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Removal of atrazine from aqueous solution onto commercial Activated Carbons

Abdoulaye Demba N'diaye

Unité de Recherche Eau, Pollution et Environnement, Département de Chimie, Faculté des Sciences et Technique,
Université de Nouakchott Al Aasriya, BP 880, Nouakchott, Mauritanie

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abdouldemba@yahoo.fr
Phone: +22247624380;

Abstract

The main objective of this work was to study the adsorption of atrazine (ATZ) from aqueous solution onto commercial activated carbon forms: powdered (PAC) and granular (GAC) activated carbon, in batch system. The experimental adsorption data was modeled by kinetic and equilibrium models. The pseudo-first (PFO) and pseudo second-order (PSO) kinetic models were applied in order to describing the mechanism of adsorption. The data were found to be best fitted to the PSO model. The equilibrium study was performed using four models including Langmuir, Freundlich, Sips and Redlich-Peterson. The results revealed that the Freundlich isotherm fitted well the equilibrium data of ATZ onto PAC, while the Langmuir isotherm fitted well the equilibrium data of ATZ onto GAC. The suitability of the Freundlich for the adsorption of ATZ onto PAC and Langmuir isotherm for the adsorption of ATZ onto GAC was confirmed by Sips and Redlich-Peterson models. Based on isotherm parameter determination, PAC and GAC adsorbents capacities of 35.82 and 18.50 mg g^{-1} were obtained, respectively. The results of the present study showed the efficiency of using PAC and GAC as an adsorbent for the removal of ATZ from aqueous solution with PAC as the preference due to their high adsorption capacity.

1. Introduction

The use of herbicides to control weeds has been recognized as a part of agricultural practices throughout the world [1-5]. Unfortunately, the indiscriminate use of these herbicides to improve agricultural production and yield may have impacts on non-target organisms, especially aquatic life forms and their environment [6-10]. Like herbicide, Atrazine [2-chloro-4-ethylamino-6-isopropylamino-s-triazine] (ATZ) is most applied to control broad-leafed weeds and grasses. Due to its long-term persistence in soil, greater leaching potential and relatively high solubility in water, ATZ is frequently detected in aquatic media [11]. However, ATZ has also negative effects on the environment and on human health, due to their carcinogenic and mutagenic characteristic [12]. Atrazine's effectiveness and safety is well documented throughout the United States and around the world, with more than 22,000 scientific studies published on sciencedirect.

Thus, much attention and effort has been devoted to the elimination of ATZ, especially by chemical oxidation [13], photocatalysis [14], photocatalytic and biological degradation [15], Fenton degradation [16], membrane [17], electrochemical [18] and adsorption [19]. However, the adsorption process is one of most effective methods among all the above-mentioned ones for the removal of ATZ from aqueous media [20]. This is due to their versatility and the growing availability of various types of eco-friendly and low-cost adsorbents. In addition, the activated carbon is mainly used as adsorbent because it presents large specific surface area and numerous pores [21].

A several type of activated carbon materials has been used in the adsorption of pollutants from wastewater, such as, granular activated carbon (GAC), powdered activated carbon (PAC), carbon cloth, fibers, felts, black carbon from wheat residues (WC), carbon black and commercial activated carbon (AC). The forms GAC and PAC are the most used since they are considered very capable and effective materials for the adsorption of a variety of pesticides. GAC has been used in many processes for the treatment of drinking water supplies and industrial wastewaters [22].

The main objective of this work focused on the study, the removal of ATZ from aqueous solutions using the batch operation system, for the elimination of ATZ onto commercial PAC and GAC. For this purpose, kinetic and isotherm studies have been carried out with the non-linear methods.

2. Methodology

2.1 *Adsorbate and adsorbents*

All chemicals used in this study were of analytical reagent grade. A stock solution containing 1000 mgL^{-1} of ATZ was prepared by dissolving appropriate amount in methanol. Solutions of different standard concentrations of the ATZ were prepared by serial dilution of the stock solutions in ultrapure water.

