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Removal of Paracetamol by Powdered Activated Carbon Synthesized From Orange Peels

İrem KONUK AKÇA [1], Rabia KÖKLÜ [*1]

Abstract

This study aims to investigate the removal of Paracetamol active ingredient from aqueous solutions with the use of powdered activated carbon obtained by $ZnCl_2$ activation of orange peels. Equilibrium values of initial paracetamol concentration (100-500 mg L-1), pH (2-10), adsorbent dose (10-500 mg) and contact time (5-120 minutes) parameters in the removal of paracetamol from aqueous solutions are evaluated. The adsorption mechanism of paracetamol is explained with the kinetic models. The highest correlation among Langmuir, Freundlich, Temkin, and Dubinin-Radushkevichi isotherms applied to experimental data was determined as Freundlich isotherm with R^2 =0.95. Pseudo-first-order and pseudo-second-order kinetic models were applied, and it was found that the latter, whose correlation coefficient is determined as R^2 =0.99, is the best model to explain paracetamol adsorption. As a result of this study, it can be seen that powdered activated carbon synthesized from orange peel is an effective adsorbent in the removal of paracetamol and can be easily applied thanks to its low cost.

Keywords: Paracetamol, powdered activated carbon, orange peel, removal

1. INTRODUCTION

In parallel with the rapid population growth in the world, the diseases that people are exposed to also increase by diversifying. In line with this increase, much more effective and faster treatment methods have been introduced with renewed technologies to sustain the supply/demand relationship in the health sector [1]. Consequently, drug use, which is only one of these treatment methods, has increased proportionally and reached significant levels [2]. The proliferation of medical drug use leads to environmental problems as well. Along with their environmental impacts, medical drugs have other impacts such as hormonal effects on aquatic organisms, gender differentiation, and a decrease in the population of organisms, as they are designed to preserve their chemical structures for a long time [3]. Various studies gained momentum in many countries upon the frequent occurrence of drug active substances in environmental areas such as aquatic ecosystems and soil [4].

Medical drugs with active ingredients such as analgesics, antibiotics, antidepressants, and antipyretics are partially or fully

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metabolized after being used in the humanbody and are discharged into wastewater through urine, stool, sweat glands, and tears [5]. At this point, it is treated with other organic and inorganic components in the wastewater or mixed with the underground and surface waters by discharge without being treated. Some measures are taken to reduce the discharge-related increase in concentrations of medical drugs in aquatic environments, increase water quality, and prevent adverse effects that may occur in ecology [4]. In recent years, various removal methods have been developed to remove resistant drugs, including chemical oxidation with ozone [6] and ozone/hydrogen peroxide [7], membrane filtration [8] and activated carbon adsorption [9] and the effectiveness of these methods have been evaluated.

Advanced treatment technologies are used to completely remove chemicals such as medical drugs from water. Activated carbon adsorption [10] is among these technologies. In addition to being a low-cost method, adsorption is more commonly preferred due to its efficiency in removing pollutants with low concentrations [11]. Activated carbons can be obtained from various raw materials such as fruit peel wastes [12] and seeds [13], nut shells [14] and coconut shells [15]. Özer and İmamoğlu [16] researched the removal of ciprofloxacin antibiotics from aqueous solutions using biochar from pumpkin peel activated with H_3PO_4. Similarly, Özer and İmamoğlu [17] investigated the removal of nickel (II) and lead (II) ions from aqueous solutions with the use of biochar, which is formed by pumpkin peel activated by H_2SO_4. In the removal of ions such as copper (II), nickel (II), and cobalt (II) from aqueous solutions, Usanmaz et al. [18] used hazelnut shell-based activated carbon with H_3PO_4.

There is a large number of studies of activated carbon synthesized from orange peel. Even the orange peel study with $ZnCl_2$ is more than a few. In addition, activated carbon can be produced from orange peel by using KOH and H_3PO_4 by chemical and hydrolysis [19]. In former studies, orange peel activated carbon was used in the removal of pollutants such as pharmaceuticals [20], dyestuff [21], and micropollutants [22] from the aquatic environment.

