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Proceeding Paper

Adsorbents Derived from Plant Sources for Caffeine Removal: Current Research and Future Outlook †

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Abstract: Pharmaceutical wastes, due to their recalcitrant nature, are emerging contaminants in wastewater that have been the focus of researchers and scientists. One pollutant of interest is caffeine, which is one of the most detected contaminants in a global context. Although commonly present in beverages such as coffee, caffeine can be harmful to both humans and animals when disposed of in water bodies. Current wastewater treatment approaches not only display ineffective results in removing the mentioned pollutant but also entail high financial costs in applying the treatment technology. Recent studies have revealed the potential of adsorbents derived from plant sources such as husks, fruit peels, and other plant fibers from biomass to effectively reduce caffeine concentrations in wastewater, with a removal efficiency in the range of 8.04 to 171.23 mg/g. Moreover, the adsorption phenomena exhibited a Langmuir isotherm model and pseudo-second-order kinetics. This review paper aims to systematically present and analyze the current literature and prospects of utilizing plant-based adsorbents in addressing the impact of caffeine on the environment. Specifically, the review will focus on the efficiency of said adsorbents in removing caffeine, considering the specific surface area, adsorbent dosage, pH level, maximum adsorption capacity, adsorption isotherms and kinetics, and the predicted optimum conditions for adsorption. The objective is to identify the most suitable adsorbents to be used in wastewater treatment plants. This study will serve as a valuable reference for future research.

Keywords: adsorbent; plant sources; caffeine; adsorption; wastewater

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1. Introduction

Pharmaceutical and personal care products are emerging contaminants in water bodies that can disrupt the physiological processes of organisms, including humans [1]. Among these pharmaceutical and personal care products, a psychoactive drug called caffeine is one of the most detected globally. The said stimulant is found in several plant products, including coffee beans, tea, and cacao. More commonly, the said drug is known to block receptors for the inhibitory neurotransmitter adenosine, stimulating brain activity [2]. Hence, approximately 80% of the world's population consumes a caffeinated product daily, stimulating the central nervous system, energizing the body temporarily, and restoring alertness [3]. However, caffeine is not entirely broken down in the body, making its way into residential wastewater.

Caffeine contamination in various aquatic environments, including seawater in the USA, the Aegean Sea in Europe, and Darwin Harbour in Northern Australia, is a growing concern due to its potential adverse effects on wildlife. Coastal and marine ecosystems have been found to contain significant concentrations of the drug. Some of the impacts on several coastal species include growth inhibition, hindered reproduction, altering metabolic activity, neurotoxic effects, oxidative stress, cellular damage, and lethality [4].

Caffeine has also been found in wastewater treatment plants' influents and effluents. However, due to their physicochemical properties, conventional wastewater treatment plants may not be effective in removing pharmaceutical products like caffeine [5]. Only about 30% of pharmaceutical products in wastewater streams are removed by treatment plants, leading to high concentrations of these substances in effluent streams. Therefore, searching for alternatives to remove caffeine from wastewater is essential. Various treatments, such as biochemical degradation, photolysis, reverse osmosis, ozonation, advanced oxidation, and adsorption, have been used for caffeine removal [6].

Among the mentioned alternatives, adsorption has become widespread due to its simple process, low consumption, and convenient operation. Various materials such as clay minerals, polymers, zeolites, metal–organic frameworks, and activated carbons (ACs) have been reported to be effective adsorbents for caffeine removal [7]. However, it is the utilization of AC that has been widely investigated in this field.

AC is a popular adsorbent due to its high porosity, specific surface area (SSA), and affinity for various chemical substances [8]. The said properties enhance caffeine's binding on its surface through hydrophobic π-π interaction and electrostatic and non-electrostatic interaction with carboxyl and hydroxyl groups [1]. However, most studies have only considered single adsorbates, and more research is needed to understand competitive adsorption between multiple adsorbates. There have been some studies on the competitive adsorption of pharmaceutical drugs on AC. However, again, more research is still necessary to understand adsorption mechanisms in complex systems, including mixtures of drugs [8].