The PAC was provided by FLUKA and GAC was supplied by PROLABO (physical and chemical characteristics described in **Table 1**). These conventional adsorbents were used without any treatment.

Table 1. Physical and chemical characteristics of the conventional adsorbents

Characteristics	PAC	GAC
Total basicity (meqg^{-1})	0.080	0.204
Total acidity (meqg^{-1})	0.704	0.580
SiO_2 (%)	7.2	15.1
Specific surface (m^2 g^{-1})	1002	1045
Particle size (µm)	10–50	2000-3000

2.2 Batch experiments

In order to investigate the effect of contact time on ATZ removal by adsorption onto activated carbons, 100 mg of GAC or 15 mg of PAC was added to 25 mL of ATZ at 5 mg L^{-1}, then the prepared solution stirring at 70 rpm at room temperature. Adsorption isotherms were obtained by varying the initial ATZ concentration from 2.5 to 100 mg L^{-1}. At the end of each experiment, the stirred solution mixture was micofiltered and the residual concentration of ATZ was determined by HPLC. The adsorption capacity (q_e) calculated using following **equation (1)**:

$$q_e = \frac{(C_i - C_e)V}{m} \qquad \textbf{(1)}$$

Where q_e is the amount of ATZ adsorbed by adsorbent (mg g^{-1}), C_i is the initial liquid-phase concentrations of ATZ (mg L^{-1}), C_e is the liquid-phase concentration of ATZ (mg L^{-1}), V is the solution volume (L) and m is the mass of adsorbent used (g).

3. Results and Discussion

3.1 Kinetic study

The effect of contact time on ATZ adsorption was examined in order to obtain experimental results in regard to kinetics of the process. The adsorbed ATZ concentration at equilibrium q_e was plotted against time for the PAC and GAC (**Figures 1-2**). We can also notice that the equilibrium is reached in 50 min for PAC and in about 80 min for GAC.