The present study investigates the removal of paracetamol (PSM), which is a non-prescription drug in Turkey and is widely used for analgesic and antipyretic purposes, from the aquatic environment by adsorption method. To this end, the orange peel was activated with $ZnCl_2$ and used as powdered activated carbon. In this study, the effects of initial PSM concentration, adsorbent dose, solution pH, and mixing time parameters are investigated. Isotherm models and kinetic properties were determined. This study aims to contribute to the removal of the analgesics in question and their types from the wastewater that come out of the conventional wastewater treatment plants.

2. MATERIAL METHOD

2.1. Instruments and Chemicals

Paracetamol (PSM), a drug active ingredient used in the experimental study was purchased from Neutec Pharma Industry and Trade Inc. Methanol (CH_3OH) (Merck, Germany) was used for PSM dissolution. Hydrochloric Acid (HCl) and Sodium Hydroxide (NaOH) (Merck, Germany) were used for pH adjusting in the process. UV-VIS spectrophotometer (Merck Pharo 300, Germany) was used to determine the concentration remaining in solution after treatment. The solutions were mixed with a multi-magnetic stirrer (Biosan ES-20, Latvia). pH measurements throughout the process were made with a pH meter (ISOLAB 8200M, Germany). The surface morphology of powdered activated carbon orange peel (PACOP) was investigated by scanning electron microscopy (SEM) (FEI, Quanta FEG 250, USA). Surface

characterization of PACOP was determined by Brunauer-Emmett-Teller (BET) analysis (Quantachrome Corporation, Autosorb-6, USA). CHN element analysis was conducted with Leco CHNS 932 (LECO Corporation, St. Joseph, MI) analyzer.

2.2. Preparation of Activated Carbon From Orange Peel

Briefly, the oranges obtained from the local markets were peeled, then the peels were washed and dried in an oven; 90 g of orange peel, 90 g of $ZnCl_2$ and 150 ml of distilled water were added and mixed. It was kept at room temperature for 24 hours and dried in an oven at 105^0C for 24 hours and pyrolyzed for 1 hour under N_2 flow (100 ml min^{-1}) in a tube furnace at 700^0C. After these processes, it was thoroughly washed with 2 M HCl and then with distilled water and dried at 105^0C [23]. The powdered activated carbon (<212 μm) derived from orange peel was used in this study.

2.3. Adsorption Studies

Lorem ipsum dolor sit amet, consetetur sadipscing elitr, sed diam Experiments were carried out for the adsorption of PSM active ingredient on PACOP derived from orange peel and the results were evaluated. In the experiment, the effects of parameters such as initial PSM concentration, pH, adsorbent dose and contact time on PSM removal were investigated. First, 500 mg of pure PSM was used to create a synthetic solution with a concentration of 500 mg L^{-1}. PSM was dissolved using 5 mL of CH_3OH and 100 °C distilled water. In hot water, paracetamol is more soluble as compared to cold water. In this reasons, was used boil water. The stock solution was prepared by completing the solution volume to 1000 mL with distilled water at room temperature. Effect of parameters such as initial PSM concentration (100-500 mg L^{-1}), pH (2-10), adsorbent dose (10-500 mg) and contact time (5-120 minutes) evaluated for the removal of paracetamol from aqueous solutions.

The remaining PSM concentration in the equilibrated solution was measured with a UV Spectrophotometer at a wavelength of 290 nm. Maximum absorbance value of PSM was measured as 290 nm in UV Spectrophotometer. The amount of PSM adsorbed on a unit of PACOP was calculated through the equation below.

$$q_e = \frac{(C_0 - C_e).V}{m} \quad (1)$$

$$\text{Adsorption}, \% = \left(\frac{C_0 - C_e}{C_0}\right).100 \quad (2)$$

where the initial and equilibrium PSM concentrations are given as C_0 and C_e (mg L^{-1}), solution volume V (L), and PACOP amount m (g), respectively [24].

