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Removal of poly- and perfluoroalkyl substances (PFAS) from water by adsorption: Role of PFAS chain length, effect of organic matter and challenges in adsorbent regeneration

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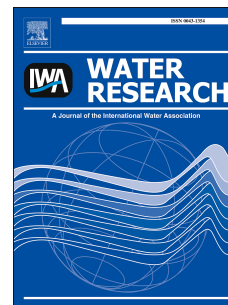
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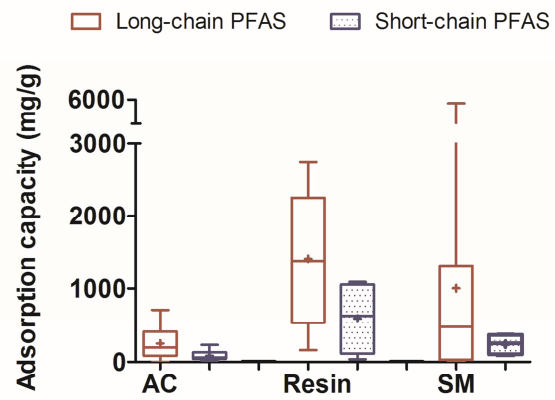
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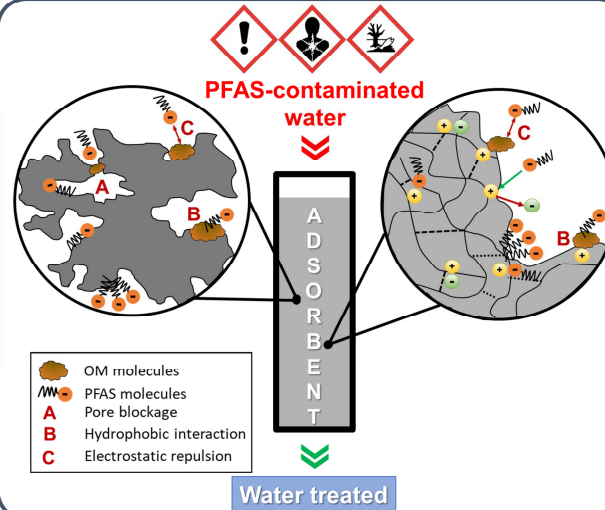
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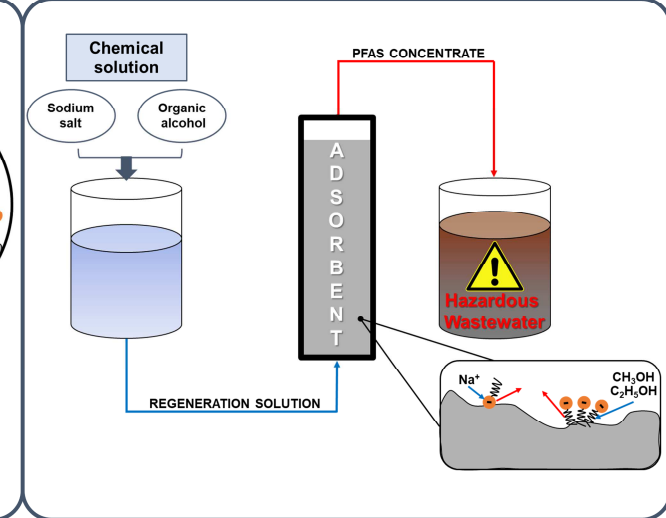
LONG- vs SHORT-CHAIN PFAS



EFFECT OF ORGANIC MATTER



CHEMICAL REGENERATION



1 **Removal of poly- and perfluoroalkyl substances**
2 **(PFAS) from water by adsorption: role of PFAS chain**
3 **length, effect of organic matter and challenges in**
4 **adsorbent regeneration**

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14 **Abstract**

15 Poly- and perfluoroalkyl substances (PFAS) are a wide group of environmentally
16 persistent organic compounds of industrial origin, which are of great concern due to
17 their harmful impact on human health and ecosystems. Amongst long-chain PFAS,
18 perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) are the most
19 detected in the aquatic environment, even though their use has been limited by recent
20 regulations. Recently, more attention has been posed on the short-chain compounds,
21 due to their use as an alternative to long-chain ones, and to their high mobility in the
22 water bodies. Therefore, short-chain PFAS have been increasingly detected in the
23 environmental compartments. The main process investigated and implemented for
24 PFAS removal is adsorption. However, to date, most adsorption studies have focused
25 on synthetic water.

26 The main objective of this article is to provide a critical review of the recent peer-
27 reviewed studies on the removal of long- and short- chain PFAS by adsorption. Specific
28 objectives are to review 1) the performance of different adsorbents for both long- and
29 short-chain PFAS, 2) the effect of organic matter, and 3) the adsorbent regeneration
30 techniques. Strong anion-exchange resins seem to better remove both long- and short-
31 chain PFAS. However, the adsorption capacity of short-chain PFAS is lower than that
32 observed for long-chain PFAS. Therefore, short-chain PFAS removal is more
33 challenging. Furthermore, the effect of organic matter on PFAS adsorption in water or
34 wastewater under real environmental conditions is overlooked. In most studies high
35 PFAS levels have been often investigated without organic matter presence. The rapid

36 breakthrough of PFAS is also a limiting factor and the regeneration of PFAS exhausted
37 adsorbents is very challenging and needs more research.

38 *Keywords:*

39 Adsorption; anion-exchange; Long- and short- chain PFAS; organic matter;
40 regeneration; adsorption mechanism.

Journal Pre-proof

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76 List of abbreviations

Main PFAS and alternative compounds

F-53B	Potassium salt of 6:2 chlorinated polyfluorinated ether sulfonate
FOSA	Perfluorooctane sulfonamide
PFAS	Poly- and perfluoroalkyl substances
PFCAs	Perfluoroalkyl carboxylic acids
PFSAs	Perfluoroalkane sulfonates
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonic acid
PFDA	Perfluorodecanoic acid
PFDoDA	Perfluorododecanoic acid
PFDS	Perfluorodecane sulfonic acid
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptane sulfonic acid
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonic acid
PFNA	Perfluorononanoic acid
PFNS	Perfluorononane sulfonic acid
PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonic acid
PFPeA	Perfluoropentanoic acid
PFPeS	Perfluoropentane sulfonic acid
PFTrDA	Perfluorotridecanoic acid
PFUnDA	Perfluoroundecanoic acid

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Adsorbent materials

2-MNPs@FG	Magnetic-nanoparticles attached into fluorographene (mass ratio of MNPs and FG is 3:5)
AC	Activated carbon
ACF	Activated carbon fiber
ARH	Aminated rice husk
BdAC	Bamboo-derived AC
CTF	Covalent triazine-based framework
DFB-CDP	Cross-linked polymer network where β -Cyclodextrin substitutes decafluorobiphenyl (DFB)
Fe ₃ O ₄ NP	Magnetite nanoparticles
Fe ₃ O ₄ @SiO ₂ -NH ₂ &F ₁₃	Silica membrane functionalized with amino group and octyl-perfluorinated chain on the Fe ₃ O ₄ NP surface
GAC	Granular activated carbon
h-BNs	Porous hexagonal boron nitride nanosheets
HDPE	High-density polyethylene