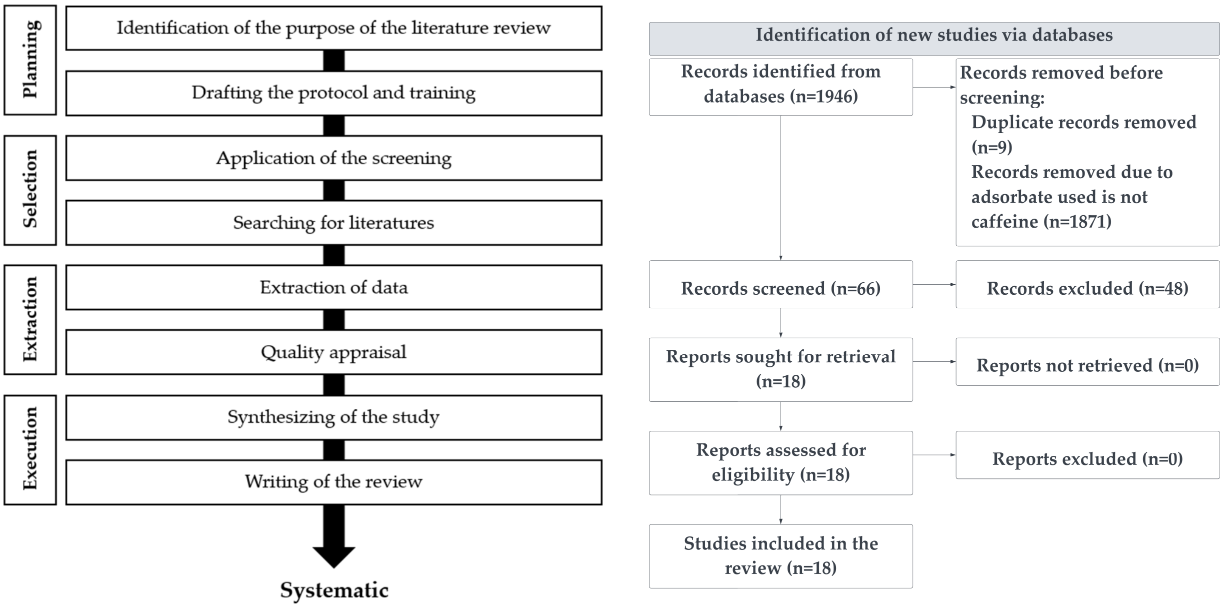
New materials with lower investment in production are needed due to the high cost of commercially available products, which limits their large-scale use [9,10]. Therefore, several non-conventional adsorbents, such as polymeric resins, nanomaterials, and biochar, have been used to remove emerging contaminants from aqueous environments [11]. In addition, agricultural residues such as husk, shells, peels, leaves, fruit seeds, and stalks have also been used either as adsorbents or as precursors to AC synthesis due to their inexpensive, renewable, and high carbon content characteristics [9,12]. New-generation adsorbents, such as nanoparticles and composites, are also being explored as an alternative. Overall, the adsorbent material to be selected should exhibit desirable characteristics such as selectivity, high surface area, high adsorption capacity, low cost, long service life, and recyclability [11].

This study focused solely on caffeine adsorption using adsorbents made from plant sources. It considered SSA, adsorbent dosage, pH level, maximum adsorption capacity (MAC), the adsorption isotherms, and kinetics. Research papers meeting these specific criteria were reviewed, and recent review articles supporting the results were scrutinized. For accuracy and objectivity, all cited research papers were published between 2000 and 2025.

2. Methodology

A review was conducted to identify research trends on caffeine adsorption from aqueous solutions, focusing on adsorbent materials derived from plant sources. The study involved a systematic literature review adapted by Okoli (2015) to assess relevant publications [13], which includes planning, selection, extraction, and execution, with all results extensively analyzed for inclusion in the study, as seen in Figure 1a.

The planning stage of the systematic review involved identifying relevant key-words, selecting publication dates, and utilizing databases to filter papers automatically. The papers that pass this initial screening are manually checked to ensure that they meet the review criteria. Following this, data extraction and cross-referencing techniques are employed to identify recurring patterns and commonalities within the literature. Finally, the data retrieved from the papers are used to conduct the review, highlighting relevant trends and common factors.



(**a**) (**b**)

Figure 1. Adapted systems and methodologies: (**a**) systematic literature review adapted from Okoli (2015) [13]; (**b**) adapted PRISMA diagram from Page et al. (2021) [14].