The compatibility of the experimental results to Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich adsorption isotherms was evaluated. The Langmuir isotherm equation is given with Eq. (3);

$$q_{max} = \frac{C_e}{q_e} = \frac{1}{K_L} + \left(\frac{a_L}{K_L}\right)C_e \quad (3)$$

where q_{max} is the maximum adsorption capacity (mg g^{-1}), Ce is the equilibrium concentration of the solution (mg L^{-1}), and K_L is a Langmuir constant [25].

The Freundlich isotherm equation is given with Eq. (4);

$$q_e = K_F C_e^{\,1/n} \quad (4)$$

where K_F is a Freundlich constant associated with the adsorption capacity and 1/n is an empirical parameter associated with the adsorption intensity [26].

The Temkin isotherm equation is given with Eq. (5);

$$q_e = \frac{RT}{b_T} \ln K_T + \frac{RT}{b_T} \ln C_e \quad (5)$$

where K_T is a Temkin constant associated with the adsorption capacity and b_T are both related to the adsorption heat [27].

The Dubinin-Radushkevich isotherm equation is given with Eq. (6);

$$\ln q_e = \ln q_m - \beta \varepsilon^2 \quad (6)$$

where β is a Dubinin-Radushkevich constant with the adsorption capacity. q_m is the theoretical monolayer saturation capacity of the adsorbent and ε is Polanyi potential [28].

3. RESULTS AND DISCUSSION

3.1. Adsorption Studies

The BET surface area of the PACOP was determined as 1570 m^2 g^{-1}. Besides, the pore volume and pore width were 0.94 cc g^{-1} and 12.2 Å, respectively. According to the results of the elemental analysis, PACOP had high carbon (78.3%), low hydrogen (2.12%), and nitrogen (2.95%) content. Fernandez et al. [29] reported that they observed high carbon, low hydrogen, and nitrogen content by weight in the elemental analysis of the activated carbon they derived from orange peel in their study.

3.2. Effect of pH

The pH of the adsorption also plays an important role in the adsorbent surface charge, along with its effects on the various ionic form exhibited by the adsorbate species [9]. For this reason, the pH change effect of the solution is accepted as an important parameter that affects the analyte charge and adsorbent surface in the interactions between the adsorbent and the drug active substance [30]. To determine the pH effect on adsorption, PSM solutions with 50 mg PACOP, 50 mL volume, and 300 mg L^{-1} concentration were studied between pH values of 2-10. pH values were adjusted using 0.1 M HCl and NaOH.

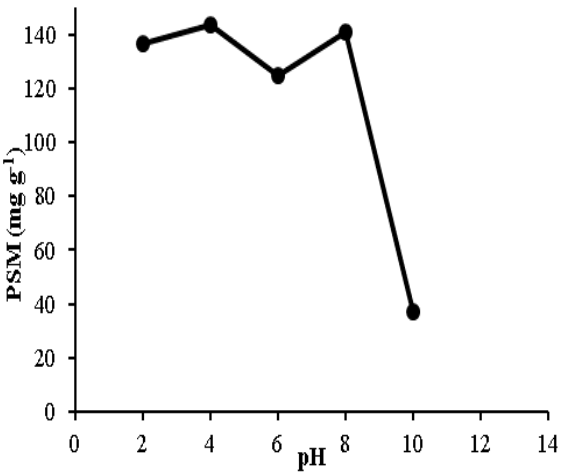


Figure 1 Effect of pH on the removal of PSM by PACOP

Figure 1 shows the effect of solution pH varying between 2 and 10 on the adsorption of PSM. It was observed that high adsorption values were obtained between pH 4 and pH 8 values. A decreasing trend in adsorption was observed above pH 8. As the pH increased towards 10, the PSM (pKa = 9.38) removal capacity of PACOP decreased again. When the pH of the solution is lower than the pKa (pH<pKa), the active ingredient is usually in a non-ionized, that is, negative form [31]. The decrease in PSM adsorption at pH values above 8 may be due to the electrostatic repulsion between the anionic PSM molecules and the negatively charged functional groups on the PACOP surface [32]. In their study on the adsorption of PSM and two other drug active ingredients, Streit et al. [33] reported that the PSM removal capacity increased gradually from pH 2 to 8 and decreased significantly at pH 9. For this reason, they preferred pH 8, which is the closest to neutral pH in their studies. In this study, pH 6 value was preferred in terms of ease of applicability for balance and kinetic studies.