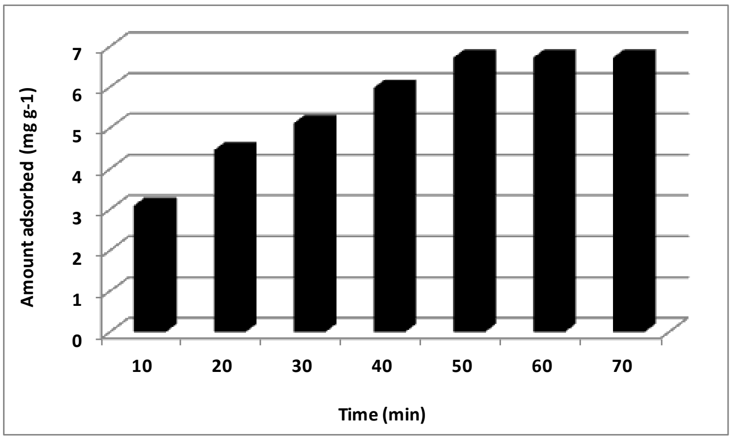

Figure 1. Variation of ATZ uptake onto commercial PAC against time

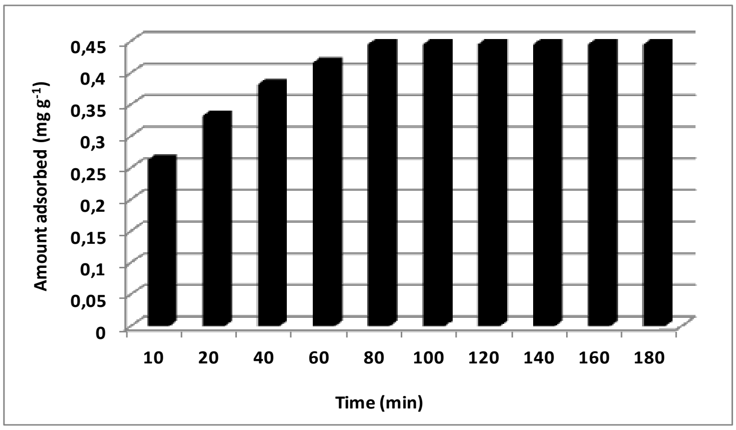

Figure 2. Variation of ATZ uptake onto commercial GAC against time

The rate and mechanism of the adsorption process was evaluated using two different kinetic models, namely pseudo-first-order model (PFO) and pseudo-second- order (PSO) model [23]. The non-linear kinetics PFO and PSO models expressed by equation (2) and (3), respectively:

$$q_t = q_e(1-\exp^{-k_1 t}) \qquad \textbf{(2)}$$

$$q_t = \frac{k_2 q_e^2 t}{1+k_2 q_e t} \qquad \textbf{(3)}$$

Where q_t is the amount of ATZ adsorbed per unit mass of adsorbent (mg g^{-1}) at time t, k_1 is the PFO rate constant (L min^{-1}), k_2 (gmg^{-1}min^{-1}) is the PSO rate constant for adsorption, q_e (mg g^{-1}), t is the contact time (min).

The adsorption data kinetics was analyzed using PFO and PSO models. **Figures 3-4** show the experimental equilibrium data and the predicted theorical kinetics for the adsorption of ATZ onto PAC and GAC. The values of two model parameters k_1, k_2 and R^2 are presented in **Table 2**. The kinetic parameters constants of experimental data were calculated using nonlinear regression with the help of Solver Excel. Figures 3-4 and Table 2 shows that a high value of regression coefficient (R^2) of PSO model was achieved for ATZ, which indicated that this adsorption model is more fitted with the experimental adsorption data than the PFO model suggesting that chemisorption is the rate controlling step. Similar phenomena have been reported for pesticides adsorption on various adsorbents [24; 25].

