

The use of activated carbon for the removal of pharmaceuticals from aqueous solutions: a review

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Published online: 2 December 2017
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Abstract The presence of pharmaceutically active compounds in surface and ground water is of concern due to the adverse effects they may have on human health, aquatic life, and the environment, emphasizing the importance of their removal from the water compartment. Activated carbon adsorption has proven to be effective for the removal of several types of inorganic and organic contaminants either as a stand-alone polishing step or in combination with other conventional and advanced water and wastewater treatment systems. This paper discusses the current status of the removal of pharmaceuticals from water using activated carbon derived from numerous precursors, providing an in-depth review of the multitude of factors (adsorbent properties, adsorbate properties,

operating conditions) affecting the adsorption process, from the preparation of the activated carbon to its regeneration. A critical assessment of the existing literature is presented, highlighting research and development needs that may ultimately lead to a more comprehensive and sustainable use of activated carbon for the removal of pharmaceuticals from the water environment.

Keywords Activated carbon · Adsorption · Pharmaceuticals · Regeneration

List of symbols

α	Initial adsorption rate constant, mg/g/s
α_{BS}	Measure of the width of sorption energy distribution in the Brouers Sotolongo model, dimensionless
β	Initial desorption rate constant, g/mg
[]	Concentration of pharmaceutical, mol/L
a	Number of neighboring sites occupied by the adsorbate, dimensionless
a_R	Redlich–Peterson isotherm constant, (L/g) ^{mRP}
b	Temkin adsorption constant, J/mol
C	Constant related to thickness of boundary layer, mg/g
C_o	Initial concentration of adsorbate, ng, μ g or mg/L
C_e	Equilibrium adsorbate concentration, ng, μ g or mg/L
D	Intraparticle diffusion coefficient, cm ² /s

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s11157-017-9456-8>) contains supplementary material, which is available to authorized users.

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E	Characteristic adsorption energy, kJ/mol	n_{AV}	Fractionary order exponent, dimensionless
G	Gibbs free energy, kJ/mol	pK_a	Acid dissociation constant, dimensionless
H	Enthalpy, kJ/mol	pH_{zc}	pH at zero charge, dimensionless
k_1	Pseudo-first order rate constant, s^{-1}	q	Amount of solute adsorbed per gram of adsorbent, mg/g
k_2	Pseudo-second order rate constant, g/mg/s	q_{BS}	Saturation adsorption value of Brouers Sotolongo model, mg/g
k_{2D}	Diffusion reaction constant, L/mg/min	q_e	Equilibrium adsorption capacity, mg/g
k_{AV}	Fractionary order kinetic constant, h^{-1}	q_m	Maximum adsorption capacity on the first monolayer, mg/g
k_{global}	Global kinetic constant (includes both kinetic constant of reaction in bulk liquid in absence of AC and the reaction occurring on AC surface), min^{-1}	q_t	Amount of adsorbate adsorbed at time t, mg/g
k_N	General order constant rate, $min^{-1} (g/mg)^{n-1}$	R	Universal gas constant, J/mol/K
K_1	Equilibrium constant for first monolayer, L/mg	S	Entropy, J/mol/K
K_2	Equilibrium constant for second monolayer, L/mg	S_a	Adsorbate solubility, mg/L
K_{BET}	Brunauer–Emmet–Teller adsorption constant, dimensionless	t	Time, s, min, h
K_{BS}	Brouers Sotolongo model constant, L/mg	T	Temperature, K
K_e	Elovich equilibrium constant, L/mg		
K_F	Freundlich equilibrium constant, $mg/g \text{ mg}^{-1/n_F} \text{ L}^{1/n_F}$		
K_g	Liu equilibrium constant, L/mg		
K_{id}	Intra-particle diffusion rate constant, $mg/g \text{ h}^{-0.5}$		
K_L	Langmuir equilibrium constant, L/mg		
K_{LF}	Langmuir–Freundlich equilibrium constant for heterogeneous solids, L/mg		
K_N	Nitta equilibrium constant, L/mg		
K_{OW}	Water dissociation constant, dimensionless		
K_R	Redlich–Peterson isotherm constant, L/g		
K_{RPI}	Radke–Prausnitz equilibrium constant, L/mg		
K_s	Sips equilibrium constant, $(L/mg)^{m_S}$		
K_T	Toth equilibrium constant, L/mg		
K_{Tem}	Temkin equilibrium constant, L/mg		
m_F	Freundlich model exponent, dimensionless		
m_L	Liu model exponent, dimensionless		
m_{LF}	Heterogeneity parameter, dimensionless		
m_N	Nitta model exponent, dimensionless		
m_{PDM}	Polany–Dubinin–Manes model exponent, dimensionless		
m_{RP}	Redlich Peterson model exponent, dimensionless		
m_{RPI}	Radke–Prausnitz model exponent, dimensionless		
m_S	Sips model exponent, dimensionless		
m_T	Toth model exponent, dimensionless		
n_o	Order of kinetic adsorption		

1 Introduction

As the world's population continues to grow, so does the demand for clean water (WWAP 2016). Meanwhile, the existing water supply sources are becoming more polluted as the use of chemicals, pharmaceuticals, and industrial compounds, accompanies economic development (Carpenter et al. 2011; Zimmerman et al. 2008). This poses a threat to the livelihood of millions around the globe, as conventional methods for water supply and irrigation have become a source of environmental concern (WWAP 2016). In addition, natural water sources in several countries throughout the world are being depleted due to various factors including global warming rendering it of even greater necessity to devise reliable methods for the reutilization of wastewater. Wastewater treatment provides a water supply source independent of weather conditions, such as rainfall and drought, and partially relieves the stress placed on natural resources (Garcia and Pargament 2015). However, wastewater contains several different types of contaminants of emerging concern, prominent among which are pharmaceuticals (Barbosa et al. 2016).

Pharmaceuticals and their degradation products have been detected, at low concentration levels (ng/L up to $\mu\text{g/L}$) in surface and groundwater throughout the world (Cabrita et al. 2010; Hughes et al. 2013; Segura et al. 2015). Pharmaceuticals are released to the aquatic environment via several pathways; these

include discharges from pharmaceutical manufacturing facilities, discharge of treated sewage effluent from sewage treatment plants, the application of sewage sludge or animal manure to agricultural land, leakage from sewage treatment plants, emissions from medical units and disposal of unwanted pharmaceuticals (Kookana et al. 2014).

The presence of pharmaceutically active compounds in water systems is of concern due to their potential carcinogenicity, mutagenicity, aquatic toxicity and other ecological effects such as the evolution of resistant bacteria (WHO 2014; Yu et al. 2016). Although some studies have investigated the possible effects on human health and aquatic organisms, these studies have not yet fully identified the risks associated with the persistent exposure to random combinations of these compounds and their toxicological significance remains, to date, an unanswered question (Backhaus 2014). Consequently, the removal of pharmaceutical residuals from drinking water and wastewater has become crucial (Taheran et al. 2016). Given the large number of pharmaceuticals in use, their varying physico-chemical properties (even within one class) and their low concentrations in the aqueous environment, several treatment methods may be needed for the elimination of these contaminants (Delgado et al. 2012). However as conventional wastewater and water treatment unit operations do not appear capable of eliminating all classes of pharmaceuticals (Taheran et al. 2016), the use of several advanced process treatment technologies may be necessary (Wang and Wang 2016).

Some of the advanced and tertiary treatment methods that have been reported to be effective for the removal of pharmaceuticals from treated wastewater and water streams include advanced oxidation techniques, membrane filtration and adsorption (Rodríguez-Narvaez et al. 2017). One of these methods, activated carbon (AC) adsorption, has been reported to proffer the most potential as a sustainable and highly effective treatment process (Sheng et al. 2016).

Adsorption refers to the buildup of a substance (adsorbate) in a fluid phase (liquid or gas) onto the surface of an adsorbent, either through physical or chemical binding (Ahmed et al. 2015). The process is well suited for the removal of low concentrations of synthetic (and natural) organic contaminants from water and wastewater streams (Cabrita et al. 2010) and is often used as a polishing step for the elimination of a

wide range of low to medium molecular mass compounds without generating by-products (Delgado et al. 2014).

Overall, the adsorption process can be described by the hydrophobic electrostatic interactions between the dissolved adsorbate and the adsorbent, which causes the pollutant to adhere to the adsorbent's surface (Nam et al. 2014). The adsorption process has several advantages, including good removal efficiency at low concentrations of organic/inorganic contaminants, possibility of regeneration and reuse, applicability to both continuous and batch processes and reliable and simple operational procedures. The process's insensitivity to toxic pollutants and the absence of toxic byproducts as well as low operational energy requirements (Ek et al. 2014) are additional advantages (Zhou et al. 2015). Nonetheless, the adsorption process also has several disadvantages such as high production and regeneration costs (Ahmed et al. 2015) and the frequency of AC regeneration/replacement, which often jeopardizes efficiency (Zanella et al. 2014). Furthermore, it has been reported that in industrial applications adsorption processes often suffer from reduced efficacy and increased adsorbent consumption arising from the presence of background organic material (BOM), whether naturally occurring (NOM) or arising from effluent organic matter (EfOM) (Margot et al. 2013).

There are several types of AC materials that have been used, primarily on a laboratory scale, for the removal of pharmaceuticals and other contaminants from aqueous solutions. These ACs have been produced from various types of carbonaceous materials precursors (Yu et al. 2016), carbon nanotubes (CNTs) of various types (Jung et al. 2015; Shan et al. 2016), clay materials (Kyzas et al. 2015), graphene oxides (Cai and Larese-Casanova 2014), and molecularly imprinted polymers (Kyzas and Deliyanni 2015). One of the most commonly used AC materials, particularly in commercial applications, is coal (Iovino et al. 2015; Lladó et al. 2015; Ruiz et al. 2010). AC is also produced from other carbonaceous materials including raw and processed agricultural waste precursors (Alahabadi et al. 2017; Cabrita et al. 2010; Mestre et al. 2009), wood (Nielsen et al. 2014), tyre pyrolysis char (Acosta et al. 2016) and paper mill sludge (Calisto et al. 2017).

This work focuses on the current status of activated carbon adsorption in pharmaceutical removal from

water/wastewater streams and its future viability. A critical assessment is conducted on the existing literature, investigating the impact of activated carbon type, preparation, properties, and various operating conditions (adsorbent dosage, pharmaceutical initial concentration, temperature, pH, ionic strength, presence of background organic matter) on the adsorption process. Other aspects, such as the mechanisms postulated for the adsorption process, competitive adsorption, arising from the presence of other pharmaceuticals (in multiple solute systems), as well as methods proposed for desorption/regeneration of the exhausted carbon are also addressed. The adsorption isotherms, kinetics, thermodynamics, and mechanism are summarized. The adsorptive capacities of the various types of activated carbon and the maximum and minimum amounts removed (presented in terms of weight of pharmaceutical adsorbed per weight of activated carbon used) for 66 pharmaceuticals are tabulated.

It is worth noting that there are a number of reviews that are available in the literature that have addressed some of the aspects considered in this review. For example Yu et al. (2016), Ahmed et al. (2015), Fu et al. (2017) and Ahmed (2017) focused on the removal of antibiotics only via AC adsorption. Kyzas and Kostoglou (2014) assessed the status of “green adsorbents” for the removal of various pollutant types: dyes, heavy metals, phenols, pesticides, and pharmaceuticals. This review paper stands out in that it comprehensively addresses removal of pharmaceuticals belonging to all therapeutic classes and AC derived from numerous precursors. Furthermore, it summarizes the available data on the adsorption of pharmaceuticals onto AC, examines the various parameters that affect the adsorption process and identifies gaps and limitations that have hindered the widespread application of this process.

2 Removal of pharmaceuticals by AC

The removal of pharmaceuticals by AC is characterized by the interplay of several elements spanning the preparation process, operating conditions, and other effects. The preparation process refers to the category of adsorbent, the type of precursor used, and the activation method employed, all of which contribute/limit the effectiveness of the process. This effectiveness is also related to the properties of the adsorbent

and the properties of the adsorbate (pharmaceutical) being studied. Furthermore, operating conditions (adsorbent dosage, temperature, pH, ionic strength, adsorbate properties and organic material) play a role in impacting the adsorption capacity of the AC.