3.3. Effect of Contact Time

To investigate the effect of contact time on the adsorption of PSM with the powdered activated carbon, PSM solutions with pH 6, 50 mg PACOP, 50 mL volume, and 300 mg

L^{-1} concentration were mixed at 180 rpm for 5 -120 min.

As presented in Fig. 2, adsorption took place rapidly between 5-60 min. and after 60 min. it reached equilibrium and remained stable. No significant difference was observed in adsorption efficiency at 90 and 120 min. 60 minutes was recorded as the time for PSM adsorption with activated carbon to reach equilibrium at 25 °C. The adsorption capacity for 300 mg L^{-1} PSM solution at equilibrium time was determined as 124.6 mg g^{-1}. This finding supports the theory that the adsorption rate is high at the first contact of PACOP and PSM solution, and it decreases with time [34]. In their study, Özer and İmamoğlu [35] explained that the deceleration of the adsorption rate is due to the inability of more adsorption molecules to enter the activated carbon, which has reached saturation in its inner pores.

3.4. Effect of Adsorbent Dosage

The adsorbent dose is an important parameter for the applicability evaluation of the adsorption process. To investigate the effect of adsorbent dose on PSM removal, solutions with 300 mg L^{-1} concentration, original pH value (pH 6), and 50 mL volume were prepared with the addition of 10-500 mg PACOP. In Fig. 3, the amount of PACOP in the solution increased while the adsorption capacity decreased.

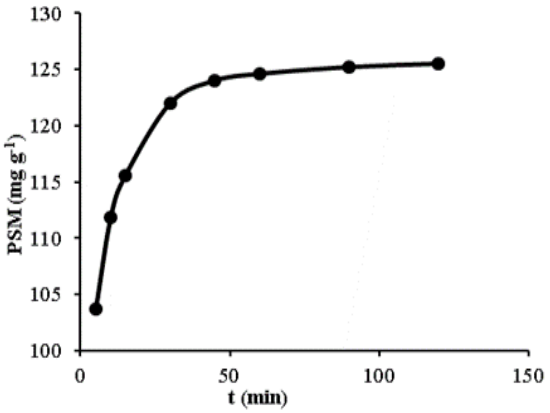


Figure 2 Effect of contact time on the removal of PSM by PACOP

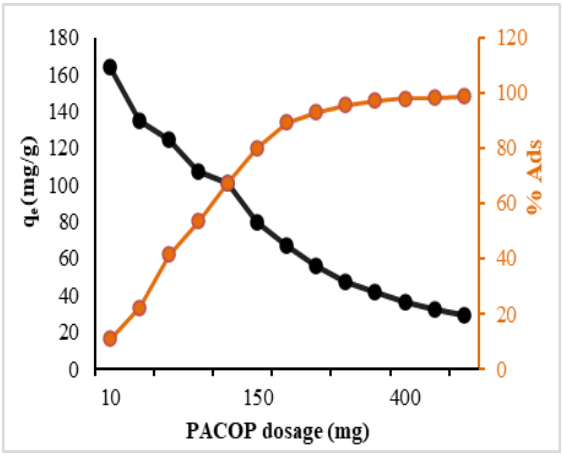


Figure 3 Effect of dosage on the removal of PSM by PACOP

The adsorption removal percentage increased in direct proportion to the adsorbent dose, contrary to the adsorption capacity. The direct proportional increase in the amount of adsorbent and the adsorption removal percentage can be explained by increased adsorption sites due to the increase in the amount of activated carbon [36]. In another study, Sajid et al. [37] used activated carbon synthesized from the CSH (Cannabis Sativum Hemp) plant for PSM removal in the aquatic environment. In this study, the adsorption efficiency increased with the increase in the adsorbent dose. Sajid et al. [37] explained the decrease in adsorption capacity depending on the number of active sites.

3.5. Effect of Initial Concentration

To determine the effect of the initial PSM concentration on adsorption, 50 mg PACOP at pH (6) of the original solution was mixed with a 50 mL solution volume at initial concentrations of 100, 200, 300, 400, and 500 mg L^{-1} with a contact time of 60 minutes.