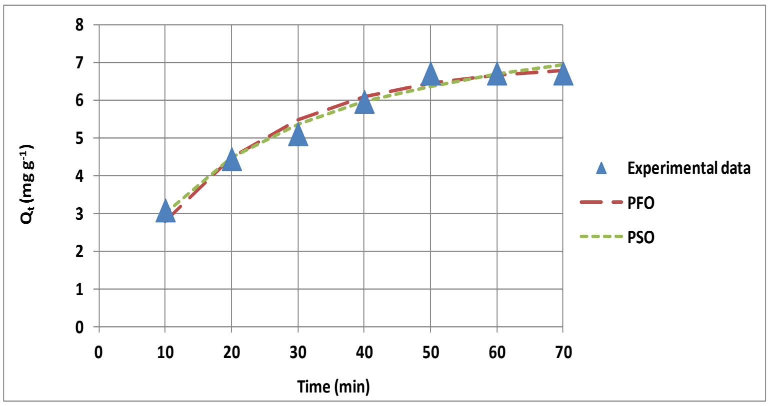

Figure 3. PFO and PSO non linear for PAC

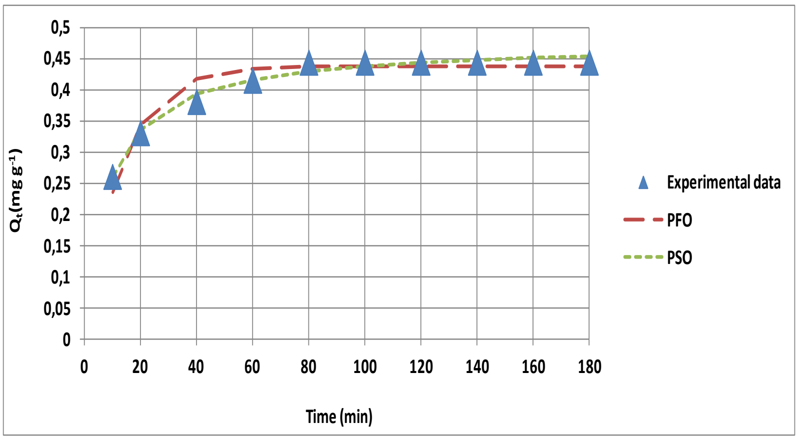

Figure 4. PFO and PSO non linear for GAC

Table 2. PFO and PSO models constants for the adsorption of ATZ by PAC and GAC

Model	Parameters	PAC	GAC
PFO	q_{exp}	6.73	0.445
	q_e	6.99	0.438
	K_1	0.051	0.077
	R^2 (%)	97.2	92.30
PSO	q_e	8.85	0.473
	K_2	0.006	0.254
	R^2 (%)	97.9	98.53

On the other hand, the PSO rate constant (k_2) for PAC were lesser as compared to GAC (**Table 1**). This result may be related to the higher silica content in the GAC, which was responsible for faster kinetic rate (**Table 2**). Similar observations have been reported by Trivedi et al., [26], indicated that the silica fraction determines the kinetics study in the adsorption process.

3.2 Adsorption isotherms

The equilibrium adsorption isotherms were studied using the Langmuir (equation 4) [27], Freundlich (equation 5) [28], Sips (equation 6) [29] and Redlich-Peterson models (equation 7) [30]:

$$q_e = \frac{q_m K_L C_e}{1 + K_L\, C_e} \qquad \textbf{(4)}$$

$$q_e = K_F C_e^{1/n} \qquad \textbf{(5)}$$

$$q_e = q_m \frac{K_S C_e^n}{(1 + K_S C_e^n)} \qquad \textbf{(6)}$$

$$q_e = \frac{K_{RP} C_e}{1 + \alpha_{RP} C_e^n} \qquad \textbf{(7)}$$

Where q_e is the quantity of ATZ adsorbed per unit mass of adsorbent (mg g^{-1}), k_L is the Langmuir constant related to the adsorption capacity (L g^{-1}), C_e is the concentration of ATZ in the solution at equilibrium (mg L^{-1}), q_m is the maximum uptake per unit mass of adsorbent (mg g^{-1}), K_F (mg g^{-1}) (L mg^{-1})^{n}, n is the Freundlich heterogeneity factor, q_m the Sips maximum adsorption capacity (mg.g^{-1}), K_S the Sips equilibrium constant (L mg^{-1}) and n the Sips model exponent describing heterogeneity. K_{RP} (L g^{-1}) and α_{RP} (L mol^{-1}) are the Redlich-Peterson isotherm constants, while n is the exponent, which lies between 0 and 1. The factor of separation of Langmuir, R_L, which is an essential factor characteristic of this isotherm was calculated by using the relation:

$$R_L = \frac{1}{(1 + k_L C_0)} \qquad \textbf{(8)}$$

The R_L value indicates the shape of the isotherm as follows: When $R_L = 1$ the adsorption is linear; when $0 < R_L < 1$, it is favourable, then if $R_L = 0$, it is irreversible, while when $R_L > 1$, the adsorption process is unfavorable.

The isotherm constants of experimental data were calculated using nonlinear regression with the help of Solver Excel. The isotherms constants related to studied isotherm models determined from the plots of Langmuir and Freundlich shown in **Figures 5-6** are listed in **Table 3**.