The references were obtained from ScienceDirect with the search equation "caffeine" AND "type" AND "adsorption isotherm". The "type" part of the equation was interchanged with the following terms: fruit, fiber, stalk, algae, agricultural, wood, fungi, vegetable, and husk to cover as many relevant references as possible. A total of 1946 studies combined were found and filtered using the Preferred Reporting Items for Systematic reviews and Meta-Analyses (PRISMA) guidelines [14], as seen in Figure 1b. Upon examining the article title and removing duplicates, 66 out of 1946 results remained, emphasizing caffeine. Abstracts were examined for eligibility of the references for the study, and 18 out of 66 papers remained. Finally, after screening, retrieval, and another examination of eligibility, 18 publications were used to prepare the data for this review.

The review of SSA, adsorbent dosage, pH level, and MAC was conducted to determine the most effective adsorbent derived from plant sources for caffeine adsorption. The common trend in the adsorption process was determined by reviewing the adsorption isotherms and kinetics.

3. Results and Discussion

The 19 publications examined resulted in 35 adsorbents analyzed for the study due to other publications experimenting with multiple adsorbents. The 35 adsorbents were investigated according to the following parameters in Table 1: SSA, adsorbent dosage, pH, MAC, isotherm, and kinetic models.

Table 1. Summary of properties of different adsorbents derived from plant sources and parameters of the adsorption process.

Adsorbent	SSA (m^2/g)	Dosage (g/L)	pH	MAC (mg/g)	Adsorption Isotherm	Adsorption Kinetics	Reference
Orange Peel (Orange)	0.801	3.5	6.9	15.188	LM	PFO	[11]
Banana Peel (Banana)	0.079	9.5	6.9	6.761	LM/SPs	PFO	
Orange Peel Composite (Orange)	14.282	2.5	6.9	25.604	LM/SPs	PFO	
Banana Peel Composite (Banana)	8.140	5.5	6.9	11.668	LM	PSO	

Table 1. *Cont.*

Adsorbent	SSA (m^2/g)	Dosage (g/L)	pH	MAC (mg/g)	Adsorption Isotherm	Adsorption Kinetics	Reference
KAC (Chichá-do-cerrado)	420.46	1	7	391	FL	PSO	[15]
CAC (Chichá-do-cerrado)	53.92	1	7	139.61	FL	PSO	
CH-KAC (Chichá-do-cerrado)	1082.41	1	7	121.9	LM	PSO	
CH-CAC (Chichá-do-cerrado)	240.79	1	7	39.53	LM	PSO	
CA-SA 400/10 (Açaí)	1150.3459	1	7	176.8	-	PSO	[16]
Pineapple ACF (Pineapple)	1031	1	5.8	152.18	LM	PSO	[17]
Peanut shell AC (Peanut)	790	0.05	5	0.63 mmol/g	LM	-	[7]
Peanut shell AC (Peanut)	790	0.05	7	1.11 mmol/g	LM	-	
GS (Grape)	6.23	25	2	89.194	SPs	-	[9]
MGS (Grape)	4.21	15	2	129.568	SPs	-	
GSAC (Grape)	1099.86	1	4.0	916.679	SPs	-	
NS900 (Norway spruce)	167.71	0.25	7	9.24	LM	PSO	[18]
WP900 (White pine)	156.08	0.25	7	11.85	LM	PSO	
SAC (Sargassum)	754	0.6	6	221.61	LM	PSO	[19]
ABC (Macrophyte)	740	1	6	117.8	LM	PSO	[20]
TWBC-SA (Tea)	576	1	3.5	15.4	FL	ELV	[21]
TWPC-800 (Tea)	2260.82	2.5	-	491.37	LM	PSO	[22]
AC_{C03} (Custard apple)	431.31	0.8	4	171.23	LM	PSO	[23]
RG (Rice husk)	63	1	4	8.04	LM	PSO	[24]
CW-C-1-800 (Coffee waste)	1212	0.2	5	274.2	LM	PFO/PSO	[1]
MBC1 (Green coconut)	474	1	-	45	SPs	PSO	[25]
ACP (Green coconut)	1242	1	-	259	SPs	PSO	
MNC (Green coconut)	1019	1	-	168	SPs	PSO	
GBC300 (Gliricidia sepium)	1.02	1	4.5	-	FL/TK	ELV/FTP	[26]
GBC500 (Gliricidia sepium)	76.30	1	4.5	-	FL/TK	ELV/FTP	
GBC700 (Gliricidia sepium)	216.40	1	4.5	16.26	FL/TK	ELV/FTP	
Pi/1:1/800/2 (Pine wood)	945	6 mg	5	500	LM	PSO	[27]
Pi/1:3/800/2 (Pine wood)	1509	6 mg	5	476.2	LM	PSO	
YC (Yeast)	823	0.6	-	130	SPs	PSO	[28]
NP-YC (Yeast)	644	0.6	-	139	SPs	PSO	
OLC (Luffa cylindrica)	-	1.67	4	59.88	LM	PSO	[3]