HMS	Hexagonal mesoporous silica
HWC	Hardwood biochar
MIMs	Macromolecular imprinted materials
MIP-CMs	Molecularly imprinted carbon microspheres
MWNT	Multi-walled carbon nanotubes
NIP-CMs	Non-imprinted carbon microspheres
OD-HMS	N-octyldichlorosilane grafted hexagonal mesoporous silica
PAC	Powdered activated carbon
PACFs	Polyacrylonitrile fiber (PANF)-derived activated carbon fibers
PAF-45	Porous aromatic framework constructed from benzene rings
PCMA	Permanently confined micelle arrays
PEI-f-CMC	Poly(ethylenimine)-functionalized cellulose microcrystals
PS	Polystyrene
PS-COOH	Poly-styrene carboxylic acid
PWC	Pinewood biochar
R-CAC	Reactivated coconut shell-based AC
SWNT	Single walled carbon nanotubes

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Other

AE	Anion-exchange
AFFF	Aqueous film-forming foam
AOPs	Advanced oxidation processes
ATRP	Atom transfer radical polymerization
BET	Brunauer, Emmett and Teller
BSA	Bovine serum albumin
DBP	Disinfection by-products
DI	Deionized water
DMC	Methacryloyloxyethyl trimethyl ammonium chloride
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
DW	Drinking water
DWTPs	Drinking water treatment plants
EBCT	Empty bed contact time
EfOM	Effluent organic matter
EWTP	Electroplating WWTP
FA	Fulvic acids
GW	Groundwater
HA	Humic acids
LW	Lake water
MB	Methylbenzene

MPLs	Microplastics
OM	Organic matter
NDMA	N-Nitrosodimethylamine
NOM	Natural organic matter
NPs	Nanoparticles
PFSOF WWW	Perfluorooctanesulfonyl fluoride washing wastewater
RSSCT	Rapid small-scale column tests
RW	River water
S _{BET}	BET surface area
SDS	Sodium dodecyl sulphate
SeaW	Seawater
SUVA ₂₅₄	Specific ultra-violet absorbance at 254 nm
SW	Surface water
TCE	Tri-chloroethylene
TEM	Transmission electron microscopy
TMFA	2-(trifluoromethyl)acrylic acid
TOC	Total organic carbon
WW	Wastewater
WWTPs	Wastewater treatment plants

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80

1. Introduction

1.1. PFAS classification

Poly- and perfluoroalkyl substances (PFAS) are a large group of anthropogenic aliphatic compounds that are of increasing concern worldwide due to their impact on environment and human health. For many decades, they have been used in an array of formulations as surfactants intermediates (i.e., firefighting foams, hydraulic fluids for aircrafts), waterproofing treatment for textiles (i.e., apparel, carpet) and household products (i.e., paper and non-stick cookware coating) (Buck et al., 2011; Rahman et al., 2014; Xiao, 2017). PFAS contamination is the result of consumer-product degradation, firefighting activities and discharges of fluorochemical manufacturing facilities (Hu et al., 2016).

PFAS are made up of a chain of varying carbon length, on which at least one (polyfluoroalkyl acids) or all (perfluoroalkyl acids) of the hydrogen atoms bound to carbon-chain in the nonfluorinated substances have been replaced by fluorine atoms (Buck et al., 2011). Their chemical structure also includes a charged functional group commonly carboxylic or sulfonic acids attached at one end. Polyfluoroalkyl acids have also been indicated as “precursors” of perfluoroalkyl ones, hence the non-fluorinated bond provides a biotic or abiotic degradation pathway (ITRC, 2017). Depending on their terminal functional group, PFAS can be distinguished in perfluoroalkyl carboxylic acids (PFCAs) and in perfluoroalkyl sulfonic acids (PFSA) (Buck et al., 2011). However, an abridged way to distinguish among PFAS subclasses is that referred to carbon-chain length. According to the Organization for Economic Co-operation and Development (OECD), the term “long-chain compound” indicates PFCAs with eight or more carbons, and PFSA with six or more carbons. Whereas, “short-chain” is related to PFCAs with

105 seven or fewer carbons and PFSA with five or fewer carbons (OECD/UNEP Global
106 PFC Group, 2013). It should be noted that PFCAs and PFSA classification, based on
107 chain length, is different in terms of number of carbon atoms. However, PFAS
108 bioaccumulation and biomagnification into biota increase with the increasing of carbon-
109 chain length, PFSA are more bioaccumulate than PFCAs with the same chain length
110 (Ahrens, 2011).

111 **1.2. PFAS properties**

112 The knowledge of PFAS physical and chemical properties is paramount to understand
113 their fate and transport in the environment. The C-F bond is the strongest covalent
114 bond in organic chemistry and it leads to PFAS thermal stability (ITRC, 2017). In
115 addition, PFAS exhibit mutually hydrophobic and lipophobic properties, which are
116 primarily attributed to the low polarizability of fluorine atoms. Moreover, the terminal
117 functional group attached to fluoroalkyl chain provides PFAS chemical stability. With the
118 increasing replacement of hydrogen by fluorine as well as the increasing carbon-chain
119 length, PFAS become more chemically inert. On the contrary, their water solubility
120 increases with the decreasing carbon-chain length (Kucharzyk et al., 2017). PFCAs and
121 PFSA are characterized by a low vapour pressure, which makes air stripping an
122 unsuitable technique for PFAS removal (Rahman et al., 2014). With the increasing C-F
123 chain length, PFAS become more lipophilic as demonstrated by octanol/water partition
124 coefficients (K_{ow}). At environmentally relevant pH, PFAS exhibit anionic species
125 according to their low values of acid dissociation constant (pK_a). Other properties, such
126 as resistance to thermal, biological and chemical degradation and redox stability, impart
127 unique features to PFAS, making them suitable for several industrial applications.
128 Information regarding chemical formulas and Chemical Abstracts Service (CAS) of

129 selected per-fluoroalkyl substances along with their physical and chemical properties
130 are reported in Table 1.

131 **1.3. Occurrence of PFAS in water**

132 In the aquatic environment, PFAS have been detected at low concentrations ranging
133 from pg L^{-1} to $\mu\text{g L}^{-1}$ (Kaboré et al., 2018; Rahman et al., 2014) and long-chain
134 compounds, such as perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid
135 (PFOA), are the most commonly measured PFAS around the world (ITRC, 2017).
136 Widespread occurrence of PFAS in urban water cycle including wastewater (WW), river
137 water (RW), lake water (LW), drinking water (DW), stormwater, and groundwater (GW)
138 has been demonstrated by several studies (Appleman et al., 2014; Arvaniti and
139 Stasinakis, 2015; Ateia et al., 2019; Hölzer et al., 2009; Munoz et al., 2017; Rahman et
140 al., 2014). The amount of PFAS not removed in wastewater treatment plants (WWTPs)
141 and, consequently, released into the receiving environment is a concern for the possible
142 presence of these compounds in water used for potable supply production.

143 A recent research suggests that the possible impacts of PFAS on GW is linked to the
144 use of treated WW for irrigation (Szabo et al., 2018). The occurrence and distribution of
145 PFAS remain not completely identified, due to the limited data about the removal
146 efficiency of conventional water treatment plants and about the transformation of PFAS
147 precursor compounds (Appleman et al., 2014; Arvaniti and Stasinakis, 2015).