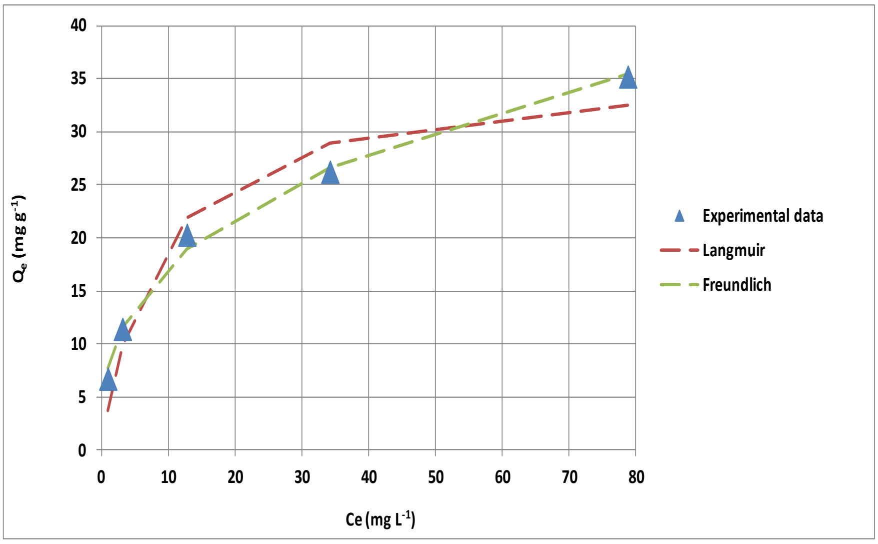

Figure 5. Langmuir and Freundlich models for ATZ adsorption by PAC

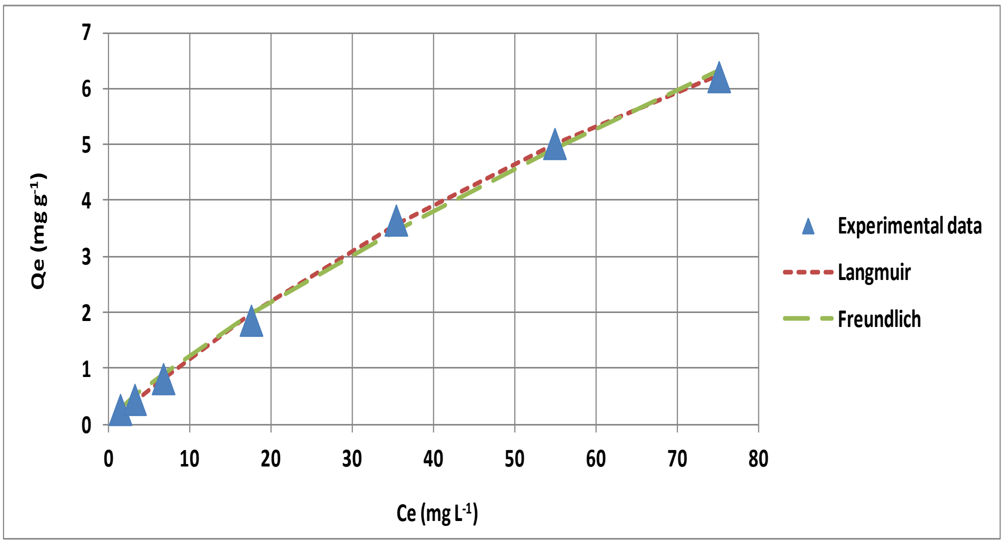

Figure 6. Langmuir and Freundlich models for ATZ adsorption by GAC

Table 3. Parameters of Langmuir and Freundlich isotherm models for ATZ retention on the PAC and GAC

Model	Parameters	PAC	GAC
Langmuir	**q_m**	35.82	18.50
	K_L	0.123	0.007
	R_L	0.075	0.58
	R^2 (%)	94.5	99.92
Freundlich	**n**	2.9	1.20
	K_F	7.86	0.20
	R^2 (%)	99.41	99.75

Comparing the correlation coefficients of the Langmuir and Freundlich indicated that the equilibrium experimental data for PAC are best fitted by the Freundlich model, while ATZ adsorption onto GAC is best follows to the Langmuir model. Sips and Redlich-Peterson have been applied to evaluate the fit by isotherm for the adsorption of ATZ onto PAC and GAC by Figures 7-8. According to Table 4, the coefficients of correlation are very good ($R^2 \geq 99$ %) for both models.