At this stage it is important to state that several isotherm models (most of which have been proposed previously for adsorbates other than pharmaceuticals) have been used to characterize the adsorption process. Isotherms quantify the interactions at equilibrium when the two phases have been in contact for a sufficient period of time such that the concentration of the adsorbate in the bulk solution balances with that at the interface (Chayid and Ahmed 2015). A detailed description of the various isotherm models is beyond the scope of this work [the interested reader is referred to Foo and Hameed (2010)]. However, a table summarizing all of the isotherms used to describe adsorption of pharmaceuticals onto activated carbon is presented in Table 1. It is worth noting that the vast majority of researchers have found that the Langmuir (1916) and Freundlich (1906) isotherm models represented the best fit for the adsorption data.

Kinetic modeling is often used to determine the rate-controlling step of the adsorption process; a number of kinetic models have been proposed for the adsorption of pharmaceuticals onto AC surfaces. A detailed description of these kinetic models is beyond the scope of this work [the interested reader is referred to Plazinski et al. (2009) and (Tan and Hameed 2017)]. However, Table 2 summarizes kinetic models that are commonly used for the adsorption of pharmaceutical onto activated carbon.

Table 3 summarizes the studies that have considered the adsorption of a single pharmaceutical onto AC surfaces, where information relating to the type of AC precursor, the Brunauer Emmett and Teller (BET) surface area of the AC and the experimental conditions (pH, temperature and initial pharmaceutical concentration) used by the researchers are provided. The table also includes the removal percentages achieved, the adsorption capacity as well as the kinetic and isotherm models used to fit the experimental data. Table 4, on the other hand, lists an almost identical set of information but for studies in which the adsorption of more than one pharmaceutical was considered by the researchers; the studies listed in Table 4, however, have investigated the adsorption of each pharmaceutical individually and have not addressed any competitive

Table 1 Commonly used isotherm models

Isotherm model	Equation	Notes
Brunauer–Emmet–Teller	$\frac{C_e}{q_e(C_o - C_e)} = \frac{1}{q_m K_{BET}} + \frac{K_{BET} - 1}{q_m K_{BET}} \left[\frac{C_e}{C_o} \right]$	Multilayer adsorption; surface is modeled as a set of nearby sites
Elovich	$q_e = q_m K_e C_e \exp\left(-\frac{q_e}{q_m}\right)$	Supposes multi-layer adsorption
Freundlich	$q_e = K_F \cdot C_e^{m_F}$	Multi-layer adsorption Heterogeneous adsorption Single molecule per site
Guggenheim–Anderson–Boer	$q_e = \frac{q_m \cdot K_1 \cdot C_e}{(1 - K_2 \cdot C_e) \cdot [1 + (K_1 - K_2) \cdot C_e]}$	
Langmuir	$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$	Monolayer adsorption Homogenous energy distribution
Langmuir–Freundlich	$q_e = \frac{q_m (K_{LF} C_e)^{m_{LF}}}{1 + (K_{LF} C_e)^{m_{LF}}}$	Combination of Langmuir and Freundlich; one molecule may occupy multiple sites At low concentrations, reduces to Freundlich isotherm; at high concentrations, predicts monolayer sorption capacity (Langmuir)
Liu	$q_e = \frac{q_m \times (K_g C_e)^{m_L}}{1 + (K_g C_e)^{m_L}}$	
Nitta	$\frac{q}{q_m} = K_N C_e \left(1 - \frac{q}{q_m}\right)^{m_N}$	Based on mass action law
Polanyi–Dubinin–Manes	$q_e = q_m \exp\left(-\frac{RT \ln\left(\frac{C_e}{S}\right)}{E}\right)^{m_{PDM}}$	
Radke–Prausnitz	$q_e = \frac{q_m K_{RPI} C_e}{(1 + K_{RPI} C_e)^{m_{RPI}}}$	
Redlich–Peterson	$q_e = \frac{K_{RP} C_e}{1 + a_R C_e^{m_{RP}}}$	Can be applied in either homogeneous or heterogeneous systems
Sips	$q_e = \frac{q_m K_S C_e^{m_S}}{1 + K_S C_e^{m_S}}$	
Temkin	$q_e = \frac{RT}{b} \ln(K_T C_e)$	Uniform distribution of binding energy Heat of adsorption of molecules in the layer decrease linearly with coverage
Toth	$q_e = \frac{q_m K_T C_e}{(1 + (K_T C_e)^{m_T})^{1/m_T}}$	Describes heterogeneous adsorption isotherm systems

Table 2 Commonly used kinetic models

Kinetic model	Equation
Diffusion reaction	$\frac{\partial q_t}{\partial t} = k_{2D}(q_e - q_t)^2$
Elovich	$q = \frac{\ln(\alpha\beta) + \ln(t)}{\beta}$
Fractionary order	$q_t = q_e \times \{1 - \exp[-(k_{AV} \times t)]^{n_{AV}}\}$
General order	$q_t = q_e - \frac{q_e}{[k_N(q_e)^{n-1} t^{(n-1)+1}]^{\frac{1}{1-n}}}$
Global first order	$-\frac{d[PC]}{dt} = k_{global}[PC]$
Intraparticle diffusion	$q_t = K_{id} + \sqrt{t} + C$
Pseudo-first order	$q = q_e(1 - e^{-k_1 t})$
Pseudo-second order	$q = \frac{k_2 q_e^2 t}{1 + k_2 q_e t}$

effects (competitive effects are addressed in section 2.4 for multiple pharmaceutical systems). For all the studies presented in Tables 3 and 4, the appropriate isotherm and kinetic model are listed where available. The mechanisms that have been proposed to explain the adsorption of pharmaceuticals onto AC along with the thermodynamics of adsorption will be addressed in the mechanism of adsorption section, Sect. 5.1.

3 The effect of materials on adsorption

AC can be synthesized from a number of precursors and prepared using different procedures thus resulting

Table 3 Summary of activated carbon adsorption studies for individual pharmaceuticals

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceutical	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Acosta et al. (2016)	Water	AC from tyre pyrolysis char and commercial	118–814	Tetracycline	5–80		422–455	6.5–7.5	288–308	Sips	Pseudo-second
Ahmed and Theydan (2012)	Distilled	AC from Albiza lebeck seed pods	1676–1824	Cephalexin	20–100		111.6–137.0	7.0	303–323	Langmuir	Pseudo-second
Aksu and Tunç (2005)	Distilled	AC (commercial powdered)	1000	Penicillin G	50–1000	12.0–78.3	330–459	5.0–8.0	298–318	Langmuir	Pseudo-second
Alahabadi et al. (2017)	Various	AC from dried pomegranate wood	1024–1029	Chlortetracycline	20–200		377.5–482.5	2.0–10.0	283–323	Redlich–Peterson	Pseudo-second
Baghdadi et al. (2016)	Wastewater	Powdered AC (commercial)	1241–1378	Carbamazepine	2	93	182.9	6.65	273–303	Radke–Prausnitz	Elovich
Belhachemi and Djelaila (2017)	Various	AC from date pits	1069–1325	Amoxicillin	20–700		244.0–424.3	2.0–11.0	295	Langmuir, Sips	Pseudo-second
Bernardo et al. (2016)	Deionized	AC from potato peels	866	Diclofenac	10–100		69–146	5.0–12.0	298	Langmuir	Pseudo-second
Bhadra et al. (2016)	Distilled	AC (commercial, chemically modified)	704–1016	Diclofenac sodium	25–100		83–487	4.2–10	298	Langmuir	Pseudo-second
Bojic et al. (2015)	Distilled	AC from Lagenaria vulgaris shell	665	Ranitidine	10–400	93–99	315.5	2.0–11.0	298	Langmuir	Pseudo-second
Carrales-Alvarado et al. (2014)	Distilled	AC (commercial)	919	Metronidazole	50–1000	–	182.2–389.8	2.0–12.0	298	Radke–Prausnitz	–
Chayid and Ahmed (2015)	Distilled	AC from <i>Arundo donax</i> (giant reed)	1065	Amoxicillin	50–450		75–345	7.0	303–323	Sips	Pseudo-second
Chu and Wang (2017)	Distilled	AC (commercial)	1234	Sulfamethoxazole	50–500	90	417	–	300–302	Langmuir	Pseudo-second
Darweesh and Ahmed (2017)	Various	GAC from date stones	817	Levofloxacin	50–250	41.4–97.01	100.4	2.0–12.0	298	Langmuir	–
de Franco et al. (2017)	Distilled	GAC (commercial)	463	Amoxicillin	10–1000		0.55–4.39	5.5	298	Sips	Pseudo-second
de Luna et al. (2017)	Various	AC from cocoa pod husks	–	Sodium diclofenac	10–30	76.0–93.6	5.53	3.0–11.0	298	Freundlich	Pseudo-second
El-Shafey et al. (2012)	Distilled	AC from date palm leaves	24.4	Ciprofloxacin	50–300	–	133.3	2.0–11.5	298–318	Langmuir	Pseudo-second
Erdinc et al. (2010)	Distilled	Activated Charcoal	1014	Thioridazine hydrochloride	4.07–4233.17		20.4–2686.4		298	Langmuir	Pseudo-second

Table 3 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceutical	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Ferreira et al. (2015)	Distilled	AC from dende and babassu coconut	484–672	Paracetamol	50	–	65–74	2.0–11.0	298	Langmuir	Pseudo-second
Fukuhara et al. (2006)	Distilled	AC (commercial)	991–1831	17β-estradiol	0.001–0.01	–	0.0003–67.6	5.0	298	Freundlich	–
Galhetas et al. (2014)	Distilled	AC from pine gassification residue	799–1171	Acetaminophen	120–480	35–60	180–222	5.0	293–313	Langmuir	Pseudo-second
Guedidi et al. (2013)	Ultrapure	GAC (commercial, chemically modified)	655–800	Ibuprofen	5–100	–	146.6–256.2	3.0–7.0	298–328	Langmuir–Freundlich	Pseudo-second
Güzel and Saygılı (2016)	Various	AC from grape industrial processing waste	1455	Tetracycline	150–500	–	417–625	5.7	288–308	Langmuir	Pseudo-second
Haro et al. (2017)	Various	GAC (commercial)	543	Atenolol	5–900	88	40	2.0–10.0	298	Freundlich	Pseudo-second
Huang et al. (2015)	Distilled	AC from <i>Thalia dealbata</i>	558–956	Streptomycin	25–400	–	170.8–247.7	2.0–11.0	303–323	Langmuir	Pseudo-second
Ilbay et al. (2015)	Wastewater	AC (plus magnetite)	100–120	Naproxen	1.0–30	67.2–89.8	8.06–87.79	3.0–11.0	293–323	Langmuir	Pseudo-second
Iovino et al. (2015)	Deionized	AC from bituminous coal (commercial)	1000	Ibuprofen	10	–	34	2.0–10.0	277–307	Langmuir	–
Jain et al. (2014b)	Distilled	AC from pine cone and deoiled canola meal	132–980	Acyclovir	400	39.5–90.3	–	4.0–11.0	288–318	–	–
Jain et al. (2014a)	Distilled	Powdered activated charcoal	1097	Acyclovir	100–400	70–90	20.2	3–11	298–318	Freundlich	Pseudo-first
Jodeh et al. (2016)	Distilled	AC from <i>Cyclamen persicum</i>	799–880	Diclofenac	0–50	–	22.22	2.0–12.0	277–307	Freundlich	–
Kim et al. (2010)	Wastewater	Powdered/granular AC (commercial)	882–1112	Trimethoprim	1.0–30	43–98	239.2–333	4.0–10.0	298	Toth	–
Kim et al. (2016)	Distilled	AC (commercial) and biochar	972–1360	Atenolol	1	–	20.6–37.5	3.5–10.5	298	Langmuir	Pseudo-second
Kong et al. (2017)	Deionized	AC from luffa sponge	834	Ofloxacin	20–80	50.1–98.5	126.74–131.93	2.0–12.0	293–313	Freundlich	Pseudo-second
Kumar and Mohan (2011)	Distilled	Granular activated charcoal	99.5	17α-ethinylestradiol	0.025–0.1	68–78	0.0075	2–10	298–328	Langmuir	Pseudo-first
Kyzas et al. (2013)	Various	AC from wood	471–2327	Pramipexole dihydrochloride	0–200	–	117	3.0–9.0	298–338	Langmuir–Freundlich	Pseudo-second