148 Although the use of PFOS and PFOA has been reduced due to their health impact, the
149 total amount of PFAS introduced into the environment has not been reduced because
150 the long-chain compounds have been replaced by short and ultra-short PFAS (Ateia et
151 al., 2019). The latter are potentially less bioaccumulated and bioconcentrated through

152 trophic levels as compared to long-chain but they are still environmentally persistent
153 (Brendel et al., 2018; OECD/UNEP Global PFC Group, 2013).

154 **1.4. PFAS toxicity**

155 PFAS are absorbed in human body after oral ingestion, they are not metabolized and
156 they are detected in human tissue and blood serum at typical concentrations of ng mL^{-1} .
157 Long-chain PFAS are taken up and stored preferentially in the liver. Half-life values of
158 PFOS and PFOA in humans range from 2.3 to 5.4 years (Hölzer et al., 2009; Stubbleski
159 et al., 2017). Epidemiological studies have shown that the occurrence of PFOA and
160 PFOS in humans is probably linked to a high incidence of thyroid disease, high
161 cholesterol, ulcerative colitis, kidney cancer, testicular cancer, and pregnancy-induced
162 hypertension (Crawford et al., 2017; Domingo and Nadal, 2017). More details
163 concerning PFAS toxicological properties are reported in literature (Dewitt, 2015).

164 Based on WHO's recommendation and on the precautionary principle, the proposal for
165 a recast of the Drinking Water Directive (98/83/CE, Directive on the quality of water
166 intended for human consumption) published on 1st February 2018, has included the
167 group of poly- and perfluoroalkyl substances in the list of regulated chemicals. The
168 proposal recommends the values of $0.5 \mu\text{g L}^{-1}$ for the total amount of PFAS and $0.1 \mu\text{g}$
169 L^{-1} for each single compound. In 2017, the Veneto Region (Italy) set drinking water
170 quality standards for PFAS (Veneto Legislation 1590/201), with a limit value of 90 ng L^{-1}
171 for PFOA and PFOS (with $\text{PFOS} < 30 \text{ ng L}^{-1}$), and a limit value of 300 ng L^{-1} for the
172 other PFAS (sum of PFBA, PFPeA, PFBS, PFHxA, PFHpA, PFHxS, PFNA, PFDeA,
173 PFUnA, PFDoA). Such regulation came after a large contamination of surface water
174 and GW due to industrial WW discharges, with the subsequent bioaccumulation of

175 PFAS in the serum of the exposed population. As a result, PFAS are emerging
176 contaminants already regulated in Italy.

177 **1.5. PFAS removal**

178 Conventional treatment processes are ineffective for PFAS degradation due to the high
179 straight of the covalent C-F bond of their chain, their low concentration in water and
180 their high hydrophilicity (Rahman et al., 2014; Ross et al., 2018). Biological treatment
181 both aerobic and anaerobic are only able to break the C-C bond, and they lead to the
182 formation of short-chain PFAS. When advanced treatment technologies are not
183 implemented, biological WWTPs affect the receiving water bodies and the concentration
184 of PFAS detected in finished water is often higher than in the untreated water (Arvaniti
185 and Stasinakis, 2015; Kaboré et al., 2018). This fact gives evidence to recalcitrant
186 behaviour of PFAS to biological treatment and, consequently, their formation via
187 biodegradation of precursor compounds (Rahman et al., 2014). According to data
188 obtained from monitoring studies, PFAS with long carbon-chain, such as PFOS, tend to
189 accumulate in sludge, whereas perfluorobutane sulfonic acid (PFBS) has been mainly
190 detected in the effluents of WWTPs and in drinking water treatment plants (DWTPs)
191 (Arvaniti and Stasinakis, 2015; Rahman et al., 2014).

192 Under typical water treatment plant conditions, disinfection by free chlorine or UV
193 irradiation are ineffective (Appleman et al., 2014; Rahman et al., 2014). PFAS are also
194 much more resistant than other micro-contaminants to the oxidation by ozone and
195 hydroxyl radicals due to the strong C-F bond and the electron withdrawing of their
196 functional groups (Nzeribe et al., 2019; Trojanowicz et al., 2018). Furthermore,
197 advanced oxidation processes (AOPs) often result in a partial degradation of PFAS,

198 with formation of PFAS with shorter perfluorinated alkyl chain (Nzeribe et al., 2019;
199 Trojanowicz et al., 2018).

200 Adsorption, anion-exchange (AE), high pressure nanofiltration and reverse osmosis
201 membrane processes are effective for substantial PFAS removal (Appleman et al.,
202 2013; Du et al., 2014; Kucharzyk et al., 2017).

203 Main issues inherent with PFAS removal through these mentioned techniques are the
204 influence of water matrix (i.e., competing anions, organic matter), frequent regeneration
205 (due to fast breakthrough), and disposal of concentrate with high PFAS concentration
206 (Appleman et al., 2013; Rahman et al., 2014; Zaggia et al., 2016).

207 Adsorption is actually an established technology for PFAS removal, both as single
208 process of point-of-use applications and as step of water treatment plants (Arvaniti and
209 Stasinakis, 2015; Eschauzier et al., 2012; Quiñones and Snyder, 2009). Moreover, it
210 shows a more economical performance compared to high pressure membrane
211 processes (Rizzo et al., 2019; Roccaro et al., 2013).

212 As previously mentioned, PFAS exist as anions at ambient pH values and this makes
213 strong base AE resins suitable for their removal. However, AE treatment efficiency
214 depends on the resin properties, including porosity, functional group and polymer matrix
215 (Deng et al., 2010).

216 To date, adsorption studies have addressed single long-chain PFAS (i.e., PFOA and
217 PFOS), even though long- and short-chain co-removal requires further investigation
218 (Ateia et al., 2019; Maimaiti et al., 2018; McCleaf et al., 2017). Moreover, many
219 compounds, commonly present in water bodies, could affect adsorption efficiency and

220 additional studies should regard their potential competition (Ateia et al., 2019; Du et al.,
221 2015, 2014; Yu et al., 2012).

222 **2. PFAS adsorption and scope of the review**

223 **2.1. PFAS adsorption mechanism**

224 Electrostatic and hydrophobic interactions are considered the predominant forces that
225 govern adsorption of PFAS on several adsorbent materials. Other less important
226 mechanisms of PFAS adsorption are hydrogen bonding and covalent bonding. PFAS
227 molecular structure, adsorbent chemistry and physical properties (i.e., surface functional
228 groups, polarity, and porosity), as well as liquid phase composition should be
229 considered as the main factors affecting PFAS adsorption efficiency (Du et al., 2014).

