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A. Tahar, J.M. Choubert, Marina Coquery

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XENOBIOTICS REMOVAL BY ADSORPTION IN THE CONTEXT OF TERTIARY TREATMENT- A MINI REVIEW

Alexandre TAHAR, Jean-Marc CHOUBERT*, Marina COQUERY

Irstea, UR MALY (Freshwater systems, ecology and pollution research unit)

5 rue de la DOUA, CS70077, 69626 VILLEURBANNE Cedex, France

{alexandre.tahar@irstea.fr , jean-marc.choubert@irstea.fr, marina.coquery@irstea.fr }

Tel: +33 (0)4 72 20 87 87, Fax: +33 (0)4 78 47 78 75.

* corresponding author

KEYWORDS

Langmuir parameters, pharmaceuticals, pesticides, batch studies, maximum adsorption capacity, wastewater treatment

ABSTRACT

Many xenobiotics, including several pharmaceuticals and pesticides, are poorly treated in domestic wastewater treatment plants. Adsorption processes, such as with activated carbons, could be a solution to curb their discharge into the aquatic environment. As adsorbent-like activated carbon is known to be expensive, identifying promising alternative adsorbent materials is a key challenge for efficient yet affordable xenobiotic removal from wastewaters. As part of the effort to address this challenge, we surveyed the literature on pharmaceutical and pesticide xenobiotics and built a database compiling data from 38 scientific publications covering 65 xenobiotics and 58 materials. Special focus was given to the relevance and comparability of the data, to the characteristics of the adsorbent materials used, and to the operating conditions of the batch-tests inventoried.

This paper gives an in-depth overview of the adsorption capacities of various adsorbents. The little data on alternative adsorbent materials, especially for the adsorption of pharmaceuticals, makes it difficult to single out any one activated carbon alternative capable of adsorbing pesticides and pharmaceuticals at the tertiary stage of treatment. There is a pressing need for further lab-scale experiments to investigate the tertiary treatment of discharged effluents. We conclude with recommendations on how future data should best be used and interpreted.

1. INTRODUCTION

Research into the issue of environmental xenobiotics has recently surged. Many xenobiotics, like pharmaceutically-active compounds (PhACs) and pesticides, have been quantified in surface waters (Zuccato et al. 2000; Kolpin et al. 2002), ground waters (Ternes 2001; Heberer 2002) and drinking water (Jones et al. 2005; Togola and Budzinski 2008). To reduce xenobiotic release into the aquatic environment, EU regulations have promoted a phased reduction of xenobiotics discharge based on a regularly revised list of priority substances including pesticides and certain pharmaceuticals.

Conventional wastewater treatment plants (WWTPs) have been singled out as one of the main point-sources of xenobiotic transfer into the aquatic environment (Bendz et al. 2005; Carballa et al. 2004; Carballa et al. 2005; Lishman et al. 2006; Vieno et al. 2006; Palmer et al. 2008; Spongberg and Witter 2008). Indeed, treated effluents released by conventional WWTPs still contain significant concentrations of several xenobiotics, due to their poor biodegradation in secondary treatments and/or their high levels in raw influents. Pesticides and PhACs are of particular concern, since many of these substances are poorly biodegradable and highly hydrophilic. For instance, there are regular reports of effluent concentrations over $0.1 \mu\text{g L}^{-1}$ for several pesticides (diuron, glyphosate, AMPA) and PhACs (e.g. sulfamethoxazol, roxithromycin, ibuprofen, carbamazepine, propranolol, acebutolol, atenolol, sotalol, paracetamol, ketoprofen, gemfibrozil, diclofenac (Ternes et al. 2004; Gabet-Giraud et al. 2010; Martin Ruel et al. 2010; Falås et al. 2012); and even reports of effluent concentrations over $1 \mu\text{g L}^{-1}$ for certain xenobiotics, including aminomethylphosphonic acid (AMPA), ibuprofen and gemfibrozil (Martin Ruel et al. 2010).

Advanced processes, such as UV or ozone oxidation, nanofiltration (NF) and reverse osmosis (RO) have been shown to improve treatment for a majority of xenobiotics, with removal efficiencies of over 90% for many xenobiotics poorly eliminated in conventional WWTPs (Verlicchi et al. 2010; Martin Ruel et al. 2011). However, these processes are not “eco-friendly” (Wenzel et al. 2008; Høiby et al. 2008) and cannot be made sustainable until we address a number of issues such as the treatment of concentrate by RO/NF processes or the formation of toxic metabolites during ozone oxidation (Verlicchi et al. 2010). Moreover, for economic reasons, these advanced processes are not yet viably adaptable to WWTPs below 50,000 population equivalents (PE), meaning alternative solutions need to be developed.

Activated carbons (AC) provide efficient retention of xenobiotics due to their microporous structure that offers very high specific areas of up to $2000 \text{ m}^2 \text{ g}^{-1}$. Filtration on granular AC (GAC) enabled removal efficiencies of up to 90% for a majority of xenobiotics, including carbamazepine and diclofenac (e.g. (Westerhoff et al. 2005; Martin Ruel et al. 2011)). High removal efficiencies were also obtained by adding powder-activated carbon (PAC) into a membrane bioreactor, with removal efficiencies above 90% for atrazine, naproxen and estrone (Snyder et al. 2007). Nevertheless, the use of ACs at the tertiary treatment stage runs into several issues, notably their price and the environmental cost of the production/regeneration step (Crisafulli et al. 2008; Verlicchi et al. 2010). Identifying alternative materials to AC is therefore a key issue for tertiary treatments to remove xenobiotics from WWTP effluent.

The last decade has seen a surge in papers on the adsorption capacities of chemical substances by various adsorbent materials (batch assay tests). Much of this literature is devoted to dyes (Crini 2006; Gupta and Suhas 2009), phenols (Ahmaruzzaman 2008) and heavy metals (Bailey et al. 1999; Babel and Kurniawan 2003). Besides ACs, reviews have also focused on

wastes from industry or agriculture (Bhatnagar and Sillanpää 2010), chitosan and chitin derivatives (Bhatnagar and Sillanpää 2009), natural zeolites (Wang and Peng 2010), sawdust (Shukla et al. 2002), biosorbents such as dry activated sludge and yeast (Aksu 2005), polymeric materials (Pan et al. 2009) and cellulose-based materials (Wojnárovits et al. 2010), but unfortunately without reporting any data on pesticide and PhAC adsorption capacities. The relevant knowledge on PhACs and pesticides is spread across papers dealing with individual results from batch assays (i.e. one adsorbent in contact with one xenobiotic).

This paper set out to draw up a qualitative and quantitative overview compiling the available literature data describing the pesticide and PhAC adsorption potential of ACs, mineral adsorbents and other adsorbent materials. We set out with three aims: i) to identify the most promising AC alternatives capable of adsorbing hydrophilic xenobiotics; ii) to identify relationships between adsorption parameters based on material characteristics so as to predict adsorption potential, and iii) to recommend technical and research directions for refining and improving the data used. We thus built a database by compiling the results of experimental adsorption assays (Langmuir parameter values) found in the literature, including meta-data such as the physical-chemical characteristics of the adsorbents and the experimental conditions applied. Special attention was devoted to selecting the data and compiling reliable results relevant to adsorption potentiality.

2. MATERIALS AND METHODS

2.1. Construction of the database

We built a database compiling published Langmuir parameter data on various adsorbents tested with aqueous solutions at lab-scale in completely mixed-batch reactors (Homem and Santos 2011). The Langmuir model (equation 1) considers a maximum adsorption capacity

(‘MAC’, in mg g⁻¹) and an affinity constant (‘b’, in L/mg) to model the equilibrium values between the xenobiotic concentrations in an adsorbent material and the liquid phase:

$$Q_e = \frac{MAC \cdot b \cdot C_e}{1 + b \cdot C_e} \quad \text{Eq. (1)}$$

The Langmuir parameters are always expressed in the same dimension, contrary to Freundlich parameters (Kf and n) for which Kf unit depends on n value (Xu et al. 2009). Hence, the use of Langmuir parameters enabled us to compare MAC and b results between different studies.

