqueous medium over Saba banana peels is well fitted by a pseudo-second-order kinetic model. Ladyfinger banana peels, meanwhile, fitted with both pseudo kinetic models. The hydrogen bonding/donor-acceptor/lewis acid-base interaction was involved to adsorb p-NP onto banana peels. The Freundlich isotherm revealed that p- NP adsorption in aqueous solution was more favourable on Ladyfinger banana peels than Saba banana peels. Thus, Ladyfinger banana peels are an effective adsorbent for the removal of p-NP from aqueous solutions.

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Author contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of conflict of interest

The authors have no disclosures to declare.

Compliance with ethical standards

The work is compliant with ethical standards.

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