As presented in Fig 4, it was observed that the adsorption capacity increased in direct proportion to the increase in PSM concentration. This increase results from the increased interaction between PSM and PACOP [38]. The decreased adsorption removal percentage indicates that the

PACOP surface is insufficient to adsorb PSM in the empty active sites [39].

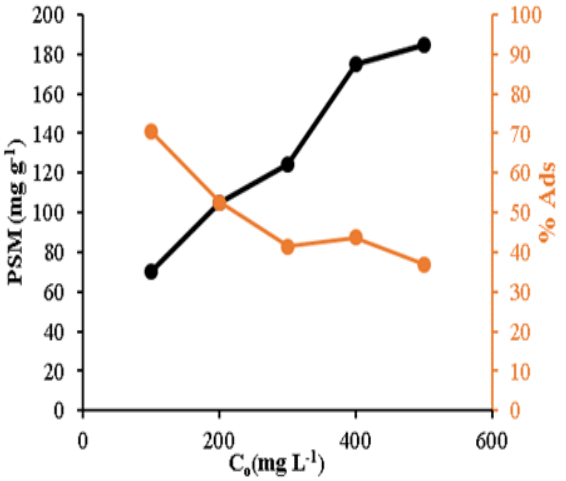


Figure 4 Effect of initial concentration of PSM on its removal

3.6. PSM Adsorption Isotherms

The relationship between adsorbent and adsorbate was determined using Langmuir, Freundlich, Temkin, and Dubinin-Radushkevich isotherm models (Fig 5).

In order to determine the compatibility of the adsorption isotherms, the R^2 values of the experimental results obtained according to the initial concentrations were compared. Table 1 presents isotherm model constants and correlation coefficients of the adsorption of PSM on powdered activated carbon synthesized from orange peel. Accordingly, the R^2 value is 0.95 and the n value is 2.41 for the Freundlich isotherm. The correlation coefficient and RL value greater than 1 indicate that the adsorbent will adsorb the PSM active substance effectively. According to the results, the best fit isotherm model was found to be the Freundlich Isotherm. In the literature, Nourmoradi et al. [40] determined that the most compatible model for PSM removal from aqueous solutions with activated carbon derived from acorns is the Freundlich isotherm model with $R^2 = 0.99$. This finding shows that the activated carbon surface synthesized from the orange peel is heterogeneous. Besides, the K_F constant showing a value as high as 16.61 indicate that the adsorption process is easy, and the adsorbent has a high adsorbing potential [41].