We then imported physical-chemical characteristics of studied substances, such as log K_{ow}, solubility, pKa, Henry’s constant, and molecular weight. These values were extracted from <http://www.syrres.com> and <http://toxnet.nlm.nih.gov> (see Online Resource 1). We also collected physical characteristics such as specific area, pore volume and diameter, particle size, density, point of zero charge ‘pH_{pzc}’, plus chemical characteristics, and price. Crucially, we documented the conditions under which the Langmuir parameters were determined. These metadata describe the operating conditions of the batch experiments: initial concentration (C₀), water type (synthetic, surface, groundwater, release from a secondary WWTP), pH, temperature, solid/liquid ratio, agitation speed, time to reach equilibrium conditions (see Online Resource 2 to see metadata on the experimental conditions applied).

2.2. Data selection strategy

We developed a strategy to exploit only selected results on Langmuir parameters values. This data selection strategy was implemented as follows:

- adsorbent materials produced from certain kinds of waste such as dried sludge (Aksu and Yener 1998) or plant material like sunflower stem waste (Sun and Xu 1997) or garlic peel (Hameed and Ahmad 2009) were excluded as they are not mechanically resistant in water and may consequently release undesirable xenobiotics into the aquatic environment;

- we chose physical-chemical conditions within the usual range characteristic of domestic wastewater, i.e. pH in the range 5 to 9 and temperature in the range 5°C to 30°C;
- we chose data on adsorption with the more frequent type and generated with spiked synthetic water for homogeneity reasons due to the lack of data on natural waters (such as rivers and groundwaters);
- we excluded results when time-to-equilibrium was longer than 24 h, as this duration would not be relevant for a tertiary treatment application;
- experiments carried out in completely mixed reactors (batch) were ignored if carried out under conditions far removed from conventional secondary effluents treatment.

Consequently, the selected dataset thus selected originates from batch studies considering the adsorption of only one substance in synthetic water free of organic matter. We found no papers reporting adsorption performances for a cocktail of xenobiotics.

The database ultimately obtained contains data extracted from 38 research papers published between 1998 and 2010 (see Online Resource 3 for the full reference list). It gathers 108 individual data on MAC values (but only 43 corresponding b parameter values) concerning 65 xenobiotics (pesticides and pharmaceuticals) and 58 adsorbent materials.

3. RESULTS

3.1. Description of the xenobiotics and the adsorbent materials in the database

3.1.1. Xenobiotics studied

Among the data gathered in our database, pesticides were the most studied substances, with 63 data items followed by PhACs (45 data items), as shown in Figure 1.

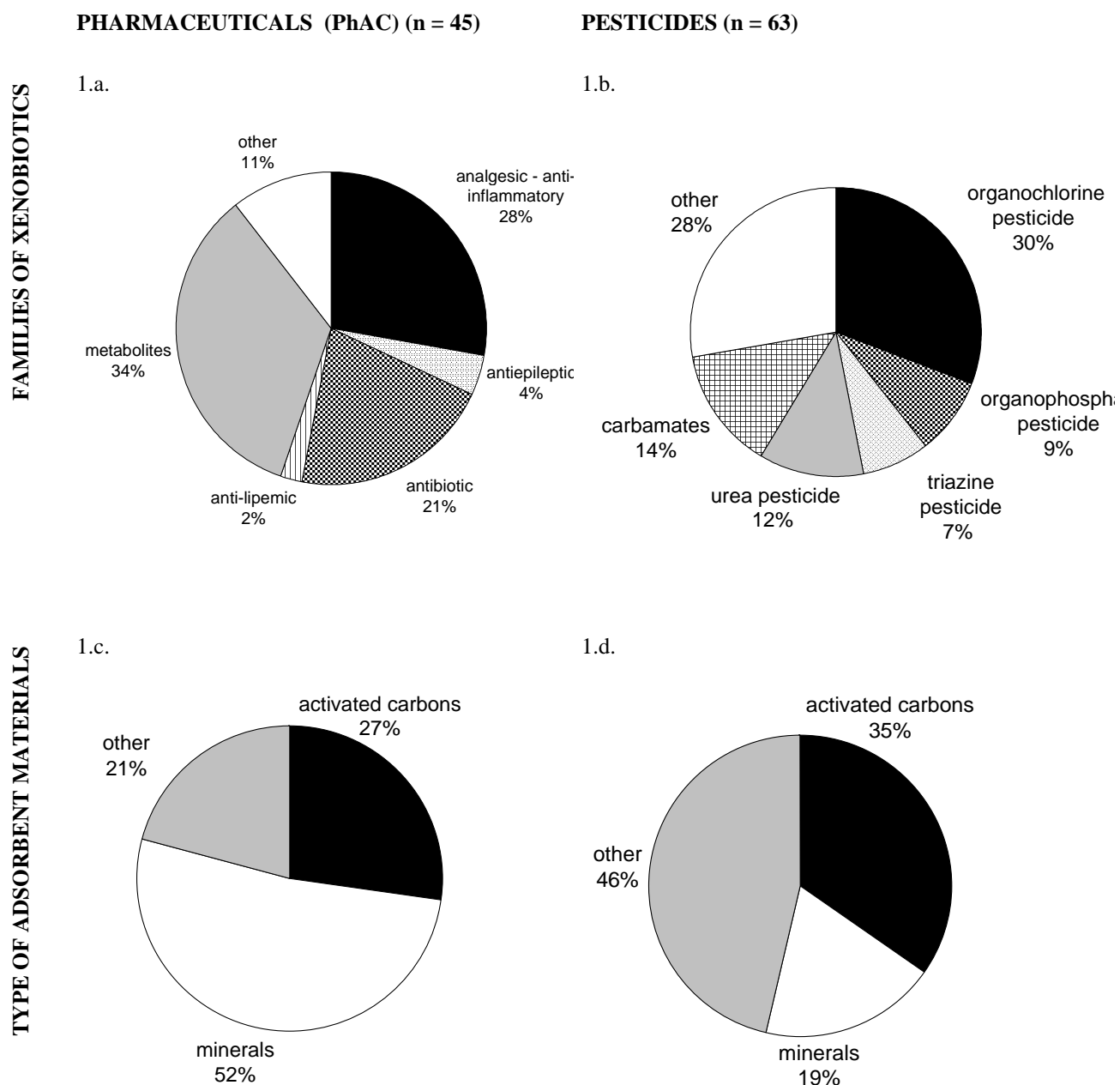


Fig 1 Distribution of MAC values in the database by xenobiotic family: a) pharmaceuticals, b) pesticides; and by type of adsorbent material - activated carbons (ACs), mineral adsorbent and other adsorbent materials- for c) pharmaceuticals and d) pesticides. (n: number of available MAC data).

The PhACs most represented were metabolites (salicylic acid and tetracycline hydrochloride; Figure 1.a), accounting for 34% of the 45 PhAC data items, followed by analgesics/antiinflammatories (ibuprofen, ketoprofen, diclofenac) and antibiotics (amoxicillin, ciprofloxacin) which represented 28% and 21% of the pharmaceuticals, respectively. The

PhACs most rarely studied for adsorption were anti-lipemic (clofibric acid) and antiepileptic (carbamazepine) substances that represented 2% and 4% of the PhAC data items, respectively.

The pesticides most represented were organochlorines (lindane, alachlor, acetochlor; Figure 1.b), accounting for 30% of the 65 pesticide data items, followed by carbamates and urea (diuron, isoproturon) which represented 14% and 12% of the pesticide data items, respectively. The pesticides least represented were organophosphates (diazinon, methylparathion) and triazines (atrazine, simazine) that represented 9% and 7% of the pesticide data items, respectively.

The literature only featured adsorption performances for a few of the xenobiotics usually measured in domestic treatment plants effluents (such as sulfamethoxazole, carbamazepine, diclofenac, isoproturon and diuron; (Ayranci and Hoda 2005; Polubesova et al. 2006; Sarkar et al. 2007; Bui and Choi 2009; Vergili and Barlas 2009)). Unfortunately, each data was measured on different adsorbent materials, making it difficult for us to use them.