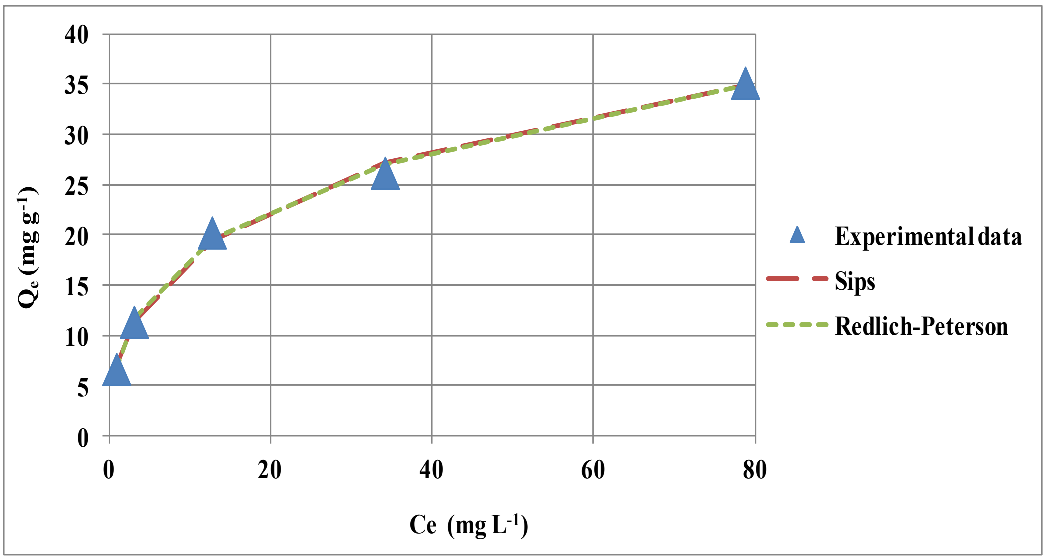

Figure 7. Sips and Redlich-Peterson models for ATZ adsorption by PAC

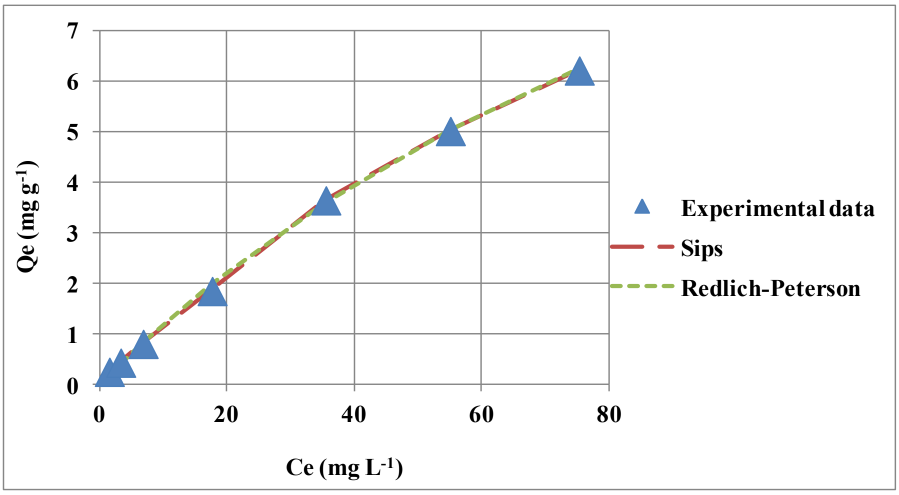

Figure 8. Sips and Redlich-Peterson models for ATZ adsorption by GAC

The suitability of the Freundlich model for the adsorption of ATZ onto PAC and Langmuir isotherm model for the adsorption of ATZ onto GAC was confirmed by the exponent values of the Sips and Redlich –Peterson models n (the exponent n, lies between 0 and 1 for PAC and. the exponent n is equal 1for GAC). The Langmuir adsorption monolayer was also reported for ATZ adsorption onto activated carbon prepared from rubber tire waste [31] and commercial activated carbon fibers [32] which were similar to results found it in this work for the adsorption of ATZ onto GAC. However, same studies report that the adsorption isotherms of ATZ for various adsorbents were best fitted by the Freundlich model [33-35] which were in coherent on the adsorption of ATZ onto PAC.