230 Electrostatic interactions occur between anionic PFAS and adsorbent material positively
231 charged. Consequently, any changes in ionic strength (i.e., co-existing inorganic anions
232 and changing solution pH) could affect adsorption efficiency, due to electrostatic
233 repulsions (Du et al., 2014). Change in solution pH has an effect on the charge
234 characteristics of PFAS molecules and surface properties of adsorbents (Liu et al.,
235 2018; Yu et al., 2008). As reported in several studies, an increase in pH solution led to a
236 decrease in adsorption capacity of most adsorbents (Deng et al., 2010; Gao et al.,
237 2017; Qian et al., 2017). Nevertheless, an increase of ionic strength ascribed to the
238 presence of monovalent and divalent cations (i.e., Na^+ , K^+ , Ca^{2+} , Mg^{2+}) might enhance
239 the PFAS removal due to the compression of the electrical double layer (Du et al., 2014;
240 Xiao et al., 2011). The salting-out effect should be also mentioned as reported in
241 previous studies (Carter and Farrell, 2010; Chen et al., 2016). This effect occurs at high
242 salt concentrations and it involves a decline of PFAS solubility and, consequently, their
243 adsorption on adsorbent surface could be encouraged (Du et al., 2014). It is clear that

244 the presence of inorganic anions (i.e., Cl^- and SO_4^{4-}) involves a competition with PFAS
245 for adsorption sites (Deng et al., 2010; Gao et al., 2017). However, the worsening of
246 PFAS removal occurred when solution pH was higher than point of zero charge of the
247 adsorbents, due to the establishment of electrostatic repulsions (Chen et al., 2017;
248 Zhou et al., 2009).

249 Electrostatic repulsions between anions PFAS and negatively charged surfaces can be
250 overcome through hydrophobic interactions of fluorinated chain (Zaggia et al., 2016).
251 Moreover, removal efficiency of individual PFAS into adsorbent materials depends on
252 the compound functional groups and C-F chain length. In detail, PFASs are removed
253 better than PFCAs, and long-chain compounds with the same functional group are more
254 efficiently removed than short-chain ones (Appleman et al., 2014; Du et al., 2014;
255 McCleaf et al., 2017; Xiao et al., 2011; Zaggia et al., 2016).

256 Hydrophobic effect has been found to increase with the increasing C-F chain length (Du
257 et al., 2014; McCleaf et al., 2017; Rahman et al., 2014). This was also demonstrated in
258 a prior work that investigated the competitive adsorption among PFAS with different
259 chain length on kaolinite (Xiao et al., 2011). The authors argued that, due to a stronger
260 hydrophobic interaction with the tested adsorbent, the longer-chained PFAS
261 outcompeted the shorter during adsorption. Based on PFAS functional group, the
262 presence of one more C-F bond in PFASs compared to corresponding PFCAs results in
263 stronger hydrophobic properties and increasing adsorption of PFASs (Chen et al., 2016;
264 Zaggia et al., 2016). Moreover, strong hydrophobic interactions favour the formation of
265 molecular aggregates during PFOA and PFOS removal, while highly hydrophilic
266 compounds, such as PFBA and PFBS, are removed as single molecules on adsorbent

267 active sites (Du et al., 2014). Exceeding the critical micelle concentration (c.m.c) may
268 lead to the formation of multilayer structures (i.e., micelles and hemi-micelles) on
269 adsorbent surfaces that may play an important role in PFAS adsorption (Appleman et
270 al., 2014; Deng et al., 2013; Du et al., 2014). However, at the PFAS environmental level
271 the formation of micelles is unlikely and only aggregates have been observed (Zaggia et
272 al., 2016). McCleaf et al. (2017) reported that PFAS adsorption process may include
273 two distinct phases. Initially removal occurs through PFAS adsorption on open sites of
274 adsorbents, gradually pore sites become clogged and removal becomes a function of
275 molecular aggregation of PFAS (McCleaf et al., 2017).
276 Since electrostatic negativity is originated from PFAS functional head and hydrophobic
277 interaction is linked to C-F chain, the electrostatic interactions seem to be dominant for
278 short-chain PFAS, while longer PFAS are adsorbed through hydrophobic interactions,
279 which favour the formation of molecular aggregate on the active surface (Deng et al.,
280 2012; Du et al., 2014; Zaggia et al., 2016).

281 **2.2. Scope of the review**

282 An important limitation of prior research on PFAS adsorption is that the evaluation of
283 adsorption efficiency has been explored at high PFAS concentrations, which are not
284 representative of the environmental concentration level. Furthermore, very high doses
285 of adsorbents have been used which do not reflect the actual water treatment plant
286 adsorption processes (Rahman et al., 2014).

287 Another relevant limitation of prior researches is that the experiments have often been
288 carried out by using deionized water (DI) spiked with selected PFAS, and the effect of
289 organic matter (OM) was not accounted for (Du et al., 2014). Recent studies have
290 shown that OM may not affect PFAS removal (SgROI et al., 2018a) or may positively

291 influence the PFAS adsorption (McCleaf et al., 2017). However, natural organic matter
292 (NOM) and effluent organic matter (EfOM) often play a competitive role in the
293 adsorption process (Appleman et al., 2013; Kothawala et al., 2017). Therefore, a critical
294 review of the published data is needed to better understand the OM effect on long- and
295 short-chain PFAS removal.

296 Furthermore, the regeneration of adsorbents exhausted by PFAS is not fully addressed
297 (Woodard et al., 2017). Since PFAS breakthrough is fast, adsorbent regeneration is a
298 very important and challenging task.

299 Previous published review papers deals with PFAS classification and origins (Buck et
300 al., 2011; Xiao, 2017), their occurrence and fate in aquatic environments (Ahrens, 2011;
301 Arvaniti and Stasinakis, 2015; Rahman et al., 2014) and the treatment technologies for
302 long-chain PFAS removal (Appleman et al., 2014; Kucharzyk et al., 2017). A critical
303 review concerning PFAS adsorption previously published is mainly concerned with
304 PFOS and PFOA (Du et al., 2014) and it does not include recent scientific findings on
305 OM effect as well as the performance and the regeneration of novel adsorbents.
306 Recently, the removal of short-chain PFAS by different processes, including different
307 adsorbents, has been critically reviewed by Ateia et al. (2019). However, such prior
308 review papers concerning PFAS adsorption were not focused on the role of PFAS chain
309 length, OM effect and adsorbents regeneration.

310 Therefore, the objective of this paper is to critically discuss data concerning PFAS
311 adsorption with particular focus on: 1) recent published data about adsorption capacity
312 of different materials for both long- and short-chain PFAS; 2) effect of OM on the PFAS
313 adsorption; 3) role of adsorbent regeneration.

3. Adsorption of long chain PFAS

3.1. PFOA and PFOS

A number of studies have been carried out worldwide in order to investigate long-chain PFAS removal by adsorption on different materials. PFOA and PFOS are commonly regarded as the most representative PFAS, due to their persistence in the environment and occurrence in industrial applications. A prior review paper indicated that long-chain PFAS (i.e., PFOA and PFOS) have been mainly removed through hydrophobic interactions and adsorbents with amine groups generally have high adsorption capacity (Du et al., 2014).

The adsorption of long-chain PFAS on different adsorbents (commercially available or synthesized) obtained from batch adsorption tests is discussed in the following sections. Table 2 reports the adsorption capacity obtained from published data fitting the Langmuir model along with adsorbent material type and dosage, long-chain PFAS initial concentrations and experimental setups.

3.2. Activated carbon and biochar

Among commercial adsorbents, activated carbon (AC) has been widely investigated for PFOS and PFOA removal (Liang et al., 2011; Ochoa-Herrera and Sierra-Alvarez, 2008; Rattanaoudom et al., 2012). In detail, powdered activated carbon (PAC) had higher adsorption capacity than granular activated carbon (GAC) for both PFOA and PFOS (Chen et al., 2017; Yu et al., 2009).