3.1.2. Adsorbent materials studied

The available MAC data for ACs, mineral and other adsorbent materials are presented for PhACs (Figure 1.c) and pesticides (Figure 1.d). Complementary metadata on the description of adsorbent materials studied are detailed in Online Resource 4.

For PhACs, ACs represented 27% of the 45 data items. Alternative adsorbent materials were extensively studied (73% of the data: 52% on mineral materials and 21% on other adsorbent materials). For pesticides, ACs represented 35% of the 63 data items. Alternative adsorbents were also widely studied (65% of the data on pesticides: just 19% on mineral materials and

46% on other adsorbent materials). The ACs studied are commercial products that were manufactured from different sources such as cork (Mestre et al. 2009), coconut shell, wood (Quesada-Peñate et al. 2009), date stones (Hameed et al. 2009) or banana stalk (Salman and Hameed 2010). The mineral adsorbent materials studied were essentially composed of different classes of modified clays (e.g., (Polubesova et al. 2006; Chang et al. 2009a), one silica (Bui and Choi 2009) and one modified zeolite (Lemić et al. 2006). Clays were thermally and chemically modified to increase their porous structure and change their surface chemistry to improve adsorption affinity. Other adsorbent materials were generally organic and composed of chitosan derivatives (Adriano et al. 2005; Ding et al. 2009), polymeric materials (Otero et al. 2004; Vergili and Barlas 2009), some agricultural wastes such as rice bran (Adachi et al. 2001), or chestnut shells (Zuhra Memon et al. 2007).

3.1.3. Substances and adsorbent materials suffering from a lack of data

No Langmuir parameters were found for some substances poorly removed by conventional WWTPs, including a number of pesticides and metabolite (glyphosate, AMPA), pharmaceuticals such as the antibiotic roxithromycin and beta-blockers (atenolol, sotalol), and some personal care products such as galaxolide (Martin Ruel et al. 2010). Moreover, we found few data for some poorly-removed PhAC, including carbamazepine (two MAC data) and diclofenac (one MAC data). This relative lack of data could be due to the fact that these substances are still not regulated under environmental quality or drinking water standards.

Several adsorbent materials, even those that show adsorption potential (e.g. large specific area and pore volume), are still without Langmuir parameter data. For example, no Langmuir parameter values were found for apatite, despite the fact that it has been shown to possess a good adsorption/precipitation capacity for phosphorus (Molle et al. 2005). Chitin-based

adsorbents such as chitosan and cyclodextrin have relatively good MAC values ($> 100 \text{ mg g}^{-1}$) for substances such as heavy metals (Babel and Kurniawan 2003) and dyes (Crini 2006), but have rarely if ever been studied for PhAC and pesticides adsorption. Peat is an organic material that has good adsorption capacities ($>100 \text{ mg g}^{-1}$) for metals such as chromium (Brown et al. 2000) but has not been studied for PhAC and pesticide adsorption. Finally, no data were found on pozzolan (natural volcanic rock with a highly-developed porous network such as zeolite) or coco fiber.

3.2. Data on Langmuir parameters

The MAC and b Langmuir parameter values available for PhACs and pesticides are presented in Table 1 and classified into 3 types of adsorbent (AC, mineral adsorbents, other adsorbents).

Table 1 Langmuir parameter values (MAC and b) included in the database

Pesticides: ^aorganochlorine, ^bother, ^ctriazin, ^durea, ^eorganophosphate
 Pharmaceuticals: ^fanalgesic/anti-inflammatory, ^gother, ^hmetabolites, ⁱantibiotics, ^jantiepileptic, ^kanti-lipemic.

abbreviations		
substances	MCPA	2-methyl-4-chlorophenoxy acetic acid
	2,4-D	2,4-dichlorophenoxyacetic acid
	MCPB	4-(4-chloro-o-tolyloxy)butyric acid
materials	AC	activated carbon
	F 400	filtrasorb 400
	S 2225	Spectracarb 2225
	F 300	Filtrasorb 300
	PAC	powder activated carbon
	GAC	granular activated carbon
	BF	Blast Furnace

Activated carbons (ACs)

Material	Substance	MAC (mg/g)	b (L/mg)	Reference
F 400	salicylic acid ^h	351	1.85E-05	Otero et al. 2004
AC (norit row 0.8 SUPRA)	amoxicilin ⁱ	190	0.03	Putra et al. 2009
AC (cork powder)	ibuprofen ^f	145	0.24	
AC (cork, strong activation)	ibuprofen ^f	378	0.10	
P (AC from PET)	ibuprofen ^f	267	0.49	
AC (coal)	ibuprofen ^f	430	0.11	Mestre et al. 2009
AC (wood)	ibuprofen ^f	292	0.17	
AC (wood, strong activation)	ibuprofen ^f	149	0.06	
AC (wood)	levodopa ^g	317	0.02	
AC (coconut shell)	levodopa ^g	285	1.12	Quesada-penate et al. 2009
AC (i-ian tree bark)	levodopa ^g	393	0.88	
F 400	lindane ^a	183	-	Sotelo et al.2002
	alachlor ^b	151	-	
	amtryn ^c	355	-	
S 2225	aldicarb ^b	422	-	Ayranci and Hoda 2005
	dinoseb ^b	302	-	
	diuron ^d	213	-	
S 2225	bentazon ^b	151	0.57	Ayranci and Hoda 2004
	propanil ^b	114	0.32	
F 300	carbofuran ^b	96.2	0.13	Salman and Hameed 2010b
	2,4-D ^a	182	0.10	
	2,4-D ^a	70.0	-	
AC (WD-extra)	MCPA ^b	2.0	-	Ignatowicz 2009
	MCPB ^b	0.5	-	
PAC 313W	lindane ^a	412	1.00	Kouras et al. 1998
darco G-60	bromoxynil ^b	500	-	Yang et al. 2004
	diuron ^d	300	-	
PAC	carbofuran ^b	190	-	Fernandez-perez et al. 2005
PAC	isoproturon ^d	104	-	Sarkar et al. 2007
AC (date stones)	2,4-D ^a	238	-	Hameed et al. 2009
AC (banana stalk)	carbofuran ^b	156	0.26	Salman and Hameed 2010
AC (palm oil)	bentazon ^b	32.7	-	Salman and Hameed 2010c
AC (charcoal)	MCPB ^b	126	-	Hu et al. 1998
	imazalil ^a	208	-	
AC (ns)	2,4-D ^a	303	-	Gupta et al. 2006
	carbofuran ^b	266	-	

Table 1 (continued)

Mineral materials				
Material	Substance	MAC (mg/g)	b (L/mg)	Reference
modified clay	tetracycline ^h	5.0	-	Polubesova et al. 2006
	oxytetracycline ^h	5.0	-	
	chlortetracycline ^h	5.0	-	
	sulfamethoxazole ^h	5.0	-	
	sulfizoxazole ^h	5.0	-	
	sulfamethizole ^h	5.0	-	
mesoporous silica	carbamazepin ^j	0.2	33.8	Bui and Choi 2009
	clofibrac acid ^k	0.1	22.2	
	diclofenac ^f	0.3	2.61	
	ibuprofen ^f	0.4	13.3	
	ketoprofen ^f	0.3	14.7	
palygorskite	tetracycline ^h	56.0	0.03	Chang et al. 2009
	tetracycline ^h	99.0	0.03	
rectorite	tetracycline ^h	140	-	Chang et al. 2009b
	tetracycline ^h	107	-	
SW y-2 Na-Montmorillonite	tetracycline ^h	355	-	Chang et al. 2009c
SA z-1 Ca-Montmorillonite	tetracycline ^h	460	-	
rectorite	tetracycline ^h	140	-	
bentonite	amoxicilin ⁱ	47.4	0.01	Putra et al. 2009
goethite (syn-FeOOH)	ciprofloxacin ⁱ	49.6	0.24	Zhang and Huang 2007
goethite (ald-Fe-OOH)	ciprofloxacin ⁱ	33.1	0.01	
goethite	salicylic acid ^h	31.7	-	Xu et al. 2007
kaolinite	salicylic acid ^h	5.2	-	
rhodic ferrasol	salicylic acid ^h	17.9	-	
haplic acrisol	salicylic acid ^h	11.8	-	
bituminous shale	2,4-D ^a	1.9	-	
modified natural zeolite	atrazine ^c	0.4	-	Lemic et al. 2006
	lindane ^a	1.0	-	
	diazinon ^e	1.3	-	