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Table 3 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceutical	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Kyzas and Delyianni (2015)	Various	AC from potato peels	N/A	Dorzolamide	0–200		52–92	2.0–12.0	298–338	Freundlich	Pseudo-second
Larous and Meniai (2016)	Various	AC from olive stones	84	Diclofenac	25–150		0.45–8.8	2.0–10.0	296	–	Pseudo-second
Ledesma et al. (2010)	Distilled	AC (commercial)	710–966	Amitriptyline	3.2–75.3	–	58.7–134	3–7	298	Langmuir	–
Li et al. (2013)	Distilled	AC from Iris tectorum	1371	Tetracycline	324.7–737.8	99	625–769	2–11	295	Langmuir	Pseudo-second
Limousy et al. (2016)	Ultrapure	AC from olive stones	1174	Amoxicillin	12.5–100	93	11.1–67.7	3.2–3.6	293–298	Sips	Pseudo-second
Liu et al. (2011)	Distilled	AC from lotus stalk	140–1289	Norfloxacin	29–87	–	6.9–294.7	3.5–10.5	293	Langmuir	Pseudo-second
Liu et al. (2012)	Distilled	AC from lotus stalk	125–1114	Trimethoprim	29–87	–	118.6–333.2	3–7.5	293	Langmuir	–
Liaó et al. (2015)	Ultrapure	AC from bituminous coal and peat	260–1234	Paracetamol	1–150		53.8–261.0	7.0	298	Langmuir	Diffusion-reaction
Martins et al. (2015)	Various	AC from macadamia nut shells	1524	Tetracycline	250–800		455.3	3.0–10.0	–	Temkin	Elovich
Marzbaei et al. (2016)	Distilled	AC from apricot nut shells	308	Tetracycline	100–200		308.3	1.5–8.5	308–338	Freundlich	Pseudo-second
Mashayekh-Salehi and Moussavi (2015)	Distilled	AC from pomegranate wood	1029	Acetaminophen	50–500	35–98	193–233	2.0–12.0	283–313	Langmuir	Pseudo-second
Mestre et al. (2009)	Ultrapure	AC from cork powder and PET residue (plastic)	879–1426	Ibuprofen	20–120		145.2–430.4	4.0–7.0	303	Langmuir, Freundlich	Pseudo-second
Mestre et al. (2016)	Various	AC from coconut shell and wood	758–907	Clofibrac acid	20–150	>70	82.6–433.2	3.0, 8.0	303	Langmuir	Pseudo-second
Miao et al. (2016)	Distilled	AC from alligator weed	736	Cephalexin	5.0–70		38–45	2.0–12.0	288–308	Langmuir	Pseudo-second
Mondal et al. (2015)	Distilled	AC from mung bean husk	405	Ranitidine hydrochloride	100	99.16	26.5	2.0–12.0	298–318	Sips	Fractionary order
Moussavi et al. (2013)	Distilled	AC from pomegranate wood	1024	Amoxicillin	10–100	54–92	262–247	2–9	283–328	Langmuir	Pseudo-second
Nabais et al. (2012)	Distilled	AC from cork, coffee and eucalyptus pulp	598–839	Amitriptyline	255	–	20–120	1–13	298	Langmuir	–
Nazari et al. (2016)	Distilled	AC from walnut shell	1452	Cephalexin	100–200		233.1	1.5–8.5	303	Freundlich, Toth	Pseudo-second

Table 3 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceutical	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Onal et al. (2007)	Distilled	AC from apricot waste	1060	Naproxen	100–500	–	106.4	5.82	298–323	Langmuir	Pseudo-second
Otero et al. (2004)	Distilled	AC (commercial)	1050	Salicylic acid	100	–	350	–	293–333	Nitta	Pseudo-second
Peng et al. (2015)	Various	AC from bamboo	509–1746	Ciprofloxacin	20–100	–	153.2–369.3	2.0–12.0	298–318	Langmuir	Pseudo-second
Pezoti et al. (2016)	Distilled	AC from guava seeds	18–2573	Amoxicillin	400–800	–	570.48	3.0–9.0	298–328	Redlich-Peterson	Elovich
Quesada-Peñate et al. (2009)	Deionized	AC from wood, coconut shell, casuarina	1175–1860	Levodopa	31–1281	–	285.3–393.3	5.5–6.5	298	Freundlich	–
Rajapaksha et al. (2014)	Distilled	Activated biochar from tea waste	0.9–576	Sulfamethazine	2.5–50	–	0.54–33.8	3–9	298	Freundlich	–
Rajapaksha et al. (2015)	Distilled	Activated biochar from <i>Sicyos angulatus</i> L.	0.85–7.5	Sulfamethazine	2.5–50	46–95	6.69–37.7	3–9	298	Temkin	–
Rigobello et al. (2013)	Artesian well	AC from babacu coconut shell	N/A	Diclofenac	1	–	–	6.5–6.7	293	Freundlich	–
Román et al. (2012)	Deionized	AC from almond tree pruning	284–870	Fluoxetine	1000	–	111.3–224.4	6.0–7.0	298	Langmuir	–
Ruiz et al. (2010)	Ultrapure	AC from coal (commercial)	889–1033	Paracetamol	120	–	13.9–48.9	5.8	303	–	Intraparticle diffusion
Saygili and Guzel (2016)	Distilled	AC from tomato industrial processing waste	1093	Tetracycline	200–400	–	73.7–500	5.7	288–308	Langmuir	Pseudo-second
Shimabuku et al. (2016)	Various	PAC (commercial), biochar from pine forestry waste	39–697	Sulfamethoxazole	50 ng/L to 1	8–98	–	7.2	–	–	–
Stoykova et al. (2013)	Distilled	Activated charcoal	267–452	Carbamazepine	1–20	25–100	4.46–14.50	–	–	Langmuir	–
Sulaiman et al. (2017)	Wastewater	Powdered/granular AC (commercial)	N/A	Diazepam	0.01–100	–	1.1–31.2	7.32	288	Langmuir	–
Sun et al. (2016)	Distilled	AC from <i>Arundo donax</i> linn and pomelo peel	675–1252	Ciprofloxacin	100–800	–	244–400	2.5–11.0	298	Langmuir	Pseudo-second
Taheran et al. (2016)	HPLC	Biochar from pinewood	14.9–853	Chlortetracycline	0.2–150	–	181.8–434.8	1.0–9.0	298	Langmuir	–
Wang et al. (2015)	Distilled	AC from bamboo	2237	Ciprofloxacin	8.42–84.2	–	108–613	2.0–12.0	298	Langmuir	Pseudo-second

Table 3 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceutical	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Wang et al. (2017)	Deionized	AC from bamboo	1228	Ciprofloxacin	0.5–70		36.02	2.5–9.5	298	Langmuir	Pseudo-second
Wuana et al. (2015)	Distilled	AC from <i>Moringa oleifera</i> pod husks	182–236	Ofloxacin	10–50	25–76	1.0–3.5	2.0–11.0	298–328	Langmuir	Pseudo-second
Wuana et al. (2016)	Distilled	AC from <i>Moringa oleifera</i> pod husks	182–236	Norfloxacin	10–50	55–90	1.4–1.9	2.0–11.0	298–328	Langmuir	Pseudo-second
Yi et al. (2016)	Various	Biochar from rice husk and wood chip	1.2–312	Levofloxacin	7.5–150		1.49–7.72	2.0–9.0	303	Langmuir	Pseudo-second
Zhang et al. (2015)	Drinking	AC from petroleum coke	11.2–1122	Tetracycline	60–200		897.6–1121.5	1.0–10.0	303–323	Freundlich	Pseudo-second
Zhang et al. (2017)	Distilled	AC from residue of desiccated rice husk	1020	Ciprofloxacin	150–350		461.9–475.7	3.0–9.0	298–318	Langmuir	Pseudo-second
Zhang and Zhou (2005)	Various	AC (commercial)	600	17 β -estradiol	0.0248	80	12.2–116	2–11	–	–	–

ACs with varying physical and chemical properties. AC can be prepared either in a powdered (PAC) form or in a granular (GAC) form (Kim et al. 2010). The primary difference between PAC and GAC is particle size; the former has typical diameters of less than 0.1 mm compared to 1.2–1.6 mm for the latter (EPA 2017a, b). AC, in either form, has been reported to achieve high removal efficiencies of several organic compounds, including pharmaceuticals, in laboratory, pilot and full-scale tests (Ek et al. 2014; Li et al. 2011; Nowotny et al. 2007). In this section the effects of the type of precursors used to produce AC, the adsorbent preparations method, the adsorbent and adsorbate properties on the efficiency of adsorption of pharmaceuticals are considered.

3.1 Type of precursors

The existing literature contains a myriad of studies that have experimented with the adsorption efficiency of AC prepared from countless precursors. Precursor material yield is affected by its carbon and ashes content, both of which contribute to the effectiveness of a material as AC (Lladó et al. 2015). Factors that should be taken into account when selecting precursor material for AC are: inorganic matter content, availability, cost, degradation rate during storage, and the ease of activation and regeneration.

Numerous studies have addressed the variety of precursors that can be used to prepare AC with the focus in recent years being directed towards the use of waste material and biomass. The waste material precursors investigated include: peach stones (Cabrita et al. 2010), de-oiled canola meal (Jain et al. 2014a), cocoa shells (Saucier et al. 2015), cocoa pod husks (de Luna et al. 2017), macadamia nut (Martins et al. 2015), coffee residue (Flores-Cano et al. 2016), almond shell (Flores-Cano et al. 2016), olive stones (Limousy et al. 2016), date pits (Belhachemi and Djelaila 2017; Darweesh and Ahmed 2017), grape industrial processing waste (Güzel and Saygılı 2016) and tomato industrial processing waste (Saygili and Guzel 2016). Similarly, other studies have looked into the use of biomass material for AC precursors such as: pinecones (Jain et al. 2014a), date palm leaflets (El-Shafey et al. 2014), bamboo (Reza et al. 2014; Wang et al. 2015), loofah scraps (Kong et al. 2015), and alligator weed (Miao et al. 2016). Furthermore, other studies have experimented with different AC types: oxidized AC

Table 4 Summary of activated carbon adsorption studies for more than one pharmaceutical

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Ahmed and Theydan (2014)	Distilled	AC from <i>Albizia lebbek</i> seed pods	1843	Ciprofloxacin	20–100	55–96	131.14	2–12	303–323	Langmuir	Pseudo-second
Álvarez-Torrellas et al. (2015)	Ultrapure	AC from peach stone	959–1216	Norfloxacin Diclofenac	100	55–98	166.99 90–180	6.3	303	Sips	–
Álvarez-Torrellas et al. (2016)	Ultrapure	Granular AC (commercial)		Carbamazepine Ibuprofen	10–100		175–335 141.2–2242.9	–	293–303	Guggenheim–Anderson–Boer	Pseudo-second
Awwad et al. (2015)	Wastewater	AC from peach stones and rice husk	N/A	Tetracycline	100–500	96.5	192.7–56,000		298	Langmuir	–
Bo et al. (2016)	Distilled Distilled Wastewater	Granular AC AC from coconut shell	916	Amoxicillin Cefuroxime axetil Clotbric Acid Carbamazepine Naproxen Diclofenac	20–300 0.5 0.5 0.5	96 63	3.3–31.25 2.48 3.00 2.74 2.52	8.2	–	Langmuir	–
Calisto et al. (2015)	Ultrapure	Powdered AC (commercial)	209–848	Carbamazepine	5	85.5	12.6–116	NA	298	Langmuir	Pseudo-second
Delgado et al. (2015)	Ultrapure	Powdered AC from coal and wood	1012–1491	Oxazepam Sulfamethoxazole Piroxicam Cetirizine Venlafaxine Paroxetine Atenolol Ibuprofen	0.08	7.8–119 1.6–118 5.2–138 8.2–101 8.5–42.5 38–90 0.07615–0.08562	3.0–11.0	298	Freundlich Polanyi–Dubinin–Manes	–	