Results about adsorption of PFOA on bamboo-derived activated carbon (BdAC) have shown that a long time (about 34 h) was required to reach adsorption equilibrium (Du et al., 2015). The comparison between BdAC and microporous AC revealed that the adsorption capacity of BdAC was higher than that of microporous AC (Du et al., 2015;

338 Wang et al., 2016). This evidence corroborates that microporous adsorbents are not
339 suitable for removing large molecules (i.e., long-chain PFSA and PFCAs) and it is in
340 agreement with results from fixed-bed column tests, as detailed below (Appleman et al.,
341 2013; Du et al., 2014; McCleaf et al., 2017; Rattanaoudom et al., 2012; Zaggia et al.,
342 2016).

343 Chlorinated polyfluorinated ether sulfonate (F-53B) is a PFOS alternative compound.
344 Both F-53B and PFOS compounds have been detected at high concentrations in an
345 actual chrome plating WW (Du et al., 2016a). Electroplating wastewater treatment plant
346 (EWTP), employing reduction reaction (adding $\text{Na}_2\text{S}_2\text{O}_5$), precipitation (adding
347 $\text{Ca}(\text{OH})_2$), and flocculation–sedimentation (adding anionic polyacrylamide, PAM), was
348 unable to effectively remove PFOS and F-53B. Hence, batch adsorption experiments
349 were performed and a commercial coconut shell based GAC (R-CAC), after an
350 activation process at low KOH/C ratio, was selected as adsorbent material for PFOS
351 and F-53B removal (Du et al., 2016a). F-53B is more hydrophobic than PFOS, due to
352 an ether unit in the molecular structure, and this led to a much higher adsorption
353 capacity of F-53B (two-fold higher than PFOS) onto R-CAC (Table 2). Single walled
354 carbon nanotubes (SWNT) are hollow cylindrical tubes, made of graphene layers and a
355 lattice of C atoms, and they have been investigated for PFOS and PFOA removal
356 (Wang et al., 2016). The comparison of microporous AC efficiency with SWNT, in
357 removing PFOA at the same concentration, revealed that microporous AC has a higher
358 adsorption capacity than SWNT. This difference may be linked to the higher surface
359 area of microporous AC than that of SWNT (Table 2). Moreover, adsorption capacities
360 of SWNT and AC for PFOS were 6 and 2 times higher, respectively, than that of PFOA,

361 probably due to the one more C-F unit of the PFOS molecule (Wang et al., 2016). In
362 addition, SWNT exhibited higher adsorption capacity than multi walled carbon
363 nanotubes (MWNT), consistent with their higher Brunauer, Emmett and Teller (BET)
364 surface area (Chen et al., 2011; Li et al., 2011).

365 PFAS adsorption on biochar has also been investigated because of its cost-
366 effectiveness and affinity with organic compounds (Guo et al., 2017; Inyang and
367 Dickenson, 2017; X. Xiao et al., 2017).

368 Pinewood (PWC) and hardwood (HWC) biochars have been tested for PFOA removal
369 and the adsorption capacity of the biochar was lower than that of AC (Table 2). Guo et
370 al. (2017) have demonstrated that the adsorption capacity of corn-straw-derived biochar
371 for PFOS removal increased with the increase of pyrolytic temperature, which is related
372 to BET surface area of biochar (Table 2).

373 Reported data from fixed-bed column tests showed the ability of GAC to remove traces
374 of PFAS (as both complex mixture or single solute) from water intended for human
375 consumption (Appleman et al., 2014; Chularueangaksorn et al., 2014; McCleaf et al.,
376 2017; Pramanik et al., 2017, 2015; Sgroi et al., 2018a; Zaggia et al., 2016). GAC
377 (Filtrisorb 400, Calgon) column tested for PFOA removal has shown that the bed
378 volumes were about 55,000 at 90% breakthrough point (Chularueangaksorn et al.,
379 2014). The same GAC (Filtrisorb 400, Calgon) has been tested for long-chain PFCAs
380 (i.e., PFOA, PFNA, PFDA, PFUnA, PFDoDA, PFTeDA) and PFSAAs (i.e., PFHxS and
381 PFOS) removal, and the observed removal efficiencies were in the range 65-80%
382 (McCleaf et al., 2017). Information about the influence of GAC pore distribution on
383 adsorption capacity of PFOS and PFOA were reported by Zaggia et al. (2016). Results

384 from continuous adsorption experiments have demonstrated the great efficiency of
385 bituminous AC, characterized by meso- and macropores, for PFOA and PFOS removal,
386 whose maximum adsorption capacity was 39.6 and 4.1 $\mu\text{g g}^{-1}$, respectively (Zaggia et
387 al., 2016). In fact, meso- and macro-porous adsorbent materials are more easily
388 accessed by long-chain PFAS resulting in higher adsorption capacity (Kothawala et al.,
389 2017; McCleaf et al., 2017; Zaggia et al., 2016). A high dosage of GAC (Filtrisorb 400,
390 Calgon) was tested in a pre-equilibrium batch study for the removal of a complex
391 mixture of long-chain PFASs (PFHxS and PFOS) and PFCAs (PFOA, PFNA, PFDA,
392 PFUnDA, PFDODA) from LW (Kothawala et al., 2017). Regardless of OM concentration,
393 long-chain PFAS removal was in the range of 50-90% (Kothawala et al., 2017).
394 However, these results were obtained from a pre-equilibrium study (contact time was 15
395 min) with a GAC dose (10 g L^{-1}) that is not typically employed at water treatment plants.

396 **3.3. Molecularly imprinted polymers**

397 Several experimental works have explored the PFAS adsorption onto synthetically
398 modified polymers, which are adsorbents conveniently synthesized to have high affinity
399 and selectivity towards the target contaminants (Guo et al., 2018; Karoyo and Wilson,
400 2016; L. Xiao et al., 2017). The macromolecular imprinted materials (MIMs), containing
401 β -Cyclodextrin (β -CD) within a urethane framework, provide two types of binding sites.
402 One binding site is a β -CD inclusion site, the second is a non-inclusion (interstitial) site
403 of the cross-linker domains, where aggregates, micelles and other inclusion complexes
404 could be formed (Karoyo and Wilson, 2016). Formation of bilayer and multilayer
405 structures is the dominant adsorption process for both PFOA and PFOS in these
406 materials, as observed in several studies (Du et al., 2014; Gao et al., 2017; Karoyo and
407 Wilson, 2016). The modification of CD-based polymer adsorbents composition may

408 allow an increasing of PFOA removal (L. Xiao et al., 2017). Nevertheless, the removal
409 efficiency of cross-linked polymer network, where β -CD substitutes decafluorobiphenyl
410 (DFB-CDP), was comparable with that of biochar tested by Inyang and Dickenson
411 (2017). Adsorption efficiency of non-imprinted carbon microspheres (NIP-CMs) for
412 PFOS removal has been compared with a surface imprinted polymer (MIP-CMs) in
413 which double functional monomers of methacryloyloxyethyl trimethyl ammonium
414 chloride (DMC) and 2-(trifluoromethyl)acrylic acid (TFMA) have been synthesized on
415 carbon microspheres (Guo et al., 2018). The adsorption capacity was higher for MIP-
416 CMs than NIP-CMs (Table 2), due to the effect of molecularly imprinting technique in
417 enhancing PFOS removal. The molecular imprinting technique has also been employed
418 in order to prepare a novel adsorbent using chitosan crosslinked for PFOS removal and
419 its adsorption capacity was higher than that observed for non-imprinted ones (Yu et al.,
420 2008).