Table 1 (continued)

Other materials				
Material	Substance	MAC (mg/g)	b (L/mg)	Reference
chisosan alginate (microgranules shells)	salicylic acid ^h	3.0	-	Ding et al. 2009
	2,4-D ^a	4.3	-	
chitosan beads	amoxicilin ⁱ	8.7	-	Adriano et al. 2005
polymeric resin (lewatit VP OC 1163)	carbamazepine ^j	3.9	-	Vergeli and Barlas 2009
	propyphenazone ^f	3.9	-	
	sulfamethoxazole ⁱ	3.9	-	
sephabeads SP207 (polymeric)	salicylic acid ^h	81.6	2.03E-07	Otero et al. 2004
sephabeads SP206 (polymeric)	salicylic acid ^h	45.2	1.27E-08	
rice bran	captan ^a	4.9	-	Adachi et al. 2001
	chloromethoxyphen ^a	4.4	-	
	chloronitrophen ^b	4.7	-	
	cypermethrin ^b	0.2	-	
	dichlofluanid ^b	4.9	-	
	fenoxycarb ^b	2.6	-	
	phthalide ^a	2.7	-	
	A-HCH ^b	4.2	-	
	hexachlorobenzen ^a	4.3	-	
	hexythiazox ^b	3.5	-	
	dicofol ^a	4.4	-	
	methopren ^b	3.9	-	
	carbendazim ^b	1.1	-	
	oxadiazon ^b	3.9	-	
	phoxim ^a	3.3	-	
	propyzamide ^b	2.0	-	
	pyrazoxyfen ^b	3.9	-	
tetradifon ^a	4.6	-		
cholorobenzilate ^b	3.9	-		
triuoram ^b	2.7	-		
clofentezin ^b	4.8	-		
chorothalonil ^a	4.2	-		
water melon peel	methyl parathion ^e	6.4	-	Memon et al. 2008
chestnut shell	carbofuran ^b	2.4	-	Memon et al. 2007
	methyl parathion ^e	5.9	-	
rice husk	triazofos ^c	0.9	0.08	Akhtar et al. 2009
rice bran	triazofos ^c	1.0	0.15	
XAD-7 (polymeric)	amitrole ^b	5.4	0.01	Kyriakopoulos et al. 2005
	prometryn ^c	3.9	0.03	
	alachlor ^b	7.4	0.10	
	trifluralin ^b	10.2	0.09	
XAD-4 (polymeric)	amitrole ^b	4.7	0.002	
	prometryn ^c	11.3	0.02	
	alachlor ^b	7.6	0.03	
carboneous (i-ustrial waste)	trifluralin ^b	11.2	0.26	
	2,4-D ^a	212	0.02	
	carbofuran ^b	208	0.02	
BF sludge	2,4-D ^a	30.0	-	Gupta et al. 2006
	carbofuran ^b	23.0	-	
BF dust	2,4-D ^a	21.0	-	
	carbofuran ^b	13.0	-	
BF slag	2,4-D ^a	0.0	-	
	carbofuran ^b	0.0	-	

3.2.1. MAC values

AC materials showed generally high MAC values for the adsorption of PhACs and pesticides. MAC values reported for Filtrasorb® 400 AC reach 137, 161 and 351 mg g⁻¹ for adsorption of 2,4-dichlorophenoxyacetic acid (2,4-D) (Kim et al. 2008), lindane (Sotelo et al. 2002) and salicylic acid (Otero et al. 2004), respectively. ACs synthesized from non-conventional materials also show high MAC values, at 238 mg g⁻¹ for the adsorption of 2,4-D in contact with AC processed from date stones (Hameed et al. 2009) or 156 mg g⁻¹ for the adsorption of carbofuran in contact with AC processed from banana stalks (Salman and Hameed 2010).

Mineral and other alternative materials generally post lower MAC values than ACs. For mineral materials, reported values are in the range 0.1 to 50 mg g⁻¹ for PhACs and 0.1 to 17 mg g⁻¹ for pesticides. Values ranged from 0.1 to 0.4 mg g⁻¹ for several pharmaceuticals (carbamazepine, clofibrac acid, diclofenac, ibuprofen, ketoprofen) in contact with a mesoporous silica (Bui and Choi 2009) and from 0.4 to 1.3 mg g⁻¹ for pesticides (atrazine, lindane, diazinon) in contact with zeolite (Lemić et al. 2006). Values in the range 5 to 50 mg g⁻¹ were obtained for 6 antibiotics (tetracycline and derivatives, and sulfamethoxazol and derivatives) in contact with modified clay (Polubesova et al. 2006) and for salicylic acid and ciprofloxacin in contact with mineral materials such as goethite, kaolinite or bentonite (Xu et al. 2007; Zhang and Huang 2007). Only thermally or chemically-treated modified clays (montmorillonites, rectorites and palygorskites) had high MAC values of 460, 140 and 99 mg g⁻¹, respectively, for the adsorption of tetracycline antibiotic (Chang et al. 2009a; Chang et al. 2009b).

For alternative materials, MAC values ranged from 1 to 30 mg g⁻¹, i.e. similar to the range measured for mineral adsorbents. Only two adsorbents showed higher MAC values: sephabeads (polymeric material) at 45 and 82 mg g⁻¹ for salicylic acid (Otero et al. 2004), and carbonaceous industrial waste at MAC values ranging from 208 and 212 mg g⁻¹ for the adsorption of two pesticides (carbofuran and 2,4-D, respectively) (Gupta et al. 2006).

3.2.2. b values

Published papers gave b Langmuir parameter values for less than a half of the 108 MAC data items collected (43 b data). Except for specially modified adsorbents, b values are generally 10-fold higher for ACs than for mineral/other adsorbent materials (around 0.1 L mg⁻¹ for AC *versus* 0.01 L mg⁻¹ for alternative adsorbent materials). Specific treatment and/or synthesis can yield very high b values for alternative adsorbents (up to 30 L mg⁻¹) showing a great affinity between adsorbent material and target xenobiotic (Bui and Choi 2009).

4. DISCUSSION

4.1 Comparing alternative adsorbent materials to activated carbons

Despite high differences between reported experiment results, our database demonstrated that ACs boast very high adsorption capacities for hydrophilic xenobiotics and clearly outperform other mineral or alternative material adsorbents. Data on alternative adsorbent materials to ACs is rare and largely inappropriate for comparing potential xenobiotic adsorbability, as there is little data available on the same xenobiotic treated with different adsorbent materials.

MAC values for both ACs and alternative adsorbents were only available for 5 pesticides (2,4-D, carbofuran, alachlor, atrazine, lindane) and 4 PhACs (amoxicilin, ibuprofen, ketoprofen, salicylic acid). ACs showed 10 to 100-fold higher MAC values than alternative adsorbent materials: for 2,4-D adsorption, AC and chitosan alginate adsorbent materials

reached MAC values of 137 and 4.3 mg g⁻¹, respectively (Kim et al. 2008; Ding et al. 2009), and for ibuprofen adsorption, AC from coal and mesoporous silica reached MAC values of 430 and 0.4 mg g⁻¹, respectively (Mestre et al. 2009; Bui and Choi 2009). The only b parameter values available for both ACs and alternative adsorbent materials concerned a pharmaceutical (ibuprofen). In this case, b value was very high (13.3 L mg⁻¹) for the alternative material as it is chemically modified, but this value remains unusual for such adsorbents.

To explore the differences between reported experiment results and get a more thorough basis for cross-comparing adsorption capacities between alternative adsorbent materials, different materials need to be tested using a common batch-test procedure. Indeed, from one study to another, the data were collected under various experimental conditions working with different initial xenobiotic concentrations (from dozens to several hundred mg L⁻¹) and different solid/liquid ratios (0.01 to 100 g L⁻¹), as shown in Online Resource 2. Besides the fact that these differences limit the scope for comparison between alternative materials, those values are very far from the concentration levels measured in WWTPs effluents (few ng L⁻¹ to µg L⁻¹). These differences may support competition phenomenon or pore blocking, and may alter the data quality and utilization of adsorption results published in literature (Limousin et al. 2007; De Ridder et al. 2011).