LM—Langmuir, PFO—pseudo-first-order, FL—Freundlich, PSO—pseudo-second-order, SPs—Sips, ELV—Elovich, TK—Temkin, FTP—fractional power.

3.1. Adsorbent Property

Specific Surface Area

The study only examined 34 out of 35 adsorbents for their SSA and MAC, as two publications did not indicate the SSA for the adsorbents used in their study. The average SSA for the adsorbents was 609.65 m^2/g, with 17 out of 32 having a low SSA of 0–500 m^2/g. Meanwhile, the same 17 adsorbents also had low MAC considering they were below 171.737 mg/g, which is the average MAC, as seen in Figure 2a.

Among the 34 adsorbents, TWPC-800 had the highest SSA of 2260.82 m^2/g and a high MAC of 491.37 mg/g. GSAC had the highest MAC of 916.679 mg/g and an SSA of 1099.86 m^2/g, which is above average but only half of the highest SSA, indicating that SSA solely does not indicate the adsorption process. Even though SSA is critical in determining MAC, other factors such as surface chemistry, pore structure, and specific interaction between the adsorbent and adsorbate molecule also affect the MAC.

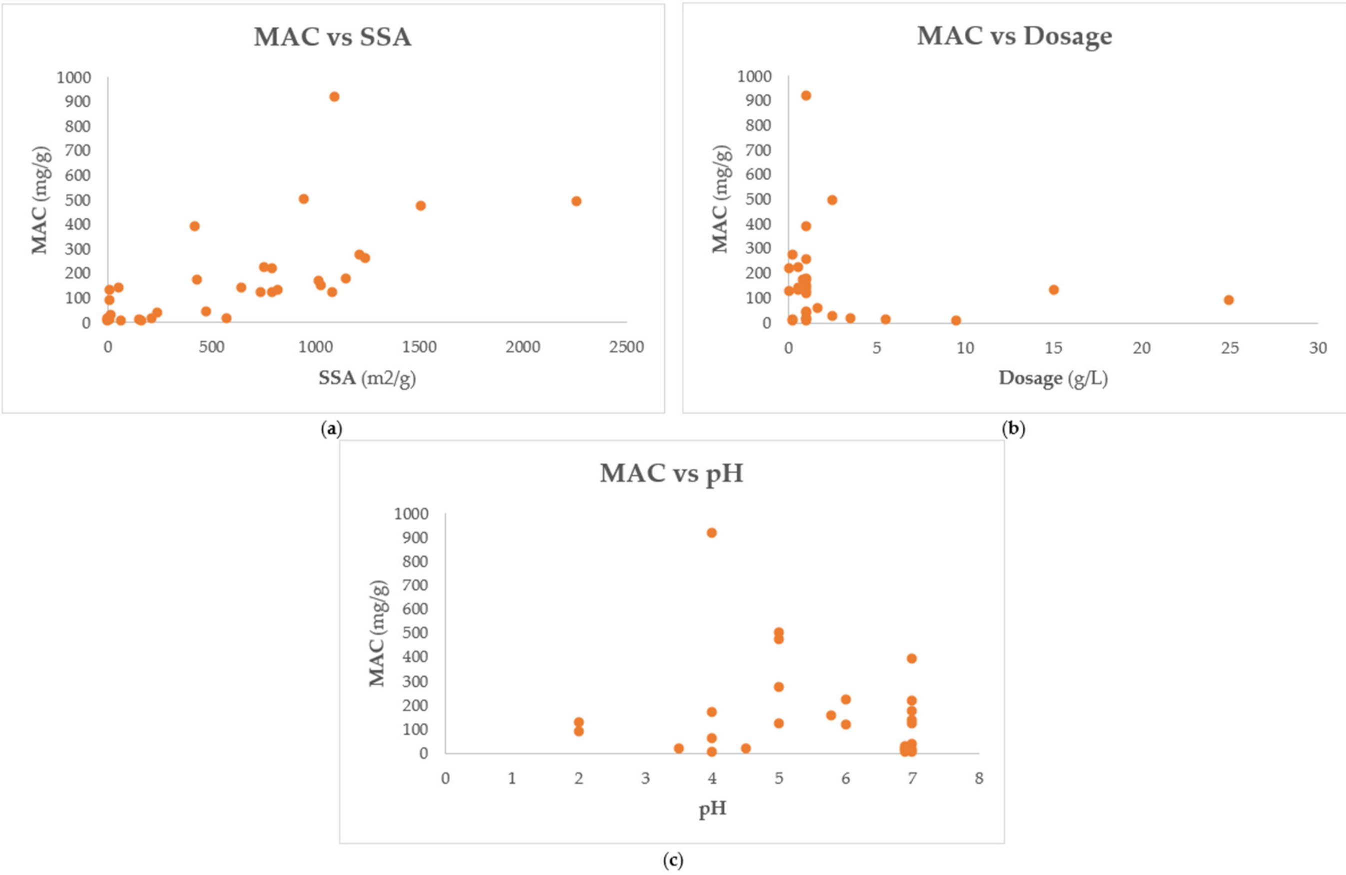


Figure 2. Adsorbents MAC against different characteristics: (**a**) MAC vs. SSA; (**b**) MAC vs. dosage; (**c**) MAC vs. pH.

3.2. Adsorption Parameters

3.2.1. Adsorbent Dosage

The study only investigated 33 of 35 adsorbents for their maximum adsorption capacity and corresponding dosage, as one publication did not indicate dosage in their study. The average dosage used among the 33 adsorbents was 2.56 g/L, with most adsorbents using a dosage below 5 g/L, particularly 1 g/L, as seen in Figure 2b. The low average dosage indicates that minimal amounts might be sufficient to acquire adequate adsorption. One exception was GS, which had a low MAC of 89.194 mg/g at a dosage of 25 g/L, which indicates that a high dosage does not necessarily mean a high MAC. Among the commonly used 1 g/L adsorbents was GSAC, which had the highest MAC, highlighting that certain adsorbents perform efficiently in capturing caffeine. The optimal dosage for achieving the desired adsorption capacity depends on the properties of the adsorbent material and the specific experimental conditions rather than simply increasing the dosage to provide more binding sites for adsorption.