Table 4 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
dos Reis et al. (2016)	Various	PAC (sludge based)	21.2–679.3	Nimesulfide	5–500		40.83–66.45	6.0–11.0	298–343	Sips	General order
Dutta et al. (1999)	Distilled	AC	920	Sodium diclofenac 7-Aminocephalosporanic acid	2722.78–5445.56		86.73–157.4 25.86–194.95	3.0–9.0	298–328	Langmuir	Pseudo-first
El-Shafey et al. (2014)	Various	AC from date palm leaflets	48–405	Cephalexin Cefadroxyl 6-Aminopenillanic acid Fexofenadine	3473.9–6947.8 3633.9–7267.8 2162.5–4325 10–250		34.04–288.33 33.79–272.54 43.25–112.66 23.92–32.89	2.0–11.0	293–318	Langmuir	Pseudo-second
Essandoh et al. (2015)	Distilled	Pine wood biochar	1.35	Diphenhydramine Salicylic acid	25–100		42.92–62.11 7.56–22.7	2.0–10.0	298–318	Sips, Toth	Pseudo-first
Fernandez et al. (2015)	Ultrapure	AC from orange peels	0.4–618	Ibuprofen Diclofenac sodium	1.55–62.26		10.74 5.73–67.12	2.0–7.0	298	Langmuir	Pseudo-second
Flores-Cano et al. (2016)	Various	AC from coffee residue/almond shells	103–222	Salicylic acid Flurbiprofen Metronidazole	6.91–276.24 12.21–488.52 25–250		12.43–91.16 148.99–202.74 22.25–152.32	7	298	Langmuir	Pseudo-first
Gao and Deshusses (2011)	Ultrapure	Powdered AC	982	Dimetridazole Diatrizoate Clofibrac acid	42.93–300.51		12.70–39.52 171.89–399.04 70–140	7	NA	Langmuir	Intra-particle diffusion
Ifebaegu et al. (2015)	Distilled	Granular AC (commercial)	N/A	Ketoprofen 17 β -estradiol	20.34–50.86 2	20–97	70–120 4.01	3–9	298	Langmuir	Pseudo-second
Liu et al. (2017)	Ultrapure	AC from maize straw	1200	17 α -ethinylestradiol Metronidazole	5–50	20–97	2.97	110.64	298	Langmuir	Pseudo-second

Table 4 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Meinel et al. (2016)	Wastewater	Powdered AC	N/A	Norfloxacin	0.00218	39–98	56–98	112.86	NA	Freundlich	
				Sulfadiazine	0.00052	31–98		147.12			
				Tetracycline	not detectable			85.29			
				Pramipexole	0.00085	12.0–84					
				Benzotriazole	0.0014						
Meinel et al. (2015)	Wastewater	Granular/powdered AC	N/A	Carbamazepine	0.0012		0.0099–0.0301	NA	NA	Freundlich	Pseudo-single
				Sulfamethoxazole 4-formylaminoantipyrine	150	12.0–84	0.0104–0.027 141.97–273.08	NA	NA	Freundlich	Pseudo-second
Mendez-Diaz et al. (2010)	Various	AC from petroleum cake	848–1301	Carbamazepine Dimetridazole	0.0012 150			6.0–7.0	298	Langmuir	Pseudo-second
Mestre et al. (2014)	Ultrapure	AC from pre-treated cork	750–1065	Metronidazole	20–150	15–80	168.24–300.54 222.56–408.31 272.00–432.23	5.0–7.0	293	Langmuir	Pseudo-second
				Ronidazole							
				Tindazole							
				Ibuprofen							
				Paracetamol							
Mestre et al. (2015)	Ultrapure	AC from sucrose	694–2431	Acetylsalicylic acid	45–300	60–98	117.2–169.5	3.0–5.0	293	Langmuir	Pseudo-second
				Clofibric acid							
				Iopamidol							
				Ibuprofen							
				Paracetamol							
Moral-Rodriguez et al. (2016)	Various	GAC (commercial)	919	Acetylsalicylic acid	100–1000	87	267–514				
				Clofibric acid							
				Iopamidol							
				Ronidazole							
				Sulfamethoxazole	40–200		212.75–542.02				

Table 4 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Nielsen et al. (2014)	Deionized	AC from coconut and wood	1042–1648	Carbamazepine	1–100		300.06–333.14	4.57–7.5	303	Langmuir–Freundlich	–
Ocampo-Pérez et al. (2015)	Ultrapure	Granular AC (commercial)	1225–1301	Sulfamethoxazole Tetracycline	100–1000		1902.13–6724.56 375.1–471.1	4.0–5.0	298	Langmuir	Pseudo-first
Ogata et al. (2012)	Wastewater	AC from Shirasagi and coal	807–1391	Oxytetracycline Chlortetracycline Sulfamethoxazole	0.4–4		252.6–413.2 65.1–309.9 92.8–160.9	5.19–5.55	298	Langmuir, Freundlich	Pseudo-second
Pouretedal and Sadegh (2014)	Distilled	AC from vine wood	13.4–54.8	Sulfamonomethoxine Sulfadimidine Sulfadimethoxine Amoxicillin	20–200	26–60	91.1–213.6 108.4–153.9 100.2–577.8 2.69	1.0–12.0	308–328	Langmuir	Pseudo-second
Rakic et al. (2015)	Deionized	Granular/powdered AC	650–1500	Cephalexin Tetracycline Penicillin G Salicylic acid	414,363	43–76 57.7–88.2 37.0–73.94	7.08 1.98 8.41 26.24–70.44	2.82–10.15	303	Sips	–
Reza et al. (2014)	Ultrapure	AC from bamboo waste	722	Acetylsalicylic acid Atenolol Diclofenac Ibuprofen	540,471 799,008 888,444 100		25.22–45.04 31.96–111.86 59.23–183.61 278.5	2.0–5.0	298–313	Langmuir	Pseudo-second
Rivera-Utrilla et al. (2009)	Distilled	AC (commercial)	848–1225	Clofibrate acid Metronidazole	50–600	–	229.35 213.94–328.6	2–11	303	Langmuir	–
	Surface Ground Wastewater			Dimetridazole Tinidazole Ronidazole			186.3–287.9 257.2–385.7 364.3–394.3				

Table 4 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	Kinetic model
Rivera-Utrilla et al. (2013)	Various	AC (commercial)	1200	Tetracycline	100–1000		375.4–471.1	2.0–11.0	298	Langmuir	–
Rovani et al. (2014)	Ground Wastewater			Oxytetracycline			252.6–413.2				
	Deionized	AC from agro-industrial waste	16.5	Chlortetracycline 17β-estradiol	2		65.1–309.9 7.584	2.0–12.5	298	Sips	General order
Saucier et al. (2015)	Wastewater	AC prepared from cocoa shell	1.1–619	17α-ethinylestradiol	10–300	0.17–97.05	63.47	7.0–10.0	298–323	Liu	General order
	Ultrapure	Biochar from agricultural waste	30.9–486	Nimesulfide Carbamazepine	5.0–60	0.21–98.75 37–80	74.81 62.7–135.1	4.0–9.0	NA	Langmuir	–
Sun et al. (2017)	Deionized	Biochar from sugarcane bagasse	1.6–388.3	Tetracycline Diclofenac sodium	5–20	20–50	45.3–94.2 20.8–329.0				
				Dimetridazole	5–20	98.0	69.65–72.17	6.67	303–323	Brunauer–Emmett–Teller	Pseudo-second
Wong et al. (2016)	–	Powdered AC from palm	555.5–960.9	Metronidazole Carbamazepine	10.0–50	88.8	16.63–23.61 107.6–114.3	6.62 7	303–323	Freundlich	Pseudo-second
	Ultrapure	Granular AC from	1030–1156	Ibuprofen Clotbric acid Naproxen	5E – 5–0.0005		117.9–166.7 58.1–78.7 2–3.2	7.5–7.9	296	Freundlich	–
Zuo et al. (2016)	Varrious	PAC (commercial anthracite) and coconut shell based	805–2514	Carbamazepine Sulfamethoxazole	7.60–119.04		1.5–3.2 165.64–572.41	6.0	298	Freundlich	–
				Sulfapyridine	19.94–117.17		136.11–563.40				

Bhadra et al. 2016), magnetic nanocomposites (Baghdadi et al. 2016), and triethoxyphenylsilane functionalized magnetic palm based PAC (Wong et al. 2016). Lingocellulosic materials are also common precursors as they exhibit low inorganic content (and thus low ash content) but relatively high volatile content; these materials include wood, sawdust, nutshells, and fruit stones (Álvarez Torrellas et al. 2015). The focus of this review will be on ACs produced either commercially or from various types of carbonaceous precursors.

In view of the different precursors used to produce AC, an important consideration is the effectiveness of these ACs in terms of removal efficiency, compared to commercially produced ACs. The literature offers contradictory data where in a number of instances the waste/biomass-derived AC outperformed commercial AC, whereas in several other cases the opposite was true. For example, Bernardo et al. (2016) compared diclofenac adsorption onto commercial AC and onto potassium carbonate (K_2CO_3) activated potato peels, with the former resulting in the greater uptake. On the other hand, Mestre et al. (2009) found that waste derived AC resulted in greater adsorption compared to commercial AC. Álvarez-Torrellas et al. (2016) studied the adsorption of ibuprofen and tetracycline onto GAC and two ACs prepared from rice husk and peach stone. Ibuprofen exhibited the greatest adsorption onto rice husk derived AC, while tetracycline removal was greatest with peach stone derived AC. Bhadra et al. (2016) studied diclofenac adsorption using oxidized AC and found it removed six times the amount removed by the commercial AC. Aside from these discrepancies, Ziska et al. (2016) found that minimal removal differences were noted when different PAC materials were tested. Overall, and for laboratory scale studies, there is no clear-cut generalization that favors commercial AC over waste/biomass-derived AC, or vice versa.

3.2 Adsorbent preparation method

It has been stated that two of the main factors influencing the removal efficiency of a particular AC are the porous structure and the surface functional groups. In turn, these factors are affected by and can be manipulated through the activation process: type of activation, activating agent, activation process temperature, and impregnation ratio (weight of activating

agent to carbon material). The activation can be used to tailor surface chemistry, particularly in the formation of basic/acidic functional groups that can promote the uptake of pharmaceuticals with compatible properties (Fernandez et al. 2015).

The activation of AC from carbonaceous sources can take place in two ways: physical (thermal) or chemical treatment. Physical treatment consists of a two-step process whereby the material undergoes carbonization at a relatively low temperature in the absence of air, followed by activation in an oxidizing atmosphere (gasification) (Álvarez Torrellas et al. 2015); the second step is crucial in order to obtain a surface area large enough to facilitate adsorption. In terms of physical treatment, carbon dioxide and steam have been shown to be suitable for micro and mesopore development (Román et al. 2012). In some cases, using air increases oxygen content (in functional groups) on the surface and increases both surface area and mesopore volume, but may lower yield. Meanwhile, chemical activation consists of mixing the raw material with an inorganic activating agent, soaking the carbonaceous material in a dehydrating agent (drying at 100–120 °C), and then activating it under high temperatures in an inert atmosphere (carbonization at 400–800 °C) (Saucier et al. 2015). Alkali hydroxides such as potassium hydroxide, KOH (Zhang et al. 2015), and sodium hydroxide, NaOH (Martins et al. 2015), are widely used chemical activating agents, but they have several disadvantages including hazardousness, corrosiveness, and high costs. For this reason the use of potassium carbonate (K_2CO_3) has increased recently as it is more benign in application (Wang et al. 2015). Phosphoric acid (H_3PO_4) has also been used as an activating agent and its major effect is that it results in an increased concentration of the acidic groups on the carbon surface (Limousy et al. 2016). The advantages of chemical treatment over the physical are: comparatively low temperature, shorter heat treatment, and higher carbon yield; however, these are usually counterweighed by high activating agent costs and an additional processing step (washing).

The process by which the carbon is prepared also affects adsorption, as illustrated by the Kyzas and Deliyanni (2015) study, where potato peels were used to create the adsorbent material; in one scenario, the peels underwent carbonization, and in the other hydrothermal treatment. The adsorption capacity of

the hydrothermally treated potato peels was greater than that of carbonized peels, with approximately a 17% enhancement. Huang et al. (2011) compared the performance of AC prepared from lotus stalk via microwave heating versus AC prepared by conventional heating and H_3PO_4 activation. The former resulted in greater surface area but lower micropore volume than the latter. Microwave heated AC was found to have smaller acidic oxygen functionalities; these textural and chemical characteristics resulted in different adsorption capacity of oxytetracycline. Jodeh et al. (2016) compared diclofenac adsorption onto *C. persicum* tubers based AC that was activated either by physical or chemical processes. The chemically activated carbon had greater surface area than the physically activated one, and exhibited improved adsorption capacity. This highlights the need for further experimentation in terms of determining the extent that the activation method can affect adsorption capacity and whether variations in the conditions investigated may provide a better understanding of how to manipulate the pre-adsorption phase in a way that may improve pharmaceutical removal rates.