4.2 Relationships between Langmuir parameters and adsorbent and xenobiotic characteristics

The aim here was to determine potential relationships between Langmuir parameters and the characteristics of substances and adsorbent materials in order to predict MAC and b parameters based on material characteristics. We ran principal component analysis (PCA) on

the dataset composed of the 22 cases where the following variables were fully documented in the database: MAC and b Langmuir parameters, specific area and pore volume of the adsorbent material (other characteristics were too rarely mentioned), log K_{ow} , molecular weight and solubility of the xenobiotics (Figure 2). The subset of data was composed of 11 data items from ACs, 6 from mineral materials and 5 from other alternative materials.

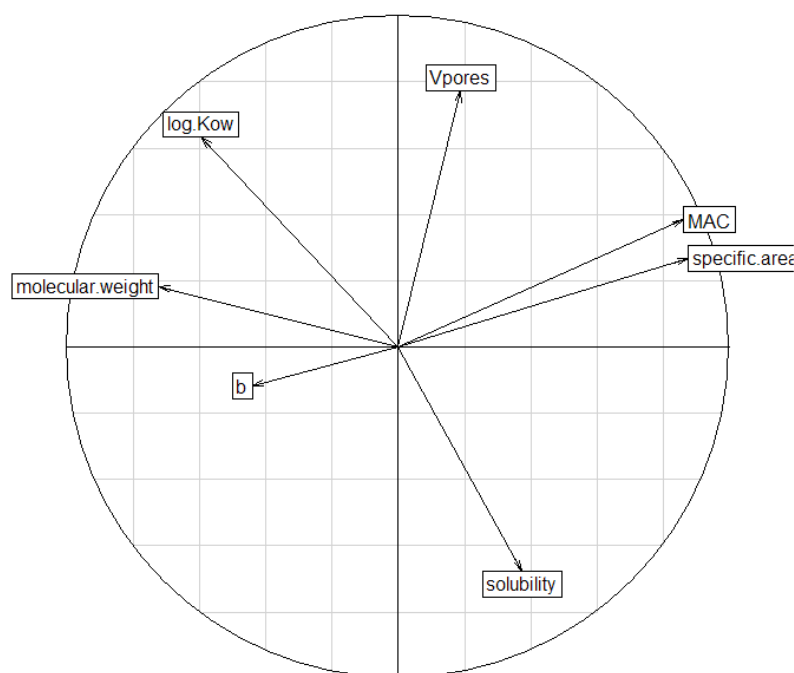


Fig 2 Correlation circle of PCA analysis (axes 1-2) with Langmuir parameters (MAC and b) and possible explanatory variables for studied adsorbent materials (specific surface area and pore volume) and for target xenobiotics (molecular weight, solubility, log K_{ow}). Data set: $n = 22$.

As shown in Figure 2, we found a relationship between MAC and specific area, as the projections of vectors onto the plane represented by axes 1-2 were collinear, and the variance was well explained. In contrast, MAC was unrelated to pore volume of adsorbent materials and unrelated to substance characteristics such as log K_{ow} and solubility. These results warrant care, as they only concern a limited dataset ($n = 22$). We therefore checked the robustness of the approach by plotting MAC *versus* specific area with a larger dataset extracted from the database ($n = 106$) (data not shown). We found no evidence of the

relationship suggested earlier (via PCA analysis) between MAC and specific surface area of the adsorbent materials. Indeed, some materials with a high specific area ($> 1000 \text{ m}^2 \text{ g}^{-1}$) showed low MAC values, and vice-versa. Thus, the MAC parameter could not be predicted based solely on specific area of the materials. The observed differences may be explained by other parameters rarely mentioned in research papers, such as pore volume, pore diameter or chemical characteristics. To go further in the interpretation and/or prediction of adsorption results, researchers should also consider xenobiotic hydrophobicity, polarizability, aromaticity and presence of H-bond donor/acceptor moieties, as with the quantitative structure–activity relationship (QSAR) model developed for one AC (de Ridder et al. 2010). The b Langmuir parameter representing xenobiotic affinity to material surface was not correlated to any of the considered variables. This could be due to the fact that this PCA was unable to integrate information on the material surface chemistry or chemical moieties of the xenobiotics.

4.3. Influence of b Langmuir parameter on adsorption

b values were unfortunately not systematically published with MAC values (108 MAC items but only 43 b items). This is nevertheless a key issue when studying xenobiotics present at very low concentrations. If we consider for instance the adsorption of pesticide 2,4-D, AC F300 gave MAC and b values of 182 mg g^{-1} and 0.1 L mg^{-1} , respectively; while chitosan alginate gave values of 4.3 mg g^{-1} and an undetermined b value (Ding et al. 2009). Based on MAC values only, AC emerges as the best material for the adsorption of 2,4-D, but for a low equilibrium concentration in liquid of $C_e = 0.001 \text{ mg L}^{-1}$, chitosan alginate showed a higher equilibrium concentration in solid ($Q_e = 0.042 \text{ mg g}^{-1}$) than AC ($Q_e = 0.018 \text{ mg g}^{-1}$). Thus, considering the strong influence of the Langmuir b parameter on Q_e at low C_e levels, we recommend systematically evaluating this parameter for use in WWTP applications.

5. CONCLUSIONS

Using an original database approach, we compiled literature values of Langmuir parameters (MAC and b) for various adsorbents in contact with synthetic water spiked with individual xenobiotics (pesticides and pharmaceuticals). A total of 108 MAC and 43 b Langmuir data items were sorted for three types of adsorbents (i.e. ACs, mineral adsorbents, alternative adsorbent materials). We found 12 and 22 MAC values for the adsorption of PhACs and pesticides on ACs but only 33 and 41 MAC values for the adsorption of PhACs and pesticides on all alternative adsorbent materials. Using our database, we demonstrated that activated carbons offer clearly superior adsorption capacities for hydrophilic xenobiotics compared to other mineral or alternative materials that perform far less well. But the lower price of alternative materials compared to activated carbons could be a driving force for their use in wastewater treatment. Indeed, activated carbons cost about 1000 € per ton, while alternative materials such as clays and zeolites cost about 30 € per ton (Rakč et al. 2012) The broad range of reported values and low number of data available make it difficult to reliably cross-compare alternative materials. To progress on the comparison of adsorption capacities between alternative adsorbent materials, we need a higher number of data, covering the full panel of potential adsorbent materials. Data produced using a common batch-test procedure would greatly improve the comparability of adsorption performances. We recommend that authors detail the physical-chemical characteristics (e.g. surface chemistry) of the adsorbent material and determine Langmuir b parameter together with MAC value, as b parameter greatly influences the calculation of the adsorbed value Q_e .

The work underlines that on the basis of the available literature data, it remains difficult to select an alternative material to AC capable of adsorbing pesticides and PhACs at a tertiary treatment stage. Further lab-scale experiments should be carried out using a common

experimental protocol for different adsorbents. Studying adsorption under the presence of a cocktail of several xenobiotics (not studied individually) would give useful information on competition and interactions.