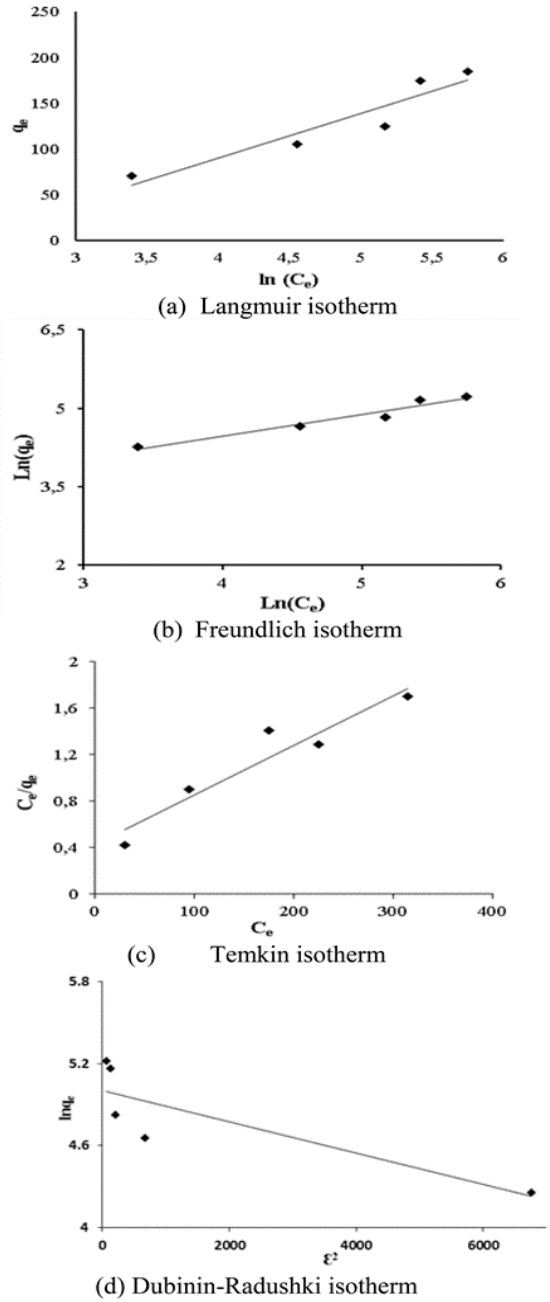


Figure 5 Linear isotherm graphs for PSM adsorption onto PACOP

Table 1 Langmuir, freundlich, temkin, dubinin-radushki isotherms constants and correlation coefficients for PSM adsorption onto PACOP

Isotherms	Isotherm Constants		R^2
Langmuir	q_{max}=238.10	K_L= 0.01	0.91
Freundlich	K_F= 16.61	n=2.41	0.95
Temkin	b_T=48.53	K_T =0.12	0.88
Dubinin-Radushkevich	ß= 0.0001	q_m=148.8	0.71

3.7. Adsorption Kinetics

PSM adsorption kinetics were calculated through pseudo-first-order and pseudo-second-order kinetic models and linear curves as presented in Fig 6. Model equations and calculated coefficients are presented in Table 2.

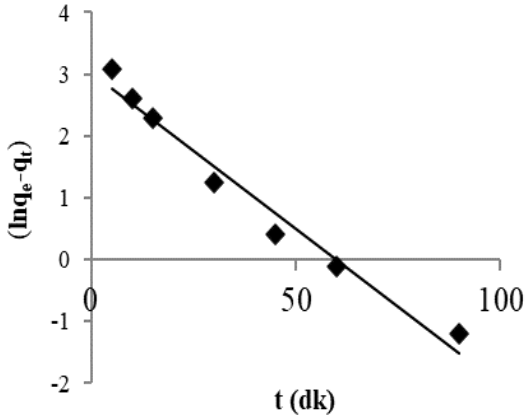
The estimated qe value according to the pseudo-second-order kinetic model was found close to the experimental qe value. The correlation coefficient of the pseudo-second-order kinetic model was calculated as 0.99.

According to Table 2, it was found that the 0.99 value of the R^2 coefficient was obtained in the pseudo-second-order kinetic model.

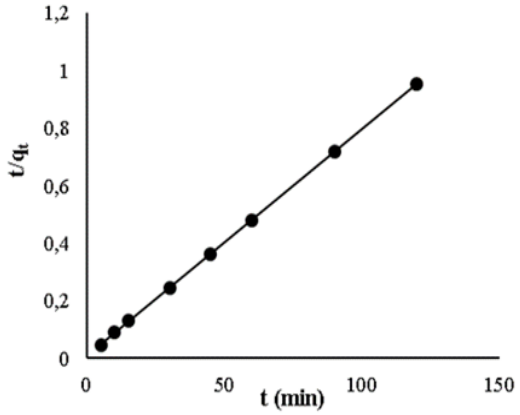
Table 2 Kinetics coefficients for PSM adsorption onto PACOP

Kinetic model	Constants		Ref.
Pseudo first order model	k_1	$5*10^{-2}$	[43]
	q_e $(mg\ g^{-1})$	20,64	
	R^2	0.97	
Pseudo second order model	k_2	$6{,}1*10^{-3}$	[44]
	q_e $(mg\ g^{-1})$	126.58	
	R^2	0.99	

This value gives the result that the absorption of the PSM active substance on the PACOP surface is more compatible with the pseudo-second-order kinetic model.