It should be noted that as the preparation processes differ, different reagents and equipment are required to produce the AC and, if applicable, to regenerate the spent AC which subsequently results in different marginal/unit costs (Ahmed et al. 2015). Cost is one of the determining factors for adsorbent usage in terms of the required treatment, regeneration, and solvent recovery. Commercial grade AC is known to be effective but expensive, highlighting the need for alternative adsorbents at more competitive and sustainable costs (Reza et al. 2014). Estimation of the production cost of AC varies between different sources and depends on a number of factors including but not limited to: the precursor used, the activation method employed and the regeneration/reuse method. It is extremely difficult to find accurate information on the costs of production as this is often considered proprietary information. Costs reported in the open literature range between 1 and 2.89 \$/kg (Lima et al. 2008; Ng et al. 2003; Toles et al. 2000).

3.3 Adsorbent properties

AC adsorption capacity is influenced by several adsorbent properties such as surface functional

groups, pore size distribution, surface charge and BET surface area (de Ridder et al. 2010).

Functional groups, acidic and basic, affect the surface charge of AC, and thus its adsorption properties. In most cases, the AC has a hydrophobic surface, but it can contain oxygenated functional groups once it undergoes an activation process. As the AC surface contains more oxygen containing functional groups, adsorption of organic compounds decreases, as these sites tend to favor water molecule adsorption over organic compounds. It seems that one of the most important surface functional groups is the carbon–oxygen surface group due to the influence it has on wettability, polarity, acidity, and reactivity of the surface (Álvarez Torrellas et al. 2015). While the presence of oxygen containing functional groups facilitates adsorption, these groups can also promote Hydrogen-bond donor and acceptor interactions between the adsorbates and adsorbent surface (de Ridder et al. 2010). However, it was reported that the removal rates of certain types of pharmaceuticals, such as ibuprofen and atenolol, were low when ACs with high oxygen containing functional groups were used (Delgado et al. 2014). In addition, the presence of chlorine, nitrogen, and sulfur groups on the surface of some ACs can affect their interaction with the adsorbed compounds (Delgado et al. 2012). Similarly, phenolic groups on the surface of the AC dictate affinity towards the pharmaceuticals; these groups increase the electron density of AC and thus promote pi–pi dispersive interactions, elevating adsorption capacity (Flores-Cano et al. 2016). High adsorption is also supported by Hydrogen-bond formation between phenolic groups on the pharmaceuticals and oxygen groups on the carbon surface (Rivera-Utrilla et al. 2013). In cases where the adsorbent surface is hydrophilic, solute removal is lower than in cases where the adsorbent is hydrophobic.

AC has a pore structure characterized by three types of pore volume: micropores (for molecule adsorption), mesopores (for molecule transportation), and macropores (for the entrance of the molecules onto AC) (Yu et al. 2016). Mesopores refer to the transport pores network that ensures accessibility of the adsorbate to the inner pores of the carbon. Micropores on the other hand are involved in the uptake of pharmaceuticals to the active adsorption site; the accessibility of these pores is determined by the pore size and adsorbate size (Ruiz et al. 2010). Cabrita et al. (2010) reported that

AC derived from plastic waste had adequate micropores and the highest affinity to acetaminophen when compared to biomass derived AC but exhibited the lowest removal efficiency and slow kinetics. On the other hand, biomass derivatives appeared to have large micro and mesopore structures with greater hydrophilic nature resulting in fast removal and large adsorption capacity. Mestre et al. (2014) considered the pore size distribution of the adsorbent and its role in the adsorption of pharmaceuticals with different structures. The results showed that chemically activated ACs with a well-developed micropore structure were able to remove 80–100% of the small molecules such as ibuprofen, paracetamol, acetylsalicylic acid and clofibrac acid, whereas ACs with a mesopore structure and higher volume of super-micropores were required for the efficient removal of bulkier compounds such as iopamidol.

Another significant parameter is the pH at point of zero charge (pH_{pzc}). Activated carbon can have an acidic, basic or neutral nature depending on the precursor type, the activation mode (chemical or physical) and pH_{pzc} . The pH_{pzc} of activated carbon is characteristic of amphoteric surfaces and is determined by the chemical and electronic properties of surface sites. The pH_{pzc} plays an important role in determining surface properties. For a given activated carbon, the surface of carbon is neutral at $\text{pH} = \text{pH}_{\text{pzc}}$, negatively charged for pH higher than pH_{pzc} and positively charged at pH below the pH_{pzc} (Pouretedal and Sadegh 2014). As such, pH_{pzc} influences the protonation/deprotonation of functional groups on the carbon surface, which can change the electric charge (Nielsen et al. 2014).

In general, increasing the surface area increases the availability of adsorption sites for the adsorbate (Larous and Meniai 2016; Mailler et al. 2016). Commercial AC, which has a greater surface area, was able to adsorb more diclofenac than a potato-peel AC; however, the latter had a higher adsorption rate because of its hydrophilic nature (Bernardo et al. 2016). On the other hand, when comparing oxidized AC to non-oxidized AC, Bhadra et al. (2016) found that the former had a greater adsorption capacity as a result of the acidic functional groups created upon oxidation even though it possessed the lower surface area. Similarly, Essandoh et al. (2015) examined the adsorption of salicylic acid and ibuprofen onto pine wood char and compared it to commercial AC, and

found that the lower surface area of pine wood char had a larger adsorption capacity than the commercial AC. Thus, it can be observed that surface area may not necessarily be directly linked to adsorption capacity (Mestre et al. 2009). AC produced from olive stones was found to have relatively low surface area, but high adsorption capacity (Larous and Meniai 2016).

The interaction between adsorbent properties and their effects on the adsorption capacity is rather complicated as there are several contradictory results in the literature. The influence of one property changes from system to system, and from pharmaceutical to pharmaceutical. For instance, ibuprofen recovery was highly dependent on the adsorbent's textural properties, whereas in the case of tetracycline, it was the chemical nature of the adsorbent which had the greater influence (Álvarez-Torrellas et al. 2016). Hence, adsorbent characteristics can be studied for the possibility of surface structure modification with novel functional groups that will promote selectivity and specificity, and thus improve adsorption capacity (Ahmed et al. 2015). The interlinking relations between the textural and chemical properties of the adsorbent and the extent of the role they play in promoting adsorption should be investigated further.

3.4 Adsorbate properties

The physical and chemical properties of the pharmaceuticals also appear to have a strong impact on adsorption; these include hydrophobicity/hydrophilicity, molecular size and structure, charge, solubility and acid dissociation constant (pKa).

The nature of the pharmaceutical compound (hydrophobic/hydrophilic) affects its ability to adsorb; hydrophobic compounds tend to have a higher affinity for adsorption compared to hydrophilic pharmaceuticals. Some ACs have been found to be well suited for the removal of non-polar or moderately polar compounds, but often exhibit relatively low adsorption capacities with polar compounds (Kovalova et al. 2013). Generally, all positively charged pharmaceuticals can be removed, regardless of their hydrophobicity, whereas the removal of negatively charged and neutral compounds is dependent on their hydrophobicity. Meanwhile, the hydrophobicity of the adsorbate is related to the octanol–water partitioning coefficient, ($\log K_{\text{ow}}$) such that the greater the hydrophobicity (larger $\log K_{\text{ow}}$), the higher the

removal. This illustrates the significance of hydrophobic interactions for negatively charged compounds in wastewater (Margot et al. 2013). When hydrophobic partitioning is less relevant (small solute, hydrophilic, charged, polar), the correlation between $\log K_{ow}$ and adsorption capacity is poor. The correlation between $\log K_{ow}$ and adsorption is strong when both the solute and adsorbent are hydrophobic (de Ridder et al. 2010). However, although $\log K_{ow}$ can be indicative of hydrophobic interaction, when the solution is within a neutral pH range, hydrophobicity is greatly impacted by the ionization of weak acids and bases (Ziska et al. 2016). Hence, it is often reported that hydrophobic compounds (high $\log K_{ow}$) are more favorably adsorbed onto nonpolar AC surfaces (Lladó et al. 2015). In particular, Lladó et al. (2015) concluded that an adsorbate's hydrophobic character does not impact its sorption propensity, but may affect its uptake rate.

Molecular size plays a role in determining which compounds are removed the fastest, such that compounds with the smallest molecular sizes are adsorbed more quickly, because they have more access to the AC pores (Delgado et al. 2014; Kovalova et al. 2013). Smaller molecules can diffuse more deeply into the carbon and adsorb to more surface area compared to larger molecules, which are restricted by pore size (Yu et al. 2016), so that compounds with small molecular sizes such as carbamazepine, sulfamethoxazole, and ketoprofen, are removed more easily than their larger molecular sized counterparts (Nebout et al. 2016). It is evident that the carbon pore size and the molecule size place restrictions on the accessibility of the adsorption sites and therefore ACs with naturally larger micropore sizes and mesopore volumes allow for greater access. Rivera-Utrilla et al. (2013) validated this notion by observing that among the studied pharmaceuticals (tetracycline, oxytetracycline, and chlortetracycline), the molecule with the smallest size, tetracycline, was the most adsorbed, whereas the largest molecule, chlortetracycline, was the least adsorbed. Meanwhile, although Ocampo-Pérez et al. (2015) studied the same compounds and observed that adsorption capacity decreased in the same order of tetracycline, oxytetracycline, and chlortetracycline, they concluded that molecular size differences were very slight and could not explain the differences in adsorption propensity.

Other properties that influence adsorption include: solubility, aromaticity, and functional groups

(Delgado et al. 2014). For example, nimesulide has a higher sorption capacity than diclofenac because it possesses a more polar surface area that interacts with the polar parts of the carbon surface (OH groups, COOH) (Saucier et al. 2015). Furthermore, compound solubility can determine how quickly a compound is removed, such that poorly soluble compounds (carbamazepine, sulfamethoxazole, ketoprofen) are more easily removed than compounds with a high affinity to water (fluoxetine, terbutaline, metoprolol) (Nebout et al. 2016). On the other hand, the lack of charge on neutral AC decreases its affinity to compounds with high solubility leading to low adsorption capacity (Nebout et al. 2016). Even the same compound can behave differently when its properties change. For example, the non-ionized species of ibuprofen has higher adsorption capacity than the ionized one (Iovino et al. 2015). Some compounds possess more than one property that facilitates their adsorption, as is the case for carbamazepine, which generally has a large adsorption propensity, resulting from the joint effect of its higher hydrophobicity and lower water solubility (Álvarez Torrellas et al. 2015).

4 The effect of operating conditions on adsorption

The following sections will address the impact that various operating conditions (adsorbent dosage, temperatures, pH, water source, ionic strength of solution and organic material content) have on the adsorption process.

4.1 Adsorbent dosage

Studies have demonstrated that the adsorption capacity of the AC and the amount of pharmaceutical removed increases with an increase in adsorbent dosage, mainly due to the increased availability of sorption sites at the higher dosage values. Eventually, at equilibrium, all active sites become saturated and the adsorption capacity reaches a plateau value at a certain quantity of activated carbon (İlbay et al. 2015; Mondal et al. 2015; Pouretedal and Sadegh 2014; Yu et al. 2008) beyond which the adsorption capacity itself decreases. This decrease arises, probably, due to the decrease in the total adsorption surface area available to the compounds, as a result of the overlapping/aggregation of adsorption sites

(Pouretedal and Sadegh 2014). Increasing AC dosage causes a greater adsorbent to adsorbate ratio, rendering the optimum dosage dependent on initial pharmaceutical concentration, whereby higher initial concentrations require greater carbon dosages (Mashayekh-Salehi and Moussavi 2015).

Several studies have been performed using different AC dosages, upholding the notion that increased dosage leads to increased adsorption (Kovalova et al. 2013; Rossner et al. 2009; Yoon et al. 2003). Kong et al. (2015) observed a similar trend but reported that the equilibrium adsorption amount decreased with increasing AC, most likely due to the competitive adsorption effect brought upon by abundant amounts of adsorbent. Larous and Meniai (2016) found that while increasing adsorbent dosage, adsorption efficiency increases, but adsorption per unit mass decreases. On the other hand, varying PAC dosage did not affect removal of acetaminophen, caffeine, ibuprofen, and naproxen in the study conducted by Sheng et al. (2016), whereas metoprolol and trimethoprim removals were significantly affected. This study demonstrated that the AC dosage effect (i.e. increased dosage results in increased adsorption) is not consistent for all pharmaceuticals, highlighting a major gap in the literature on varying dosage effects, with respect to certain pharmaceuticals. This also gives rise to the challenge of designing systems that apply to a broad range of pharmaceuticals as classes of pharmaceuticals and, often individual compounds within a class, behave differently under the same operating conditions.