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Supplementary Material 1 References of the data contained in the final database (data summarized in Table 1)

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Supplementary Material 2 Xenobiotics studied via adsorption batch assays in literature

Xenobiotic	Family	Raw formula	Molecular mass (g/mol)	Solubility (mg/L)	Saturation vapor pressure (Pa)	Henry constant (Pa.m ³ /mol)	Log Kow (-)	pKa (-)
Pharmaceuticals (PhACs)								
2,4-dichlorophenoxyacetic acid	organochlorine	C8 H6 Cl2 O3	221	23180	1,9E-05	1,3E-05	-0,8	2,8
2-methyl-4-chlorophenoxy acetic acid (MCPA)	aryloxyacid	C19 H9 Cl O3	201	293900	4,0E-04	5,5E-05	-0,8	3,7
4-(4-chloro-o-tolyloxy)butyric acid (MCPB)	organochlorine	C11 H13 Cl O3	229	48	5,8E-05	2,7E-04	-	6,2
alachlore	amide	C14 H20 Cl N O2	270	187	2,9E-03	8,3E-05	3,5	-
aldicarb	carbamate	C7 H14 N2 O2 S	190	6000	1,0E-02	-	1,4	-
ametryn	triazine	C9 H17 N5 S	227	209	3,7E-04	2,4E-04	3,0	4,1
amitrole	triazole	C2 H4 N4	84	280000	5,9E-05	2,2E-08	-0,9	4,2 ; 10,7
atrazine	triazine	C8 H14 Cl N5	216	33,5	3,9E-05	1,5E-04	2,7	1,7
bentazon	benzothiadiazone	C10 H12 N2 O3 S	240	570	1,8E-01	7,2E-05	-0,5	3,3
bromoxynil	hydroxybenzotrile	C7 H3 Br2 N O	277	90	1,7E-04	5,3E-04	1,0	3,9
captan	organochlorine	C9 H8 Cl3 N O2 S	301	4,9	4,2E-06	3,0E-04	2,6	-
carbendazim	Carbamate	C9 H9 N3 O2	191	29	1,0E-07	2,1E-06	1,5	4,2
carbofuran	Carbamate	C12 H15 N O3	221	700	2,7E-03	2,5E-05	1,5	-
chlornitrofen	organochlorine	C12 H6 Cl3 N O3	319	0,764	7,0E-05	1,9E-02	5,0	-
chlorobenzilate	organochlorine	C16 H14 Cl2 O3	325	13	negligible	-	4,7	-
chloromethoxyphen	organochlorine	C13 H9 Cl2 N O4	314	0,3	1,9E-03	1,5E-03	-	-
chlorothalonil	organochlorine	C8 Cl4 N2	266	0,6	7,6E-05	2,0E-01	3,1	-
clofentezine	tetrazine	C14 H8 Cl2 N4	303	1	1,3E-07	-	3,1	-
cypermethrin	organochlorine	C22 H19 Cl2 NO3	416	0,009	2,3E-07	2,4E-02	5,3	-
diazinon	organophosphate	C12 H21 N2 O3 P S	304	60	0,0E+00	-	3,1	-
dichlofluanid	pesticide	C9 H11 Cl2 F N2 O2 S2	333	1,3	1,5E-05	3,8E-03	3,7	-
dicofol	organochlorine	C14 H9 Cl5 O	370	0,8	5,3E-05	2,4E-02	5,0	-
dinoseb	phenol derivate	C10 H12 N2 O5	240	52	7,0E-03	4,6E-07	3,6	4,6
diuron	urea	C9 H10 Cl2 N2 O	233	38,7	9,2E-06	5,1E-05	2,7	-
fenoxycarb	carbamate	C17 H19 N O4	301	6	1,7E-05	1,0E-03	4,8	-
hexachlorobenzene	organochlorine	C6 Cl6	285	0,005	1,0E-03	-	5,5	-
hexythiazox	thiazolidinone	C17 H21 Cl N2 O2 S	353	0,5	3,4E-06	1,2E-02	2,7	-
imazalil	organochlorine	C14 H14 Cl2 N2 O	297	184	1,6E-07	1,1E-04	3,7	6,5
isoproturon	urea	C12 H18 N2 O	206	67,6	3,3E-06	1,5E-05	2,7	-
lindane (alpha-HCH)	organochlorine	C6 H6 Cl6	291	7,3	1,2E-03	-	3,7	-
mecoprop (MCPP)	aryloxyacid	C10 H11 Cl O3	215	620	1,6E-03	1,8E-08	3,1	3,1
methoprene	pesticide	C19 H34 O3	310	1,4	3,1E-03	6,9E-01	5,5	-
methyl parathion	organophosphate	C8 H10 NO5 PS	263	37,7	4,7E-04	1,0E-02	2,9	7,2
oxadiazon	oxadiazole	C15 H18 Cl2 N2 O3	345	1	1,0E-03	3,6E-07	4,8	-
phoxim	organophosphate	C12 H15 N2 O3 P S	298	4,1	2,1E-03	1,5E-01	4,4	-
phthalide	organochlorine	C8 H2 Cl4 O2	272	2,5	3,0E-06	3,2E-04	3,2	-
prometryn	triazine	C10 H19 N5 S	241	33	2,7E-04	1,3E-03	3,5	4,1
propanil	amide	C9 H9 Cl2 N O	218	152	1,2E-02	-	3,1	-
propyzamide	benzamide	C12 H11 Cl2 N O	256	9	5,8E-05	9,8E+00	3,4	-
pyrioxifen	pesticide	C20 H29 N O3	321	0,367	1,4E+01	-	5,4	-
tetradifon	organochlorine	C12 H6 Cl4 O2 S	356	0,078	1,0E-03	-	4,6	-
thirame	organosulfur	C6 H12 N2 S4	240	0,0165	2,3E-03	3,3E-02	1,7	8,2

triazophos	triazine	C12 H16 N3 O3 P S	313	39	3,9E-04	4,8E-03	3,3	-	
trifluralin	pesticide	C13 H16 F3 N3 O4	335	0,184	6,1E-03	1,0E+00	5,3	-	
Pesticides									
amoxicillin	antibiotic	C16 H19 N3 O5 S	365	3430	6,3E-15	2,5E-16	0,9	-	
carbamazepine	antidileptic	C15 H12 N2 O	236	17,68	2,3E-05	1,1E-05	2,2	<1 ; 14	
chlortetracycline hydrochloride	antibiotic	C22 H23 Cl N2 O8, xCl-H	515	-	-	-	-	3,3	
ciprofloxacin	antibiotic	C17 H18 F N3 O3	331	30000	2,2E-10	5,2E-14	0,3	6,1 ; 8,7	
clofibrilic acid	hypolipemic	C10 H11 Cl O3	215	582,75	1,0E-02	2,2E-03	2,7	3,0	
diclofenac	analgesic	C14 H11 Cl2 N O2	296	2,3775	5,5E-06	4,8E-07	3,5	4,2	
ibuprofen	analgesic	C13 H18 O2	206	21	2,5E-02	1,5E-02	3,4	4,9	
ketoprofen	analgesic	C16 H14 O3	254	51	5,0E-05	2,2E-06	3,1	4,5	
levodopa	parkinson disease	C9 H11 N O4	197	5000	1,7E-08	1,3E-13	-2,4	2,3	
oxytetracycline hydrochloride	antibiotic	C22 H24 N2 O9, Cl-H	497	-	-	-	-	3,3 ; 7,3 ; 9,1	
propyphenazone	analgesic	C14 H18 N2 O	230	3000000	6,9E-04	1,8E-04	1,9	-	
salicylic acid	metabolite	C7 H6 O3	138	2240	1,1E-02	7,3E-09	2,2	3,0	
sulfamethizole	antibiotic	C9 H10 N4 O2 S2	270	1050	2,7E-07	2,6E-09	0,5	-	
sulfamethoxazole	antibiotic	C10 H11 N3 O3 S	253	610	9,2E-06	6,5E-08	0,9	1,8 ; 5,6	
sulfisoxazole	antibiotic	C11 H13 N3 O3 S	267	300	6,8E-06	1,1E-07	1,0	-	
tetracycline hydrochloride	antibiotic	C22 H24 N2 O8, Cl-H	481	231	4,8E-23	-	-1,3	3,3	

Supplementary Material 3 Experimental conditions of adsorption batch assays found in literature