421 **3.4. Anion-exchange resin**

422 Several studies have indicated the suitability of the AE process for PFAS removal.
423 Otherwise, PFAS removal efficiency varied greatly among AE resins, due to their
424 properties, such as polymer matrix, functional group and porosity (Zaggia et al., 2016).
425 It was demonstrated that large pores of macroporous resin allow a faster diffusion of
426 PFOS than the gel one (Deng et al., 2010). This evidence is confirmed by comparing
427 the adsorption capacity of IRA910 (polystyrene-DVB, macroporous-type) and IRA400
428 (styrene-DVB, gel-type) for PFOS removal (Maimaiti et al., 2018; Wang et al., 2016; Yu
429 et al., 2009). Moreover, polymer matrixes led to different intraparticle diffusion and a
430 polyacrylic resin has shown higher efficiency for PFOS removal than polystyrene one
431 (Deng et al., 2010). IRA910 exhibited a good selectivity also for PFHxS as

432 demonstrated by Maimaiti et al. (2018). When IRA67 (polyacrylic-DVB, gel-type) has
433 been employed, its adsorption capacity was higher than styrene type resins (Table 2).
434 Gao et al. (2017) demonstrated the great applicability of polyacrylic resin for F-53B
435 removal, since the adsorption capacity of IRA67 (about 2400 mg g⁻¹) was higher than
436 that of R-CAC in batch experiments (Table 2).

437 As regards long-chain PFCAs, PFOA removal has been investigated on different kinds
438 of strong AE resins (Du et al., 2015; Maimaiti et al., 2018; Wang et al., 2016; Yu et al.,
439 2009). Based on Table 2, the highest adsorption capacity was obtained when IRA910
440 (polystyrene-DVB, macroporous-type resin) was employed (Maimaiti et al., 2018). The
441 adsorption capacity of IRA67 was higher than that of BdAC, both adsorbents tested at
442 the same PFOA initial concentration (Table 2). The efficiency of IRA67 may be ascribed
443 to the amine groups on the resin surface, which involve the PFOA adsorption via AE
444 mechanisms. Besides, the open pore structure of IRA67 also allowed a better diffusion
445 of PFOA molecules. Moreover, as demonstrated by Zaggia et al. (2016), the selectivity
446 of larger and more hydrophobic anions (i.e., PFOS and PFOA) increased with the
447 increasing of alkyl chain length of the functional group of AE resins. Differences in terms
448 of equilibrium exchange capacity of different type of resins are linked with the strength
449 of hydrophobic interactions between resin functional groups and PFAS molecules.
450 Considering hydrophobicity of resin, highly hydrophobic resins (such as A532E) favour
451 PFOA and PFOS passage from hydrated state to adsorbed state as confirmed by the
452 highest adsorption capacity values obtained on both batch and continuous-flow pilot
453 scale experiments (Zaggia et al., 2016). The comparison between the performance of

454 AE resin and GAC for long-chain PFAS removal has also been carried out by employing
455 the fixed-bed column tests.
456 Particularly, as demonstrated by Chularueangaksorn et al. (2014), PFA300 (AE resin,
457 polystyrene crosslinked DVB) could treat a higher amount of water than GAC (Filtrisorb
458 400, Calgon) before the saturation (at a 90% breakthrough point, bed volumes were
459 119,880 and 55,080, respectively). A532E (polystyrene-DVB gel-type, bifunctional
460 quaternary amine) exhibited the highest adsorption capacity for both PFOS and PFOA
461 among several adsorbent materials tested (i.e., coconut based GAC, bituminous coal
462 based GAC, mildly hydrophobic and non-hydrophobic resins) during continuous
463 experiments (Zaggia et al., 2016). Removal efficiencies of AE resin (A600, polystyrene-
464 DVB gel-type) for long-chain PFCAs (PFOA, PFNA, PFDA, PFUnA, PFDoDA, PFTeDA)
465 and PFSA (PFHxS and PFOS) were in the range 33-94% after 50,704 bed volumes
466 (BVs), whereas removal efficiencies of GAC (Filtrisorb 400, Calgon) column were in the
467 range 28-94% after 49,523 BVs (McCleaf et al., 2017).

468 **3.5. Synthetized materials**

469 With the purpose of identifying the best suitable adsorbent for PFAS removal, it is
470 currently practice to synthesize new materials with properties and structures easy to
471 control and modify. To date, new adsorbents with higher selectivity and specificity
472 designed towards PFAS removal are also becoming commercially available for water
473 treatment applications. For instance, a porous aromatic framework constructed from
474 benzene rings (PAF-45) was employed for PFOS removal (Luo et al., 2016). Despite
475 the remarkable adsorption capacity (Table 2), PAF-45 feasibility was influenced by pH
476 solution and ionic strength since the adsorption capacity was higher at acidic pH values
477 (i.e., pH ~ 3). Moreover, the adsorption isotherm experiments have been carried out at

478 PFOS concentrations in the range of 50-200 mg L⁻¹ (Luo et al., 2016), which are much
479 higher than the PFAS concentrations typically detected in the water environment
480 (Rahman et al., 2014).

481 Covalent triazine-based framework (CTF) has been investigated for removing different
482 long-chain compounds (PFOA, PFHxS and PFOS), due to its large surface area and
483 rigid pore structure (Wang et al., 2016). Among the three long-chain PFAS tested,
484 Wang et al. (2016) have demonstrated that the adsorption capacity of CTF was highest
485 for PFOS (Table 2). The electrostatic interaction between PFAS anion head and triazine
486 groups of CTF (positively charged) represents the main driving force for PFAS
487 adsorption on CTF. Based on electrostatic interactions, which are the main adsorption
488 mechanism of short-chain PFAS, CTF exhibited a good adsorption capacity for PFBS
489 and PFBA removal, as it will be discussed below.

490 Boron nitride, characterized by lightness structure and thermal stability, has been
491 synthesized as porous hexagonal boron nitride nanosheets (h-BNs) for PFOS and
492 PFDA removal (Feng et al., 2016). The efficiency of this adsorbent material has been
493 demonstrated to be linked with chain length and functional group of PFAS, since PFDA
494 was more favourable adsorbed than PFOS (adsorption capacity of 0.16 mg m⁻² and
495 0.04 mg m⁻², respectively). Among novel adsorbent materials investigated, magnetic-
496 nanoparticles attached into fluorographene (2-MNPs@FG, mass ratio of MNPs and FG
497 is 3:5) exhibited the highest adsorption capacity for PFOS instead of PFOA (Table 2).
498 However, adsorbent dosage was not coherent with that really employed in full-scale
499 treatment plants (Wang et al., 2018).

500 Quaternized cotton and aminated rice husk (ARH) both prepared with atom transfer
501 radical polymerization (ATRP) technique were tested for PFOS and PFOA removal
502 (Deng et al., 2013, 2012). Results have demonstrated that quaternized cotton had
503 higher adsorption capacity than ARH for both PFOS and PFOA (Table 2).