4.2 Temperature

Temperature is an important parameter controlling the adsorption process. The effect that temperature will have is often dictated by the energetics of the adsorption process, i.e. whether the process is endothermic or exothermic. Overall, temperature can affect the adsorption process in two major ways: altering molecular activity at the adsorption interface and interfering with interactions between the solute and adsorbent. Generally, for endothermic processes, high temperatures correlate to an increase in adsorption capacity, as it results in an increase in molecular activity at the boundary layer interface boosting the solute diffusion rate. In cases where temperature results in increased adsorption capacity, the increase is

a result of increasing molecular diffusion rate and decreasing solution viscosity, effects which facilitate molecular diffusion across the external boundary and into the internal pores (Saygili and Guzel 2016). It has been observed that for hydrophobic compounds low temperatures resulted in a substantial reduction in the removal rate of certain pharmaceuticals (Nam et al. 2014).

The effect of temperature varies with the pharmaceuticals considered. For example, cephalexin adsorption is an endothermic process exhibiting a slight increase in adsorption with the increase in temperature (Miao et al. 2016). Huang et al. (2014) found that increasing temperature promoted the adsorption capacity, providing the potential activation energy needed for tetracycline to form activated complexes. However, for ciprofloxacin, adsorption capacity slightly decreased with higher temperatures. Iovino et al. (2015) showed that an increase in temperature decreases ibuprofen dissociation, increasing the non-ionized species, and thus the adsorption capacity; Guedidi et al. (2013) obtained the same result. Kovalova et al. (2013) investigated the effect of temperature on the adsorption of 5-fluorouracil and cytarabine by commercially available PAC; lower temperature resulted in a higher adsorptive uptake. Mondal et al. (2015) studied the effect of temperature on the adsorption of ranitidine hydrochloride onto superheated AC at four different temperatures; the increase in the temperature resulted in a decrease in the removal efficiency of the AC. However, it seems that temperature does not always strongly impact the adsorption process. For example the adsorption of cephalexin was found to be independent of temperature in the study by Kong et al. (2015), while Kong et al. (2017) reported that the effect of temperature on the adsorption of ofloxacin by luffa sponge AC was negligible.

4.3 pH

The pH of a solution directly affects adsorption, and is one of the most important factors controlling the adsorption capacity of activated carbon (Guedidi et al. 2013). The role of pH on the adsorption of pharmaceuticals can be explained by considering the point of zero charge of the adsorbent as well as the disassociation of the functional groups at the specific pH conditions. In other words, at pH values greater than

pH_{pzc} , excess H^+ ions will protonate the adsorbent's surface functional groups. On the contrary, at pH values lower than pH_{pzc} , the presence of OH^- deprotonates these functional groups. Furthermore, although the surface charge of an adsorbent intrinsically depends on its pK_a value, solution pH can alter the surface charge. Pharmaceuticals are essentially neutral molecules at pH below the pK_a value, however, when the solution pH is above the pK_a value, the ionization degree of the pharmaceutical changes due to dissociation of the molecules, which in turn will influence the affinity of the solute towards the adsorbent. Potential adsorption of a compound onto AC due to ion exchange via protonation highlights the interaction between surface chemistry and solution pH (de Ridder et al. 2010).

The adsorption capacity of the AC will vary over a pH range, indicating an optimum pH value for a specific AC (Essandoh et al. 2015; Pouretedal and Sadegh 2014; Zhang et al. 2010). This optimum pH is often a function of the adsorbent type (Saucier et al. 2015). In some cases, almost identical removal rates are obtained within a range of pH values; for example, Nebout et al. (2016) varied the pH from 3 to 7, and found that lower removal was obtained at the lower end of the range, and very similar removal rates were obtained at pH 5 and 7. On the other hand, some pharmaceuticals, such as diclofenac, appear to have a higher propensity for adsorption at the lower solution pH (Jodeh et al. 2016). For Ibuprofen, increasing pH produces electrostatic interactions that repel the anionic ibuprofen species from the negatively charged carbon surface, resulting in minimal adsorption at basic pH and ideal removal under acidic conditions (Iovino et al. 2015; Reza et al. 2014; Wong et al. 2016). For the adsorption of streptomycin (Huang et al. 2015), the adsorption capacity increases in the basic range of the spectrum. On the other hand, in the cases of tetracycline, (Rivera-Utrilla et al. 2013; Zhang et al. 2015) and clofibrac acid (Reza et al. 2014), removal decreases with increasing pH.

However, in other cases, pH does not seem to have any effect on adsorption, such as in the adsorption of acetaminophen where Mashayekh-Salehi and Mousavi (2015) found no significant change in adsorption for the pH range 2–10 (adsorption decreased from 90 to 83%), however, for the pH range 10–12, the adsorption decreased more drastically from 83 to 70% with the increase in pH. Similarly, pH was not found to

have a major impact on the adsorption of several other pharmaceuticals, such as cephalexin (Miao et al. 2016), ciprofloxacin (Wang et al. 2015) and carbamazepine (Jung et al. 2013).

It is widely understood that several system characteristics, specifically pH, are very important in determining adsorption capacity as they contribute to altering adsorbate properties and thus play a role in determining the electrostatic interactions or dispersion forces controlling the mechanism. For example, hydrophobic compound removal is found to be independent of pH, whereas the removal of compounds such as acetaminophen, sulfamethoxazole, and sulfamethazine depends on electrostatic interactions that are affected by pH (Nam et al. 2014).

4.4 Ionic strength

The effect of the ionic strength (a measure of the concentrations of electrolytes) of the aqueous solution, on the adsorption of pharmaceuticals on AC surfaces is not very well understood. The experimental evidence is contradictory where several studies have shown that ionic strength has no effect on the adsorption process (Bo et al. 2016; Kim et al. 2016a), others have reported an increase in the adsorption capacity with the increase in ionic strength (Carrales-Alvarado et al. 2014; Kim et al. 2016b), while others have reported a decrease in the adsorption capacity with the increase in ionic strength (Baghdadi et al. 2016; Li et al. 2013; Mansouri et al. 2015).

It has been postulated that the increase in adsorption with increased ionic strength may be due to one or two mechanisms occurring in series or in parallel: the “salting out effect”, where the solubility of the pharmaceutical decreases with increased salt concentration resulting in an increased availability of the active surface sites and the “screening effect” whereby the electrically charged anions/cations can act as screens to the charged surface of the adsorbent thus eliminating repulsive forces between the pharmaceutical and the AC surface resulting in enhanced adsorption capacity (Kim et al. 2016b; Saygili and Guzel 2016; Wong et al. 2016).

On the other hand, the following mechanisms have been proposed for the decrease in adsorption capacity with increased ionic strength: the “screening effect”, but in this instance, the electrically charged anions/cations interact with the AC surface in such a way so as

to reduce electrostatic attraction between the pharmaceutical and the AC surface, the “competitive effect” where the anions/cations compete with the pharmaceuticals for binding sites and the “water cluster effect” where water adsorption on the surface of the AC reduces the availability of active sites (Baghdadi et al. 2016; Kovalova et al. 2013; Li et al. 2013). It is worth noting that the type and valence of cations present may also have an effect on the adsorption capacity where it has been reported that hardness salts, calcium and magnesium, significantly decreased the adsorption capacity of several types of pharmaceuticals (Coutu et al. 2015; Zhang et al. 2015).

It is extremely difficult to draw definitive conclusions with respect to the effect of ionic strength as the effects often vary for the same pharmaceutical, for example Saygili and Guzel (2016) have reported an increase in adsorption for the antibiotic tetracycline with the increase in ionic strength, while Peng et al. (2015) reported the same trend but observed that beyond a threshold ionic concentration, increasing the concentration of salts in solution had no effect on the adsorption process. On the other hand, Li et al. (2013), Zhang et al. (2015) and Alahabadi et al. (2017) have reported a decrease in adsorption with the increase in ionic strength.

4.5 Organic material in aqueous solutions

The effects background organic material (BOM), whether naturally occurring (NOM) or arising from effluent organic matter (EfOM), on adsorption of pharmaceuticals have been investigated by several researchers and the results reported are not in agreement. Generally, the presence of organic matter is believed to reduce AC adsorption capacity and kinetics thus reducing pharmaceutical removal rates (Delgado et al. 2012). It appears that the impact of NOM on adsorption may vary with the hydrophobicity/hydrophilicity (expressed in terms of the partition coefficient K_{ow}), molecular mass, the chemical charge of the pharmaceuticals in question (de Ridder et al. 2009; Nam et al. 2014; Oh et al. 2013) and with the amount and characteristics of the NOM in the source waters (Altmann et al. 2014; Westerhoff et al. 2005). In addition, the pore size of the adsorbing material as well as the pore size distribution may also have marked effects when NOMs are present. It has been reported that GACs with larger pore size

distribution tend to permit high organic matter loadings, resulting in lower adsorption capacities (Yu et al. 2009), while Mailler et al. (2016) have reported an opposite effect where a mesoporous structure with large pore size distribution limited organic matter competition.

Decrease in removal from surface water, arising from the competitive inhibition of organic matter, was more pronounced for hydrophobic compounds, as opposed to hydrophilic compounds (Nam et al. 2014). Thus, adsorption in surface water is lower than that in deionized water as a result of the competing hydrophobic adsorption of organic matter. Results reported by Rodriguez et al. (2016) and Solak et al. (2014) lend further support to this conclusion. It has been reported that increasing the AC dosage is necessary to compensate for the inhibitory effects of organic matter (Nam et al. 2014). In addition, the presence of low molecular weight (LMW) acid and/or neutral organics is said to explain the adsorption differences between water types (Zietzschmann et al. 2015).

On the other hand, several studies have shown that differing NOM compositions seems to have little impact on the removal of pharmaceuticals by adsorption on AC (Jung et al. 2013; Kim et al. 2016b; Saravia and Frimmel 2008). Zhang et al. (2016) studied the adsorption of 28 antibiotics onto PAC from both deionized and surface water; removal efficiencies in both types of water were high, ranging from 86.8 to 99.9%. Mailler et al. (2016) observed that organic matter present in the water did not necessarily explain competitive adsorption, and that the nature of the organic matter should also be considered. Water containing greater concentrations of organic matter did not reduce removal rates for some compounds, indicating that their mere presence does not account for full competitive adsorption. Similarly, Ziska et al. (2016) studied the removal of 14 pharmaceuticals from three different wastewater types using four different PACs and observed that natural organic matter had minimal effects on removal.

5 Mechanisms, competitive adsorption and desorption

The following sections will address the various mechanisms proposed for the adsorption of pharmaceuticals on AC surfaces, the competitive adsorption

phenomena in multiple pharmaceutical aqueous systems and the various methods used for the desorption of the adsorbent and the regeneration of the activated carbon

5.1 Adsorption mechanism

The adsorption mechanism depends on the properties of the AC and on the nature of the pharmaceutical compound (Delgado et al. 2014) amongst other factors. It has been postulated that the adsorption mechanism consists of four major steps: bulk transport in the liquid phase, film transport across the boundary layer, intra-particle transport, and adsorption onto the active surface sites (physical or chemical in nature); the second step is often assumed to be the rate-controlling step (Ahmed et al. 2015; Onal et al. 2007). The rate determining step is dictated by the molecular diffusion through the boundary layer and defines the adsorption capacity of the AC and the accessibility of the pharmaceutical molecules to the inner surface (Ahmed et al. 2015). Initial adsorption increases rapidly, consistent with the theory that vacant surface sites are widely available; however, with time, as the system equilibrates, it becomes difficult to occupy the sites as a result of repulsive forces between molecules in the bulk and the adsorbent (Fernandez et al. 2015). However, this conclusion is not always applicable as there is a growing number of publications that have noted that the rate controlling step differs from one system to another and will depend on several forces and the nature of these forces, which will then determine whether the adsorption process is classified as physical (physisorption), interactive adsorption or chemical (chemisorption) or a combination of two or more.