3.2.1.1. pH Level

Only 29 adsorbents were evaluated for their MAC at a specific pH level, as two publications did not specify the pH level used in their study. The average pH level of the 29 adsorbents was 5.46, indicating an acidic nature. Although pH level 7 is the most used, as seen in Figure 2c, it was observed that the MAC varied from low to above average, indicating that pH level alone cannot determine the MAC. The study using GSAC showed that an acidic pH level of 4 resulted in the highest MAC, highlighting the significance of adsorbent performance in more acidic conditions. Neutral pH was avoided due to the possibility of desorption, as noted by Portinho et al. (2017) [9].

3.3. Adsorption Models

3.3.1. Adsorption Isotherms

Only 34 adsorbents were considered for the isotherm model study, as one publication did not study isotherms. Among the 34 adsorbents, 18 were fitted for Langmuir only, 8 were fitted for Sips only, 2 were for Langmuir and Sips, and 6 utilized other isotherms. GSAC, which has the highest MAC, was fitted on Sips, while the two adsorbents following it for the highest MAC, TWPC-800, and Pi/1:1/800 were fitted on Langmuir. The adsorbents close to average MAC, Pineapple, and MNC were fitted on Langmuir and Sips, respectively. The three with the lowest MAC, banana peel, banana peel composite, and WP900, were all fitted on Langmuir, while banana peel was also fitted on Sips.

This result demonstrates the flexibility of both Langmuir and Sips models to be fitted on different adsorbents, especially varying from high to low MAC. It suggests that adsorption happens on homogenous binding sites and monolayer adsorption while also applicable to heterogeneity and multilayer adsorption.