(a) Pseudo first order model



(b) Pseudo second order model

Figure 6 Kinetics graphs for PSM adsorption onto PACOP

At the same time, the closest result to the experimental qe value in PSM adsorption on PACOP is the qe value calculated in the pseudo-second-order kinetic model, which also supports the compatibility of the adsorption kinetics with the pseudo-second-order kinetic model. Wong et al. [9] reported the compatibility of the kinetic model of PSM removal with activated carbon derived from waste tea leaves. Hashemian et al. [42] also conducted kinetic studies on the adsorption of activated carbon derived from orange peel and almond peel and found that the result was compatible with the pseudo-second-order kinetic model.

Table 3 presents the properties of various activated carbons derived from different

natural materials, activated by different methods, in PSM adsorption to compare the adsorption capacity of the powdered activated carbon used with other adsorbents.

Table 3 Comparison of the PSM adsorption studies

Activated carbon	Activation	Isotherm/ Kinetic
Waste tea leaf	H_3PO_4	Langmuir/ Pseudo second order
Acorn	KOH	Freundlich/ Pseudo second order
Commercial activated carbon	-	Langmuir / Avrami fractional degree
Rice husk	$ZnCl_2$ NaOH	Langmuir and Redlich-Peterson / Pseudo second order
Coffee bark Brazil	$ZnCl_2$ NaOH	Langmuir and Redlich-Peterson / Pseudo second order
Nut shell	$ZnCl_2$ 1.0:1.0	Liu / Avrami fractional degree
Orange peel	$ZnCl_2$	Freundlich/ Pseudo second order

Taking stand from the data presented in Table 3, the lowest value of PSM capacity was observed at 45.45 mg g^{-1} in the adsorption study of Nourmoradi et al. [40] conducted with acorn activated carbon; the highest value of 309.7 mg g^{-1} was observed in the adsorption study of Lima et al. [14] conducted with Brazil nutshell activated carbon. In the present study, the result of 185 mg g^{-1} recorded in PSM adsorption with orange peel activated carbon is in the range of the two highest and lowest values.

Table 4 Comparison of the PSM adsorption studies

Qmax (mg g^{-1})	pH	Contact Time (min)	Ref.
59.17	3	60	[9]
45.45	3	150	[40]
221	7	5	[10]
50.25	5.8	60	[45]
48.31	5.8	60	[45]
309.7	7	30	[14]
185	6	60	This study

4. CONCLUSIONS

In the adsorption study of the PSM drug active substance, which was conducted with activated carbon synthesized from orange peels, various parameters were calculated in the process. The adsorption mechanism was explained using equilibrium and kinetic models under certain conditions (pH 6, 300 mg L^{-1} PSM, 50 mg PACOP). Among the adsorption isotherms (Langmuir, Freundlich, Temkin and Dubinin-Radushkevich) applied under experimental conditions, it was seen that the most compatible model was the Freundlich model (R^2 = 0.95). The results of the modelling study performed in the kinetic evaluation were examined by applying applied to pseudo-first-order and pseudo-second-order kinetic models for the evaluation. As a result of the correlation coefficients of the models, it was determined that the process was suitable for the pseudo-second-order kinetic model. Finally, in consideration of these analyses, it was concluded that PACOP synthesized from orange peel bears the capacity to remove PSM drug active substance from aqueous solutions in PSM adsorption on activated carbon. The findings obtained show that activated carbon synthesized from orange peel is a compatible adsorbent. This feasible method can be used to overcome the pharmaceutical problem in wastewater as it ensures an

effective removal both economically and as an adsorbent.

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Authors' Contribution
The authors contributed equally to the study.

The Declaration of Conflict of Interest/ Common Interest
No conflict of interest or common interest has been declared by the authors.

The Declaration of Ethics Committee Approval
This study does not require ethics committee permission or any special permission.

The Declaration of Research and Publication Ethics
The authors of the paper declare that they comply with the scientific, ethical and quotation rules of SAUJS in all processes of the paper and that they do not make any falsification on the data collected. In addition, they declare that Sakarya University Journal of Science and its editorial board have no responsibility for any ethical violations that may be encountered, and that this study has not been evaluated in any academic publication environment other than Sakarya University Journal of Science.

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