The rate controlling step can be determined by evaluation of time dependency and is often related to the kinetics of adsorption. If the first order kinetic constant is found to be dependent on time, the controlling step of the adsorption rate is assumed to be of the intraparticle diffusion type (Weber and Morris 1963) and the diffusion model is used to explain the kinetics. On the other hand, if the constant is independent of time, surface reaction is assumed to control adsorption. The pore volume diffusion model is based on the notion that intraparticle diffusion only comes from pore volume diffusion and this is due to the concentration gradient in the liquid phase. The

geometry of the pores thus affects molecular diffusion. The intraparticle diffusion model consists of external mass transfer (diffusion of the adsorbate to the boundary layer) followed by the diffusion and adsorption of the adsorbate through the pores of the AC until equilibrium is reached (Ruiz et al. 2010). Compliance with the pseudo second order kinetic model indicates that the rate-limiting step involves sharing/exchange of electrons between the pharmaceutical and the carbon. Increased adsorption with higher temperature arises from the reduced boundary layer thickness and de-solvation of the adsorbing species (El-Shafey et al. 2014).

Physical adsorption occurs mostly under the influence of the following forces: Van der Waals force, hydrophobicity, hydrogen bonds, polarity, steric interaction, dipole–dipole interaction, pi–pi interaction, or a combination of these forces (Cai and Larese-Casanova 2014; Zhou et al. 2015). When adsorption takes place chemically, the adsorbent and adsorbate share electrons, thus forming a chemical bond (Zhou et al. 2015). Surface functional groups and their charges also affect adsorption via wettability, colloid stability, the promotion/repulsion of electrostatic interactions, and alteration of electron donating nature (Cai and Larese-Casanova 2014). However, chemical sorption depends on other system characteristics. For example, the sorption mechanism consisting of hydrogen bonding via dipole–dipole interactions between oxygen moieties on the pharmaceuticals and hydrogen atoms on the carbon is highly dependent on solution pH (Wong et al. 2016). In certain situations, it was found that low range temperatures functionalities (hydroxyl and carboxylic groups) control the adsorption mechanism (Álvarez Torrellas et al. 2015). In the absence of special interactions at the adsorbent-adsorbate interface, adsorption is mainly dictated by surface area (Bhadra et al. 2016). The adsorption mechanism is a result of electrostatic and non-electrostatic interactions that depend on adsorbate, adsorbent, and solution properties. Additionally, while the carbon micropores promote adsorption of adsorbates fitting the pore size, carbons with large pores and a large amount of oxygen containing functional groups can also remove pharmaceuticals via reactive adsorption (Nielsen et al. 2014).

It is worth noting that process thermodynamics also plays a role in specifying the nature of adsorption where it has been reported that when the change in the

free energy, ΔG , value is in the range of 0 to -20 kJ/mol then physisorption is assumed to predominate while values in the range of -80 to -300 kJ/mol reflect chemisorption (Wong et al. 2016). Similarly, the change in enthalpy, ΔH , values can also be used to identify the type of adsorption taking place, such that ΔH values within the range of 2.1–20.9 kJ/mol are indicative of physical adsorption (Ahmed and Theydan 2012). Endothermic processes favor higher temperatures that enhance the diffusion rate across the external boundary layer and within the pores of the adsorbent (Chayid and Ahmed 2015). Negative changes in entropy suggest a decrease in randomness at the sorbate-solution interface. The type of change in entropy, positive or negative, also depends on the adsorbent type (Ahmed and Theydan 2012).

The exact/specific mechanisms of adsorption are not fully understood (Quesada-Peñate et al. 2009). Numerous studies have observed that it is a complex interplay between several forces and rate controlling steps, such as electrostatic, chemical, physical, non-electrostatic interactions and diffusion through liquid and solid boundaries all of which depend on adsorbent and adsorbate properties, and thus differ from system to system. Although considerable understanding of the dominant adsorption mechanisms have been gleaned from current studies, further mechanistic studies (whether theoretical or experimental) are necessary because the currently available studies are limited to a small number of pharmaceutical compounds.

5.2 Competitive adsorption: multiple pharmaceutical systems

The vast majority of studies have examined adsorption in single-solute systems, i.e. the adsorption of one pharmaceutical at a time. Competitive adsorption, between different pharmaceutical compounds, has received much less attention despite the fact that treated wastewater effluents contain a “cocktail” of multiple types of pharmaceuticals that are often recalcitrant to conventional wastewater treatment methods.

Table 5 summarizes the laboratory-scale studies that have examined competitive adsorption. The presence of multiple components in water and wastewater streams alters the single solute-adsorbent interactions that would otherwise occur and thus the

adsorbed amounts differ indicating the action of a competitive mechanism (Ogata et al. 2012). The presence of more than one component affects the levels of the energy field in the active sites, causing competition between solutes (Sotelo et al. 2014). The physico-chemical properties of the carbon and pharmaceutical both play a role in determining which of the compounds is more efficiently adsorbed. Generally, compounds with a higher octanol–water partition coefficient have a higher chemical affinity for the AC and appear to be favored under competitive adsorption conditions (Chang et al. 2015; Sotelo et al. 2014). In addition, compounds with high water solubility and low molecular weight tend to have lower removal rates. Furthermore, the polarity of the compounds plays a role as AC favors the adsorption of non-polar compounds (Masson et al. 2016; Sotelo et al. 2014). The size of the compounds and the pore size distribution of the carbon play a role in the general adsorption process, and thus contribute to the competitive effect, whereby adsorption potential onto microporous carbon increases with smaller pore size because molecules are closer to the AC surface (Chang et al. 2015; Sotelo et al. 2014). Thus, in a binary system, for two adsorbates with different volumes, a sieving effect is observed such that the adsorbate with the smaller molecules accesses the AC pores in greater quantities, resulting in greater removal rates for the co-adsorbate with the smaller volumes; this effect is more prominent in ACs that have narrow pore size distributions. Overall, diffusion into the porous network depends on molecular volume of the adsorbate, while the diffusion time depends on the affinity of the adsorbate towards the carbon (Masson et al. 2016).

Ruhl et al. (2014) studied adsorption onto different kinds of PAC in water that contains multiple pharmaceutical compounds; it was observed that the percent decrease in removal per pharmaceutical varied with the type of carbon and the compound being considered. This observation was also surmised by Mansouri et al. (2015) who reported that the affinity of individual compounds for the AC was paramount in the competitive adsorption in a binary system. Chang et al. (2015) studied adsorption in single, binary, and tertiary systems. In binary systems, adsorption replacement took place, where the compound with the larger K_{ow} replaced the compound with the smaller K_{ow} value; smaller-sized compounds, however, were able to occupy micropores. In a tertiary system,

Table 5 Summary of competitive adsorption studies

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	kinetic model
Ahmed et al. (2017a)	Water	Functionalized biochar from bamboo biomass	0.5–1.12	Sulfathiazole	0.33–50		30.18–237.71	1.5–10.0	294–303	Langmuir and Freundlich (single), Langmuir (competitive)	Pseudo-second (single), intra-particle diffusion (competitive)
Ahmed et al. (2017b)	Deionized	Functionalized biochar from eucalyptus wood	–	Sulfamethoxazole Sulfamethazine Sulfamethazine	0.25–20		23.76–88.10 17.71–88.10 20.71	1.5–10.9	298	Langmuir, Freundlich	Pseudo-second
Álvarez-Torrellas et al. (2017)	Lake water Synthetic wastewater	AC from rice husk and peach stones	1102–1521	Sulfamethoxazole Sulfathiazole Chloramphenicol Carbamazepine	100		28.29 45.2 21.35 26.0–310.2	6.5	303	Sips, Guggenheim–Anderson–De Boer	
Calisto et al. (2017)	Ultrapure	PAC (commercial)	414–848	Ciprofloxacin Carbamazepine	100–500		19.7–101.6 75.13–144.12		298	Langmuir	Pseudo-second
Chang et al. (2015)	Deionized	Granular AC from bituminous coal	1156	Paroxetine Oxazepam Acetaminophen	5.0–40		43.14–89.57 62.22–116.12 83.4–453.37	–	298	Langmuir	Pseudo-second
Jung et al. (2013)	Deionized	Powdered AC (commercial)	972.3	Diclofenac Sulfamethoxazole 17 α -ethynylestradiol Sulfamethoxazole Carbamazepine Diclofenac Ibuprofen	25.32–126.65 23.62–118.13 29.61–148.07 20.63–103.14		266.53–379.07 126.65–445.80 45.0–50.0	3.5–10.5	293	Freundlich	Pseudo-second

Table 5 continued

Study	Water type	AC (precursor)	BET (m ² /g)	Pharmaceuticals	Initial conc (mg/L)	Removal (%)	Adsorption capacity (mg/g)	pH	Temperature (K)	Isotherm	kinetic model	
Jung et al. (2015)	Distilled	Activated biochar from loblolly pine chips	1120	Diclofenac	5.9	–	109–372	7.0	–	Langmuir	–	
Mansouri et al. (2015)	Ultrapure	AC from olive stones	903–1106	Naproxen	4.6	–	52.6–290	–	–	–	–	
				Ibuprofen	4.1	–	161–311	–	–	–	–	–
				Ibuprofen	100	–	144.40–282.62	4.3	298	Langmuir	Pseudo-second	
Masson et al. (2016)	Ultrapure	AC cloth	1050–1690	Amoxicillin	–	–	120.58–204.62	–	–	–	–	
				Ibuprofen	0.83–2.06	–	127.9–288.81	7.5	298	Langmuir-	Pseudo-second	
Oh et al. (2013)	Pure	Granular AC (commercial)	1120	Carbamazepine	0.95–2.36	–	9.83–437.1	–	–	–	Freundlich	–
				Ofloxacin	1.45–3.61	–	39.87–216.82	–	–	–	–	–
				Diclofenac	1.18–2.96	–	5.86–411.65	–	–	–	–	–
				Clofibric acid	0.04–1	–	0.2–046	7	293	Langmuir	–	
Ruhl et al. (2014)	Ultrapure	Powdered AC (commercial)	900–1300	Propyphenazone	–	–	0.36–3.3	–	–	–	–	
				Acesulfame	2	7.1–29.1	–	7.8	293	–	–	
	Wastewater			Diclofenac		77.4–96.9						

competition adsorption was greatly affected by the macropores. Masson et al. (2016) studied the adsorption of nine contaminants in single, binary, and complete mixtures. Adsorption mainly takes place in micropores, particularly in the case of small molecules, which more easily diffuse in narrow pores. In binary mixtures, competition and sieving effects are observed. Chang et al. (2015) also found that some systems fall between noncompetitive and competitive adsorption, as is the case of acetaminophen-sulfamethoxazole system, where adsorption density is underestimated by the former and overestimated by the latter.

It is evident that the study of competitive adsorption of pharmaceuticals by AC in multi-solute systems is extremely challenging as the possible combinations are endless. However, more work is certainly required in order to understand the mechanisms involved in competitive adsorption. This may involve examining classes of pharmaceuticals individually, or in combination with other classes in mixtures containing one representative pharmaceutical from each class and then a combination of two more from each class and so on. Furthermore, additional studies should be done to determine the underlying mechanisms that account for the reduction/improvement of adsorption and whether these forces are dependent on the particular system characteristics and/or pharmaceutical combination.

5.3 Desorption and regeneration

One of the major issues associated with the use of activated carbon for the adsorption of conventional and emerging contaminants is the cost associated with the regeneration and disposal of the saturated and exhausted activated carbon (Cabrita et al. 2010; Yuen and Hameed 2009). Several methods have been proposed for the regeneration of exhausted activated carbon. These regeneration methods are based on thermal (steam, carbon dioxide, hot water, inert gas, pyrolysis), chemical (catalytic oxidation, solvents, pH swing), biological/microbiological, electrochemical, ultra-sound or microwave processes (Ahmed et al. 2015; Aktas and Cecen 2007; Ania et al. 2007; Yuen and Hameed 2009; Zanella et al. 2014; Zhou et al. 2015). Some of these regeneration methods, such as microwave-assisted processes, remain in the development phase (Yuen and Hameed 2009), while others, such as the thermal methods, have been used on an

industrial scale, albeit not specifically for pharmaceutically-saturated activated carbons (Zanella et al. 2014). A detailed description of the advantages and disadvantages of these methods is beyond the scope of this work and the interested reader is referred to a number of excellent review papers such as those by Aktas and Cecen (2007); Yuen and Hameed (2009); Zanella et al. (2014). In this section studies investigating the regeneration of activated carbon saturated with pharmaceuticals will be addressed and the emphasis will be on the regeneration efficiency, number of cycles, method of regeneration, and where applicable the chemical/solvent used for regeneration.