3.3.2. Adsorption Kinetics

In the study of the kinetics model, only 30 adsorbents were considered, as two publications did not examine kinetics. Of the 30 adsorbents, 22 were fitted on pseudo-second-order (PSO) kinetics only, 3 were fitted on pseudo-first-order (PFO) only, 1 was fitted to both PSO and PFO, and 4 were fitted to other kinetic models. The paper using GSAC did not include kinetic studies; however, the two adsorbents with the second highest MAC, TWPC-800, and Pi/1:1/800 were fitted on PSO. Pineapple and MNC were fitted on PSO, with MAC close to the average. Banana peel, banana peel composite, and WP900 with the lowest MACs were fitted on PFO, PSO, and PSO, respectively. These results indicate that most adsorbents at varying MACs were fitted on PSO, suggesting that PSO might be the more efficient way to describe the overall rate-limiting step in the adsorption process for different materials.

3.4. Cost Analysis

The economic viability of plant-based adsorbents for caffeine removal is significantly enhanced by their favorable cost dynamics. The abundant availability and often negligible or even negative cost associated with procuring these adsorbents, which are frequently derived from agricultural and industrial byproducts or waste, substantially reduces raw material expenditures [1,11]. This cost advantage is further amplified by the inherently simpler and less energy-intensive production processes for plant-based adsorbents compared to their commercial counterparts, leading to lower operational costs [25]. The potential for regeneration and multiple reuse cycles, as demonstrated in various studies, further contributes to the cost-effectiveness of these adsorbents by extending their operational lifespan and reducing the need for frequent replacements [28].

3.5. Life Cycle Assessment (LCA)

The utilization of plant-based adsorbents for caffeine removal aligns seamlessly with the principles of sustainable environmental management. The reliance on agricultural and industrial waste streams as the primary feedstock for these adsorbents effectively minimizes waste generation and promotes resource recovery, contributing to a circular economy model [28]. The reduced energy consumption and diminished chemical utilization during the production phase translate to lower greenhouse gas emissions and a lessened overall environmental burden [25]. The biodegradability of these adsorbents further enhances their environmental sustainability, ensuring minimal long-term ecological impact upon reaching their end-of-life [11,29].

3.6. Future Outlook

Among the analyzed studies, promising results for caffeine removal were found with GSAC, TWPC-800, and Pi/1:1/800/2, which were grape stalks, tea wastes, and pines, respectively. These adsorbents are cost-effective alternatives to commercial AC as they are readily available waste products that require minimal processing. They were derived from agricultural residues, promoting sustainability and reducing dependence on non-renewable resources.

The said adsorbents were performed in an acidic condition except for tea wastes, which was not mentioned in the study. Different processing techniques, as demonstrated with tea wastes, used carbonation and activation. Grape stalks, on the other hand, used an additional thermal treatment, which can be used to tailor plant-based absorbents' properties for different applications. Additionally, alternative processing techniques like gasification, as shown with pines, can expand the range of potential adsorbents for a more efficient method.

The different processes and methods mentioned increased SSA, which increased efficiency. Dosage was also on the lower side compared to the other adsorbents analyzed in the study. A combination of these characteristics, methods, and parameters indicates that it should be used in further studies to maximize its potential.

Overall, with the findings of this review, we are likely to see future researchers exploring other natural sources with similar properties to grape stalks, tea wastes, and pines. Additionally, further research could focus on optimizing parameters such as temperature, concentration, and the presence of other pollutants/contaminants to enhance the effectiveness of these adsorbents. As more established methods arise in the literature, it is only a matter of time before these alternatives reach the commercial market.

4. Conclusions

Different adsorbents derived from plant sources showed their potential and capacity for caffeine removal. Different adsorbent properties and adsorption parameters affect how the adsorption proceeds. It was found that the SSA contributes to a higher adsorption capacity, but surface chemistry still affects the adsorption. Additionally, a combination of a low dosage of an adsorbent and an acidic condition is seen to be sufficient to obtain optimal adsorption, but it is crucial to determine the optimal conditions due to variations depending on the material. Langmuir and Sips isotherms highlight the capabilities of adsorbents to have homogenous and heterogenous binding sites. Finally, the kinetics results show that PSO efficiently describes the adsorption process on various adsorbent materials derived from plant sources.

This study demonstrates the limitation of relying on a single parameter instead of optimizing different parameters, which is the ideal approach for adsorption studies. Among the studies analyzed, grape stalks, tea wastes, and pines indicate promising results for caffeine removal. However, at present, there are only a limited number of studies to determine the actual efficiencies of naturally derived adsorbents. Effective adsorbents presented here must be pursued for upscaled experiments, while the search for other potentially effective adsorbents continues. Nonetheless, it is expected that a trend in naturally derived adsorbents for caffeine will arise as methods become more established.

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