Adsorbent material	Xenobiotic	Water type	p H (°C)	Initial concentration (mg/L)	Rotation frequency (rpm)	Solid / liquid ratio (g _{material} /L)	Time to equil.	Kinetic	Reference
Activated carbons (ACs)									
AC (WD-extra)	2,4-D	synthetic	- 20	-	130	-	24 h	-	Ignatowicz, 2009
	MCPA	synthetic	- 20	-	130	-	24 h	-	
	MCPA	synthetic	- 20	-	130	-	24 h	-	
AC (cork powder)	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	Mestre et al, 2009
AC	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	
AC (recycling PET)	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	
commercial AC (charcoal)	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	
commercial AC (wood)	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	
AC	ibuprofen	synthetic	- 30	20-120	-	-	4 h	pseudo 2nd order	
commercial AC (Spectracarb 2225)	ametryn	synthetic	- 25	103,84	-	-	8-10 h	pseudo 2nd order / pseudo 1st order	
commercial AC (313W)	aldicarb	synthetic	- 25	103,96	-	-	8-10 h	-	Ayranci et Hoda, 2005
	dinoseb	synthetic	- 25	22,38	-	-	8-10 h	-	
	diuron	synthetic	- 25	32,27	-	-	8-10 h	-	
commercial AC (charcoal based)	lindane	synthetic	- 25	0,01	60	0,02	1 h	-	Kouras et al., 1998
commercial AC (darco G-60)	MCPB	synthetic	- 25	0,5	-	-	24 h	-	Hu et al., 1998
	imazalil	synthetic	- 25	0,5	-	-	24 h	-	
commercial AC (Spectracarb 2225)	bromoxynil	synthetic	- 25	-	40	-	24 h	-	Yang et al., 2004
	diuron	synthetic	- 25	-	40	-	24 h	-	
commercial AC (Spectracarb 2225)	bentazon	synthetic	7 25	20,9	-	-	8-10 h	1st order	Ayranci and Hoda., 2004
	propanil	synthetic	7 25	23,52	-	-	8-10 h	1st order	
commercial AC	carbofuran	synthetic	- 25	2-234	-	0,2	24 h	-	Fernandez-perez et al., 2005
commercial AC	isoproturon	synthetic / surface	- -	1-12	-	0,3	120 min	-	Sarkar et al., 2007
AC (date stones)	2,4-D	synthetic	- 30	50-400	130	1,0	9h30	pseudo 1st order	Hameed et al., 2009
AC (banana stalk)	carbofuran	-	7 30	250	120	1,5	24 h	pseudo 2nd order (Ho)	Salman and Hameed, 2010
commercial AC (F300)	2,4-D	synthetic	- 30	50-300	120	1,0	8 h	-	Salman and Hameed, 2010 b
commercial AC (F400)	salicylic acid	synthetic	- -	-	-	-	24 h *	-	Otero et al., 2004
commercial AC (norit row 0,8 SUPRA)	amoxiciline	synthetic	7 -	-	-	-	300 - 400 min	-	Putra et al., 2009
AC (palm oil)	bentazon	synthetic	- 30	100	120	3,0	-	-	Salman and Hameed, 2010 c
commercial AC	2,4-D	synthetic	- 25	-	-	1,0	90 min	-	Gupta et al., 2006
	carbofuran	synthetic	- 25	-	-	1,0	90 min	-	
commercial AC (wood)	levodopa (Parkinson disease)	synthetic	5 5 6 5	31-1281	-	1,0	24 h	-	Quesada-penate et al., 2009

commercial AC (coconut)	levodopa (Parkinson disease)	synthetic	5 5 - 25 6	31-1281	-	0,5	24 h	-	
commercial AC (tree bark)	levodopa (Parkinson disease)	-	5 - 25 6 5	31-1281	-	0,5	24 h	-	
Mineral materials									
modified zeolite	atrazine	synthetic	- -	-	-	-	2 h	-	
	lindane	synthetic	- -	-	-	-	2 h	-	Lemic et al., 2006
	diazinone	synthetic	- -	-	-	-	2 h	-	
mesoporous silica (SBA-15)	carbamazepine	synthetic	5 25	0,1	200	2	few minutes	pseudo 2nd order	
	clofibric acid	-	5 25	0,1	200	2	few minutes	pseudo 2nd order	
	diclofenac	-	5 25	0,1	200	2	few minutes	pseudo 2nd order	Bui and Choi, 2009
	ibuprofen	-	5 25	0,1	200	2	few minutes	pseudo 2nd order	
	ketoprofen	-	5 25	0,1	200	2	few minutes	pseudo 2nd order	
bituminous shale	2,4-D	synthetic	8 -	4 10 ⁻⁴ M	-	20	24 h	-	Ayar et al., 2008
modified montmorillonite	tetracycline hydrochloride	synthetic	- -	5-50	-	10	-	-	
	oxytetracycline hydrochloride	synthetic	- -	5-50	-	10	-	-	
	chlortetracycline hydrochloride	synthetic	- -	5-50	-	10	-	-	Polubesova et al., 2007
	sulfamethoxazole	synthetic	- -	5-50	-	10	-	-	
	sulfisoxazole	synthetic	- -	5-55	-	10	-	-	
palygorskite	tetracycline hydrochloride	synthetic	5 6 8	200	150	5	2 h	pseudo 2nd order	Chang et al., 2009
	tetracycline hydrochloride	synthetic	7	200	150	5	2 h	pseudo 2nd order	
rectorite	tetracycline hydrochloride	synthetic	5 -	50-1000	150	5	24 h	pseudo 2nd order	Chang et al., 2009 b
	tetracycline hydrochloride	synthetic	8 7	50-1000	150	5	24 h	pseudo 2nd order	
Na-montmorillonite	tetracycline hydrochloride	synthetic	5 -	100-3000	150	5	24 h	-	
Ca-montmorillonite	tetracycline hydrochloride	synthetic	5 -	100-3000	150	5	24 h	-	Chang et al., 2009 c
rectorite	tetracycline hydrochloride	synthetic	5 -	50-1000	150	5	24 h	-	

Other materials

	captan	-	-	50	-	10	1,5 h	-	
	chloromethoxyphen	-	-	50	-	10	-	-	
	chloronitrophen	-	-	50	-	10	-	-	
	cypermethrin	-	-	2	-	10	-	-	
	dichlofluanid	-	-	50	-	10	-	-	
	fenoxycarb	-	-	50	-	10	-	-	
	fthalide	-	-	50	-	10	-	-	
	lindane	-	-	50	-	10	-	-	
	hexachlorobenzene	-	-	50	-	10	-	-	
	hexythiazox	-	-	50	-	10	-	-	
rice bran	kelthane	-	-	50	-	10	-	-	Adachi et al., 2001
	methoprene	-	-	50	-	10	-	-	
	carbe-azim	-	-	50	-	10	-	-	
	oxadiazon	-	-	50	-	10	-	-	
	phoxim	-	-	50	-	10	-	-	
	propyzamide	-	-	50	-	10	-	-	
	pyrazoxyfen	-	-	50	-	10	-	-	
	tetradifon	-	-	50	-	10	-	-	
	chlorobenzilate	-	-	50	-	10	-	-	
	triuram	-	-	50	-	10	-	-	
	clofentezine	-	-	50	-	10	-	-	
	chorothalonil	-	-	50	-	10	-	-	
watermelon peel	methyl parathion	-	6	1000	100	5	60 min	1st order	Memon et al., 2008
chestnut shell	carbofuran	-	6	840	100	4	30 min	1st order	Memon et al., 2007
	methyl parathion	-	6	1200	-	4	30 min	-	
rice husk	hostathion	-	6	10	100	8	60 min	1st order	Akhtar et al., 2009
rice bran	hostathion	-	6	10	100	8	60 min	1st order	
rice bran	methyl parathion	-	6	100	100	4	90 min	1st order (Lagergren)	
rice husk	methyl parathion	-	6	100	100	4	90 min	1st order (Lagergren)	Akhtar et al., 2007
fly ash	methyl parathion	-	6	100	100	4	90 min	1st order (Lagergren)	
moringa olifeira husk	methyl parathion	-	6	100	100	4	90 min	1st order (Lagergren)	
	carbamazepine	synthetic	-	25	0,02	150	5	2 h	-
polymeric resin (lewatit VP OC 1163)	propyphenazone	synthetic	-	25	0,02	150	5	4 h	-
	sulfamethoxazole	synthetic	-	25	0,02	150	5	18 h	-
chisosan alginates	2,4-D	synthetic	7	-	$0.3-5.6 \cdot 10^{-4} \text{ M}$	-	4 h	-	Ding et al., 2009
	salicylic acid	synthetic	7	-	$0.3-5.6 \cdot 10^{-4} \text{ M}$	-	4 h	-	
sephabeads SP207	salicylic acid	synthetic	-	-	-	-	24 h *	-	Otero et al., 2004
bentonite	amoxiciline	synthetic	7	-	-	-	300 - 400 min	-	Putra et al., 2009
goethite (syn-FeOOH)	ciprofloxacin	synthetic	-	-	-	-	48 h *	-	Zhang and Huang 2007
goethite (ald-Fe-OOH)	ciprofloxacin	synthetic	-	-	-	-	48 h *	-	
chitosan	amoxiciline	synthetic	-	-	-	-	200 - 300 min	-	Adriano et al., 2005
goethite	salicylic acid	synthetic	-	-	-	-	22 h *	-	Xu et al., 2007
kaolinite	salicylic acid	synthetic	-	-	-	-	22 h *	-	