Table 4. Parameters of Sips and Redlich-Peterson
Sips isotherm models for ATZ retention on the PAC and GAC

Model	Parameters	PAC	GAC
Sips	**Q_m**	93.26	18.35
	K_s	0.084	0.007
	n	0.45	1
	R^2 (%)	99.67	99.92
Redlich-Peterson	**α_{RP}**	2.19	0.009
	K_{RP}	22.16	0.127
	n	0.71	0.94
	R^2 (%)	99.77	99.91

The values of R_L and K_L are in between 0 and 1 indicates the favorability of the adsorption of ATZ onto both adsorbents. Which approved by the value of the magnitude of the exponent n. It is generally stated that values of n in the range 2–10 represent good, 1–2 moderately difficult, and less than 1 poor adsorption characteristics. The studied PAC is a good adsorbent for ATZ (n=2.9) than GAC (n = 1.2) (**Table 3**). A higher value of K_L for PAC indicates a higher affinity of the adsorbent towards ATZ than GAC. The adsorption capacities of PAC and GAC were 35.82 and 18.50 mg g^{-1} respectively. From these values, the adsorption efficiency of PAC for ATZ is 2 times higher than those of GAC, suggesting the higher effectiveness of PAC for the adsorption removal of ATZ. According to **equation 8**, the values of R_L obtained from both adsorbents are range between 0 and 1 (**Table 3**). This range, ($0 < R_L < 1$) shows that the process of adsorption is favorable for the two adsorbents. Therefore, it can be inferred that adsorption is the most effective on PAC and is the highest as the separation factor R_L follows the order of PAC< GAC. This result also supports the maximum adsorption capacity q_m and K_F of the adsorbents (PAC > GAC).

Thus, if we consider **Figures 5–6**, we can notice that the better ATZ retention was achieved onto the PAC even if this latter offers the same specific area than the GAC one (**Table 1**). Functional groups on the surface may account for the difference in ATZ retention between the two types of carbons. In this study, the titrations test showed that the numbers of acidic groups onto the GAC and PAC surfaces are respectively of 0.580 and 0.704 meq g^{-1} (**Table 1**). This result showed that the surface functional groups cannot explain the difference in ATZ retention onto the activated carbons. This difference may be attributed to the pore size of the porous material since the GAC and the PAC have the same specific area and the number of functional groups. Similar observations are reported by Llad'o et al, [36]. On the other hand, the PAC with higher carbon content (99.95 %) than the GAC offers more binding sites, confirming the higher effectiveness of PAC for the adsorption removal of ATZ. This hypothesis has been described by Trivedi et al., [26] reports that carbon fraction in the adsorbent determines the capacity of adsorption.

Conclusion

The removal of ATZ from aqueous solution using PAC and GAC has been investigated. Atrazine adsorption was expressed well by the PSO kinetics model. The equilibrium study was performed using three models including Langmuir, Freundlich, Sips and Redlich-Peterson. The adsorption of ATZ onto PAC was best described by the Freundlich model, while the adsorption of ATZ onto GAC was best described by the Langmuir. The suitability of the Freundlich for the adsorption of ATZ onto PAC and Langmuir isotherm for the adsorption of ATZ onto GAC was confirmed by the exponent values of the Sips and Redlich-Peterson models n. Based on isotherm parameter determination, PAC and GAC adsorbents capacity of 35.82 and 18.50 mg g^{-1} was obtained, respectively. The results of the present study showed the efficiency of using PAC as an adsorbent for the removal of ATZ from aqueous solution.

Disclosure statement: *Conflict of Interest:* The authors declare that there are no conflicts of interest. *Compliance with Ethical Standards:* This article does not contain any studies involving human or animal subjects.

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