504 Polyacrylonitrile fiber (PANF)-derived activated carbon fibers (PACFs) showed a
505 remarkable adsorption capacity for PFOS and PFOA (Chen et al., 2017). The
506 adsorption affinity could be ascribed to its high specific surface area and micro- and
507 mesoporous structure which allowed the establishment of multilayer adsorption
508 (formation of hemi-micelles and micelles). The latter was also the main adsorption
509 mechanism on crosslinked chitosan beads employed for PFOS removal, as
510 demonstrated by Zhang et al. (2011).

511 Poly(ethylenimine)-functionalized cellulose microcrystals (PEI-f-CMC) have been tested
512 for removing long-chain PFAS at environmentally relevant concentration level (Ateia et
513 al., 2018). Long-chain PFAS removal percentage was in the range of 80-98%. Despite
514 its low surface area, PEI-f-CMC exhibited a good adsorption affinity both in DI and in the
515 presence of co-existing OM, as discussed in section 5.

516 The main drawback of those synthesized materials is the application at full-scale
517 treatment plants because their production is not industrialized and, probably, they are
518 not commercially available, unlike AC and AE resins. Economic assessment for new
519 synthesized materials is needed.

520 **3.6. Nanoparticles materials**

521 The adsorption of PFAS into nanoparticles (NPs) has recently been investigated due to
522 their co-existing in aquatic environments (i.e., marine debris plastic) or their high surface
523 area and reactivity (i.e., iron oxides). As demonstrated by Lu et al. (2016), nano-oxides

524 such as titania (TiO_2), iron oxides (Fe_2O_3), alumina (Al_2O_3) and silica (SiO_2) have the
525 ability to sorb PFOS. Among the different nano-oxides tested, Al_2O_3 showed the highest
526 PFOS adsorption capacity ($1.1 \mu\text{g m}^{-2}$), whereas SiO_2 exhibited the lower adsorption
527 capacity ($0.1 \mu\text{g m}^{-2}$). This difference in terms of PFOS removal should be ascribed to
528 the different hydroxyl groups and surface area. Furthermore, the main adsorption
529 mechanism of PFOS on nano-oxides was not only electrostatic interaction, but also the
530 formation of hydrogen bonds between the PFOS sulfonic terminal group and nano-
531 oxides surface (Lu et al., 2016).

532 Magnetite nanoparticles (Fe_3O_4 NP) were used as the substrate of a novel magnetic
533 nanocomposite ($\text{Fe}_3\text{O}_4@ \text{SiO}_2\text{-NH}_2\&\text{F}_{13}$), in which silica membrane functionalized with
534 amino group and octyl-perfluorinated chain was synthesized on the Fe_3O_4 NP surface
535 (Zhou et al., 2016). Based on kinetic data, the predominant adsorption mechanism
536 between the magnetic adsorbent and PFAS tested was chemisorption, which includes
537 both electrostatic and F-F interaction (Zhou et al., 2016). In detail, the selective F-F
538 interaction between octyl-perfluorinated chain on the magnetic composite surface and
539 perfluoroalkyl dipole shell in PFAS molecule was enhanced by electrostatic attraction of
540 -NH_2 and the anionic -COOH or $\text{-SO}_3\text{H}$ of PFCAs and PFSA molecules, respectively.
541 Due to size exclusion effect, PFCAs were better adsorbed than PFSA on $\text{Fe}_3\text{O}_4@ \text{SiO}_2\text{-}$
542 $\text{NH}_2\&\text{F}_{13}$. Considering perfluoroalkyl chain length, PFUnDA exhibited the highest
543 adsorption capacity onto $\text{Fe}_3\text{O}_4@ \text{SiO}_2\text{-NH}_2\&\text{F}_{13}$, due to the formation of aggregate
544 structure between long-chain PFAS which could stick together and, consequently, the
545 amount of PFAS adsorbed increased (Zhou et al., 2016).

546 A recent experimental work investigated the capability of microplastics (MPLs) to sorb a
547 complex mixture of PFAS from RW (Ebro river) and seawater (SeaW), simulating
548 realistic environmental conditions (Llorca et al., 2018). High-density polyethylene
549 (HDPE), polystyrene (PS) and poly-styrene carboxylic acid (PS-COOH) were selected
550 due to their wide occurrence as marine debris plastic. Results of batch adsorption
551 experiments showed low removal percentages (< 27%) of long-chain PFAS in RW after
552 7 days (with few exceptions of PFTrA and PFTeA whose percentages were between
553 65% and 70%). Moreover, MPLs efficiency in SeaW was affected by water chemistry
554 such as salinity and OM concentration. This point will be deeply explained later (see
555 section 5). Overall, carboxylated compounds with more than 11 C atoms were the most
556 removed (Llorca et al., 2018). In detail, HDPE exhibited the worst adsorption capacity
557 due to its granular shape, which limited the intraparticle diffusion (Llorca et al., 2018). In
558 RW, PFOA, PFNA, PFHxS and PFOS did not present any adsorption on both HDPE
559 and PS-COOH throughout the experiment period. On the contrary, affinity between
560 carboxylated compounds with less than 10 C atoms and HDPE was higher in SeaW.
561 Adsorption rates of PFCAs on PS-COOH in RW increased with the increasing of chain
562 length. Comparing PFCAs to PFSA, with the same chain length, the carboxylated
563 compounds were less removed (Llorca et al., 2018). Whereas, in SeaW, the presence
564 of carboxylic group in the PS-COOH surface implicated an increase of adsorption rates
565 of carboxylated compounds. Adsorption kinetics were influenced by adsorbents size,
566 therefore PS exhibited faster kinetic than HDPE, since it had the smallest particles
567 (Llorca et al., 2018).

4. Adsorption of short chain PFAS

4.1. Challenges in short chain PFAS adsorption

Short-chain PFAS and their precursors are as persistent as those long-chained. Therefore, although they show less bioaccumulation potential, their environmental occurrence is of concern (Ateia et al., 2019; Brendel et al., 2018; Zhao et al., 2016). While long-chain PFAS are binding with particles present in solution, those short-chain are mainly in the dissolved phase resulting in long-range transport in aquatic environments (Ahrens, 2011). Therefore, short-chain PFAS have higher mobility in water bodies than long-chain compounds. Specifically, short-chain compounds could significantly affect drinking water supplies, increasing the human exposure to PFAS compounds. Considering the properties of short-chain PFAS (Table 1), it is expected that the large-scale remediation treatments, currently suitable for long-chain PFAS, are ineffective for short-chain PFAS (Ateia et al., 2019; Brendel et al., 2018). This evidence is consistent with data from full-scale drinking water treatment plants (Arvaniti and Stasinakis, 2015; Rahman et al., 2014). Moreover, it is widely reported that carbon-chain length influences adsorption capacities of PFAS onto various materials, since adsorption rate seems to increase with the increase of molecular size (Appleman et al., 2014).

Data collected from full-scale treatment plants (drinking and wastewater facilities) suggested that the removal of short-chain compounds is still a challenge task, due to their early and faster breakthrough (Arvaniti and Stasinakis, 2015; McCleaf et al., 2017; Zaggia et al., 2016). To date, a restricted number of published data have addressed short-chain PFAS adsorption by using different materials, because most of the experimental works have focused on the removal of long-chain PFAS (i.e., PFOA and

592 PFOS). Adsorption capacities obtained by fitting the Langmuir model along with type
593 and dosage of adsorbent materials, initial concentration of short-chain PFAS and
594 experimental setup are reported in Table 3. Comparisons between several adsorbent
595 materials tested for short-chain PFAS are reviewed based on both batch experiment
596 studies (Table 3) and fixed-bed column tests.