rhodic ferrasol	salicylic acid	synthetic	-	-	-	-	22 h *	-	
haplic acrisol	salicylic acid	synthetic	-	-	-	-	22 h *	-	
XAD-7 (polymeric)	herbicide	synthetic	-	15-30	200-1000	50-100	4 h	-	Kiriakopoulos et al., 2005
XAD-4 (polyeric)	herbicide	synthetic	-	15-30	200-1000	50-100	4 h	-	
carbonaceous material	2,4-D / carbofuran	synthetic	6						
			5						
			7	-	-	-	1	90 min	2nd order
blast furnace (BF) sludge	2,4-D / carbofuran	synthetic	5						
			6						
			7	-	-	-	1	90 min	2nd order
blast furnace dust	2,4-D / carbofuran	synthetic	6						
			6						
			7	-	-	-	1	90 min	2nd order
blast furnace slag	2,4-D / carbofuran	synthetic	7						
			6						
			7	-	-	-	1	90 min	2nd order
			8						

* reaction time but not specified to be
equilibrium time

Supplementary Material 4 Adsorbent materials studied via adsorption batch assays in literature

Adsorbent material	pH pzk	Specific area (m ² /g)	Particle size (mm)	Pore volume (mL/g)	Pore diameter (nm)	Density (kg/m ³)	Porosity	Availability of surface chemistry	Reference
Activated carbons (ACs)									
	-	1000	1,25	0,90	-	-	-	No	
AC (WD-extra)	-	-	1,3	-	-	-	-	No	Ignatowicz, 2009
	-	-	1,35	-	-	-	-	No	
AC (cork powder)	7,5	891	-	0,42	-	-	-	Yes	
AC	9,9	1060	-	0,57	-	-	-	Yes	
AC (PET recycling)	10,7	1426	-	0,58	-	-	-	Yes	
commercial AC (charboal)	9,2	1156	-	0,65	-	-	-	Yes	Mestre et al, 2009
commercial AC (wood)	8,2	899	-	0,63	-	-	-	Yes	
AC	4,5	879	-	0,55	-	-	-	Yes	
commercial AC (spectracarb 2225)	-	2500	-	-	-	-	-	No	Ayranci and Hoda, 2005
commercial AC (PAC 313W)	-	800	-	-	-	-	-	Yes	Kouras et al., 1998
	-	1340	-	-	-	-	-	No	
commercial AC (charcoal based)	-	-	-	-	-	-	-	No	Hu et al., 1998
	-	-	-	-	-	-	-	No	
commercial AC (darco G-60)	-	776	-	-	-	-	-	No	Yang et al., 2004
	-	-	-	-	-	-	-	No	
commercial AC (spectracarb 2225)	-	-	-	-	-	-	-	No	Ayranci and Hoda., 2004
	-	-	-	-	-	-	-	No	
commercial AC	-	-	-	-	-	-	-	No	Fernandez-perez et al., 2005
commercial AC	-	-	-	-	-	-	-	No	Sarkar et al., 2007
AC (date stones)	-	763	-	0,42	261,8	-	-	No	Hameed et al., 2009
AC (banana stalk)	-	981,62	-	0,46	2,88	-	-	No	Salman and Hameed, 2010
commercial AC (F300)	-	731,48	-	0,45	-	-	-	No	Salman and Hameed, 2010 b
commercial AC (F400)	-	1050	0,65	-	-	425	-	No	Otero et al., 2004
commercial AC (norit row 0,8 SUPRA)	-	1092	-	-	-	-	-	Yes	Putra et al., 2009
AC (palm oil)	-	1237,13	2	0,67	2,16	-	-	No	Salman and Hameed, 2010 c
commercial AC (charcoal)	-	710	-	-	-	-	-	No	Gupta et al., 2006
commercial AC (wood)	6,2	1860	-	0,77	-	-	-	No	
commercial AC (coconut)	9,7	1175	-	0,47	-	-	-	No	Quesada-penate et al., 2009
commercial AC (tree bark)	11	1230	-	0,53	-	-	-	No	
Mineral materials									
modified zeolite	-	-	-	-	-	-	-	No	Lemic et al., 2006
mesoporous silica (SBA-15)	-	737	-	1,03	8	-	-	No	Bui and Choi, 2009
bituminous shale	-	11	0,38	-	-	-	-	Yes	Ayar et al., 2008
modified montmorrillonite	-	-	-	-	-	-	-	No	Polubesova et al., 2007
palygorskite	-	126	-	-	-	-	-	No	Chang et al., 2009
	-	363	-	-	-	-	-	No	
rectorite	-	-	-	-	-	-	-	Yes	Chang et al., 2009 b

Na-montmorillonite	-	725	-	-	-	-	-	Yes	
Ca-montmorillonite	-	717	-	-	-	-	-	Yes	Chang et al., 2009 c
rectorite	-	363	-	-	-	-	-	Yes	
Other materials									
rice bran	-	0,14	-	-	-	-	-	No	Adachi et al., 2001
watermelon peel	-	23,4	-	-	-	-	-	No	Memon et al., 2008
chestnut shell	-	42	-	-	-	-	-	Yes	Memon et al., 2007
rice husk	-	11	-	0,70	41	-	-	Yes	Akhtar et al., 2009
rice bran	-	19	-	0,72	78	-	-	Yes	
rice bran	-	28	-	0,66	88	-	-	Yes	
rice husk	-	17	-	0,69	51	-	-	Yes	
fly ash	-	25	-	0,71	77	-	-	Yes	Akhtar et al., 2007
moringa olifeira husk	-	27	-	0,72	86	-	-	Yes	
polymeric resin (Iewatit VP OC 1163)	-	1300	0,5	0,70	5	-	-	Yes	Vergili and Barlas, 2009
chisosan alginates	-	-	-	-	-	-	-	No	Ding et al., 2009
sephabeads SP207	-	627	0,4	-	-	1180	-	No	Otero et al., 2004
sephabeads SP206	-	557	0,4	-	-	1190	-	No	
bentonite	-	92	-	-	-	-	-	Yes	Putra et al., 2009
goethite (syn-FeOOH)	-	159	-	-	-	-	-	No	Zhang and Huang 2007
goethite (ald-Fe-OOH)	-	82	-	-	-	-	-	No	
chitosan	-	-	2,12	-	-	1,005	0,82	No	Adriano et al., 2005
goethite	-	-	-	-	-	-	-	Yes	
kaolinite	-	-	-	-	-	-	-	Yes	
rhodic ferrasol	-	-	-	-	-	-	-	Yes	Xu et al., 2007
haplic acrisol	-	-	-	-	-	-	-	Yes	
XAD-7 (polymeric)	-	450	0,42	-	30	-	0,50	No	Kiriakopoulos et al., 2005
XAD-4 (polyeric)	-	750	0,42	-	5	-	0,40	No	
carbonaceous material	-	380	-	-	-	-	-	No	
blast furnace (BF) sludge	-	28	-	-	-	-	-	No	Gupta et al., 2006
blast furnace dust	-	13	-	-	-	-	-	No	
blast furnace slag	-	4	-	-	-	-	-	No	