Huang et al. (2014) studied desorption with hydrochloric acid and sodium hydroxide and found the latter to perform better, suggesting that the process took place mainly through strong chemisorption bonds. Reza et al. (2014) used different eluents (water, hydrochloric acid, sulfuric acid, acetic acid, methanol and ethanol) to determine the efficacy of desorption of ibuprofen and clofibrac acid; methanol desorption, performed at room temperature, proved to be the most efficient. Essandoh et al. (2015) studied the sorption of the pharmaceuticals from pine wood biochar via methanol stripping, achieving the ability to recover 93% (salicylic acid) and 88% (ibuprofen) of the adsorbed concentrations. The regenerated biochar was then reused in four cycles and retained 76 and 72% of the initial adsorption capacities of salicylic acid and ibuprofen. Kyzas and Deliyanni (2015) reported that hydrothermally treated carbon loses less adsorption capacity after reuse (11%) compared pyrolyzed carbon (35% less), after 20 reuse cycles. Bhadra et al. (2016) successfully recycled oxidized AC using methanol solvent washing and was able to use it for 5 cycles; adsorption capacity decreased from the first two runs, and steadied at the third.

Álvarez-Torrellas et al. (2016) used a sodium hydroxide solution for the regeneration of several types of activated carbon saturated, separately, with ibuprofen and tetracycline. For ibuprofen, desorption efficiencies were in the range of 42.2–70.4%. On the other hand, for tetracycline, the desorption efficiencies were lower and were in the range of 18.3–41.7%. For both pharmaceuticals, only a single subsequent adsorption cycle was performed after regeneration. Cai and Larese-Casanova (2014) used ethanol to promote carbamazepine desorption from activated carbon where a recovery of 93% was achieved,

indicating that carbamazepine molecules were not chemically transformed and were likely to have been only bound through physisorption. El-Shafey et al. (2012) used hydrochloric acid for the regeneration of activated carbon saturated with ciprofloxacin where the desorption efficiency reached 83%.

Liu et al. (2011) used several chemicals (sodium chloride, magnesium chloride, methanol and sodium hydroxide) to investigate norfloxacin desorption from a number of commercial and lotus-stalk derived activated carbons. Desorption by sodium hydroxide proved to be the most effective. Saygili and Guzel (2016) conducted regeneration studies using sodium hydroxide solution to test the reusability of tomato waste derived activated carbon saturated with the antibiotic tetracycline. The adsorption–desorption cycle was repeated five times and the results showed that the adsorption capacity of the AC decreased for each new cycle after desorption and by the end of the fifth cycle it dropped to 45% of the original adsorption capacity. Effectiveness of desorption also differs with the pharmaceutical being removed; Reza et al. (2014) reported that desorption of ibuprofen via methanol was very effective (96%) and allowed for adsorbent reusability; on the other hand, clofibric acid desorption with methanol was only 60%. Similarly, regeneration differences arise with the use of different adsorbents. For example, Wong et al. (2016) found that the AC they used can be regenerated thermally at relatively low temperatures and reused with sustainable adsorption rates and capacities, as compared to palm based PAC, which was largely exhausted.

There is limited information concerning alternative low cost and effective regeneration techniques (Ahmed et al. 2015). Addressing this need is crucial if AC adsorption is to become a widespread treatment alternative, especially where large plant scale treatment is concerned, as cost-effectiveness and reusability are of particular importance. In addition, there are a number of areas that appear to have received scant attention in the literature; these include the regeneration of ACs that have been used for the removal of several pharmaceuticals simultaneously, comparison of the efficiencies of several regeneration methods for a single, or a group of pharmaceuticals, and the effectiveness of one regeneration method over a wide spectrum of individual or a group of pharmaceutical compounds.

6 Final considerations and further needs

This review presented the current status of AC adsorption in the realm of pharmaceutical removal from aqueous solutions (water/wastewater) and shed light on a number of gaps that require further experimental and theoretical study. Whilst the current studies have elucidated several aspects of the use of AC for the removal of pharmaceuticals, such as the kinetics and thermodynamics of the process and the effects of operating conditions, adsorbent and adsorbate properties on the adsorption process, there are several areas that require further investigation. These include: (a) more pharmaceuticals need to be investigated (currently there is a small subset of pharmaceuticals that have been investigated); (b) the impact of a mixture of pharmaceuticals on the adsorption process; (c) the mechanisms involved in competitive adsorption; (d) experimental conditions which mimic industrial and environmental conditions; (e) the effects of the water matrix (nature of water, ionic strength, presence of natural organic material), parameters as well as physical system (adsorbent type) characteristics on the process (f) cost effectiveness of AC adsorption, taking into account regeneration; (g) novel and cost effective regeneration methods; (h) AC in combination with other advanced removal technologies; (i) fixed bed, pilot and industrial scale investigation. What follows is a summary of the salient points of this review along with some recommendations for future research.

The maximum and minimum mass (in mg) of pharmaceuticals removed from aqueous solutions per mass of AC adsorbent used (in g) for 66 pharmaceuticals are shown in Fig. 1. It is noted that these values are obtained from all the studies included in the literature review and summarized in Tables 3, 4, and 5 and that the individual references for each of the maximum and minimum values for all 66 pharmaceuticals are included in the supplementary materials Table S1. An examination of the figure reveals a wide discrepancy between values from different studies for the same pharmaceutical, highlighting the need for an established baseline or systematic means of experimentation that will yield values that can be compared on a more rigorous basis. Furthermore, and overwhelmingly so, the most commonly studied pharmaceuticals belong to the anti-infectives and antibiotic class (J class in accordance with ATC classification),

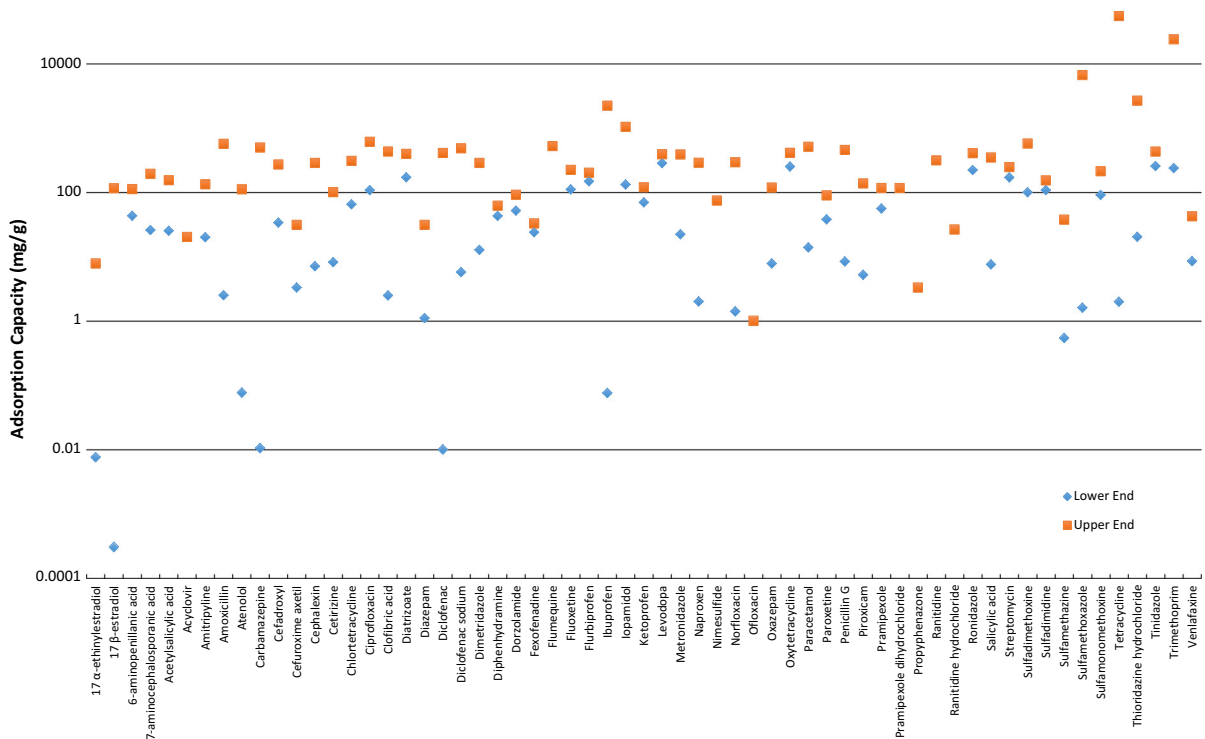


Fig. 1 Maximum and minimum adsorption capacity of various types of activated carbons for the removal of 66 pharmaceuticals (in mass of pharmaceutical per mass of AC used)

followed by pharmaceuticals belonging to the nervous system class (N class). Additionally, it is observed that a subset of these pharmaceuticals displayed greater removal rates than others. This subset is comprised of pharmaceuticals that can be removed at values greater than 100 mg/g on the lower end or a value greater than 300 mg/g on the higher end (these cutoffs were derived from perusing the values and determining a proper cutoff point with respect to the range of values); the majority of the pharmaceuticals in this subset belong to the anti-infectives and antibiotics class. However, these conclusions cannot be considered comprehensive because almost half of the original set of pharmaceuticals belongs to the same class, and so it is expected that they appear more so than the others. It is worth noting that the 66 pharmaceuticals represent a reasonable percentage of the number of pharmaceuticals that have been detected in the environment [203 pharmaceuticals according to Hughes et al. (2013)] but is a very small subset of the total number of pharmaceuticals that are currently in use [over 4000 according to Hughes et al. (2013)]. It is therefore evident that further adsorption studies which include a

larger number of pharmaceuticals belonging to a wider span of therapeutic classes are needed.

From the results reported in Tables 3, 4 and 5, it is evident that the vast majority of laboratory based adsorption studies were conducted using concentrations of pharmaceuticals in the mg/L range. Pharmaceuticals in the environment occur at the $\mu\text{g/L}$ and ng/L range and therefore, it is recommended that more laboratory scale studies are conducted using these concentration levels. In addition, studies examining adsorption in systems that contain several pharmaceuticals, often referred to as competitive absorption, are also limited. However, competitive adsorption arises not only from the presence of other pharmaceuticals, but also from other pollutants (such as dyes and heavy metals) and organic material already present in the water. Wastewater effluent and surface water contain a “cocktail” of pharmaceuticals, NOM as well as a variety of other organic and inorganic contaminants and, if AC adsorption is to be used effectively, experimental studies, on the lab or pilot scale, must reflect these operating conditions. This emphasizes the need for research addressing both

competitive adsorption and the use of real wastewater, as opposed to simulated wastewater or distilled water. Furthermore, and as can be gleaned from the various studies considered, there are a very limited number of studies that have dealt with the use of PAC (and GAC) for the removal of emerging contaminants from operating wastewater and water treatment plants. It is therefore, recommended that on-site adsorption studies should be conducted (and reported in the open literature) and that the scalability of both commercial and waste/biomass based AC should be investigated further.

There is great ambiguity and contradictory results in terms of the impacts that interactions between different operating parameters have on the adsorption process, aside from the interactions between adsorbent and adsorbate properties, and the overall interactions between these two categories. These somewhat uncertain relationships, make it difficult to fully understand, and consequently design and scale-up adsorption processes in an efficient and economic manner. Further studies are needed at both the laboratory (especially column studies) and pilot scale to: (1) understand the interplay between parameters on a general level, (2) investigate how these interactions differ from pharmaceutical to pharmaceutical, and (3) determine how a mixture of pharmaceuticals alters expected behavior and the specific parameters that cause such alterations.

Information on the costs of AC systems and how that changes for different feed waters and environments is highly lacking in the literature. Bases for comparison with other processes are very limited, and as such, information that is very important when considering potential AC applications is unavailable. In particular, the regeneration of AC has not been extensively investigated, especially for ACs generated from agricultural wastes. Effective regeneration of the spent AC is crucial for the sustainability of the process as disposal of the waste AC material and the use of “fresh” AC pose environmental and economic constraints that may hinder the widespread application of adsorption by AC. As such, there is a need for full characterization of regenerated ACs to be compared with fresh samples; this will determine the effectiveness of the regeneration method implemented and its viability cost-wise for efficiency purposes. In addition, there is very little, if any, information on the relation between nature and mechanisms of adsorption and the

regeneration methods used thereafter; studies may be performed to determine the choice of regeneration method based on the nature of adsorption (chemical or physical); in this way, the nature of the process itself can be used to facilitate regeneration in a manner that maximizes reuse and minimizes cost. Finally, and is evident from the discussion above, there is a need for in-depth cost–benefit analysis of the various types of adsorbents, accounting for the different methods of production, operation and regeneration.

Acknowledgements The authors acknowledge the financial support of the University Research Board (URB) at the American University of Beirut.

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