597 The adsorption capacity of materials tested for PFBA removal observed during batch
598 tests (Table 3) follows the order: microporous AC < CTF < IRA910 (strong AE resin).
599 However, results obtained by continuous experiments have demonstrated that the
600 adsorption capacity of meso-porous GAC (meso-porous bituminous coal-derived AC)
601 was higher than A532E (strong AE resin) (4.3 and $3.3 \mu\text{g g}^{-1}$, respectively) (Zaggia et
602 al., 2016). In the following sections the removal of short chain PFAS by adsorption is
603 discussed based on the type of adsorbent.

604 **4.2. Activated carbon, biochar and other adsorbents**

605 The performance of BdAC and coal-based AC (microporous type), used for removing
606 PFHxA, has been compared. The adsorption capacity of BdAC was 13 times lower than
607 microporous AC probably due to its low BET surface area and the presence of OM in
608 the solution tested (Du et al., 2015; Wang et al., 2016). The adsorption capacities of
609 HWC and PWC biochars for PFBA and PFOA removal were investigated by Inyang and
610 Dickenson (2017). Results from adsorption kinetic tests showed that the amount of
611 PFBA adsorbed on both HWC and PWC was 3-4 times lower than that of PFOA. This
612 evidence suggests the incapability of biochar for short-chain PFAS removal and it may
613 prove the competition between long- and short-chain PFAS for the adsorption sites
614 and/or competition with OM (Inyang and Dickenson, 2017).

615 Short-chain PFAS adsorption on GAC and PAC has also been investigated by using
616 batch test (Hansen et al., 2010). Tests have been carried out at real concentrations of
617 short-chain PFAS (range about 73 – 320 ng L⁻¹) and using an adsorbent dose
618 comparable to that used in real water treatment plants (range about 25 – 125 mg L⁻¹).
619 Results have demonstrated that PAC has higher adsorption capacity than GAC (more
620 than twice), due to the shorter internal diffusion distances and its higher BET surface
621 area (Hansen et al., 2010).

622 MWCNTs functionalized in different ways (MWCNTs-PRI, MWCNTs-COOH, MWCNTs-
623 OH) were tested for PFBS removal (Deng et al., 2015a). Removal percentage of PFBS
624 on MWCNTs was about 30% in DI water, whereas when humic acids (HA) and phenol
625 co-existed in the solution, this percentage was lower than 20% (Deng et al., 2015a).
626 Moreover, due to the weak hydrophobicity of PFBS, its percentage removal was lower
627 than that of long-chain compounds (Deng et al., 2015a). Results of adsorption isotherm
628 of PFOS and PFBS on boehmite showed that PFOS was better removed than PFBS,
629 since the effect of surface aggregation enhanced long-chain PFAS removal (Wang et
630 al., 2015).

631 Among fixed-bed column experiments carried out in order to assert the short-chain
632 PFAS removal, McCleaf et al. (2017) reported faster breakthrough and lower removal
633 percentage of short-chain PFCAs on GAC column (F400, Calgon) than long-chain ones.
634 The removal efficiencies of the short-chain PFCAs tested (i.e., PFBA, PFPeA, PFHxA
635 and PFHpA) were lower than 19% on GAC column after 49,523 BVs. Whereas, the
636 removal efficiency of PFBS (short-chained PFASs tested) was ~10%. Those results
637 suggest that short-chain PFAS removal depends mainly on the availability of active

638 sites, since they do not bind with other particles and, consequently, the formation of
639 aggregates does not occur (McCleaf et al., 2017; Zaggia et al., 2016). Moreover,
640 desorption of short-chain PFAS has been also hypothesized, due to the competition of
641 long-chain PFAS and/or OM. Similar fast breakthrough of short-chain PFAS has also
642 been observed in rapid small-scale column tests (RSSCT) carried out on different types
643 of GAC columns (Appleman et al., 2013). Among the three activated carbons tested,
644 F300 (microporous bituminous coal-derived) exhibited the lowest efficiency for the
645 short-chain PFCAs tested (i.e., PFBA, PFPeA and PFHxA), due to its pores distribution
646 (mainly microporous type) and BET surface area. Others GAC tested, coconut based
647 GAC (AquaCarb 1240) and bituminous coal based ones (F600), performed better for
648 the removal of selected short-chain PFAS (Appleman et al., 2013).

649 PFBA uptake on different types of biochar was influenced by BET surface area. Indeed,
650 the highest percentage removal has been found on the hardwood sawdust pellets
651 gasified at 900 °C (HWC), which had the highest BET surface area among the different
652 biochars tested (Inyang and Dickenson, 2017). However, results obtained from batch
653 adsorption kinetic tests have demonstrated that GAC removed PFBA better than HWC
654 (Inyang and Dickenson, 2017).

655 Short-chain PFAS adsorption on microplastics (HDPE, PS and PS-COOH) has recently
656 been investigated in simulated realistic environmental conditions (Llorca et al., 2018).
657 Due to the increase in ionic strength, short-chain carboxylates compounds have been
658 better removed by PS-COOH in SeaW than in RW. Whereas, short chain PFAS were
659 almost not adsorbed onto PS and HDPE (Llorca et al., 2018).

660 Removal of PFBS (short-chain PFASs) has been investigated at batch mode by means
661 of AC (i.e., microporous AC and Calgon F400) and CTF (Ochoa-Herrera and Sierra-
662 Alvarez, 2008; Wang et al., 2016). The difference of adsorption capacity between CTF
663 and AC may be related to their different structure and, consequently, adsorption
664 mechanisms established, since the electrostatic interactions between PFAS anion head
665 and triazine groups of CTF involved short-chain PFAS adsorption on CTF (Wang et al.,
666 2016). However, CTF exhibited a lower affinity with PFBA than ARH (Table 3).
667 Adsorption capacity of ARH was 4 times higher than CTF (Deng et al., 2013; Wang et
668 al., 2016).

669 A novel adsorbent poly(ethylenimine)-functionalized cellulose microcrystal (PEI-f-CMC)
670 has been synthesized and its efficiency for short-chain removal has been evaluated at
671 environmental relevant concentration levels (Ateia et al., 2018). Results have
672 demonstrated that short-chain with sulfonate terminal group were removed better than
673 short-chain PFCAs, in agreement with previous experimental studies (McCleaf et al.,
674 2017; Rahman et al., 2014).

675 **4.3. Anion-exchange**

676 The removal efficiency of three PFCAs owning different chain lengths, including PFHxA
677 (with 6 C atoms), PFHpA (with 7 C atoms) and PFOA (with 8 C atoms) from industrial
678 WW (perfluorooctanesulfonyl fluoride washing wastewater, PFSOF WWW) by AE resin
679 has been evaluated during batch experiments conducted by Du et al. (2015). Results
680 demonstrated that adsorption sites on IRA67 were occupied by PFOA molecules prior
681 to the others PFCAs and, therefore, the adsorption rates of short-chain were lower.
682 Consequently, short-chain PFCAs reached the equilibrium very fast (Du et al., 2015).
683 Moreover, comparing the efficiency of two different anion-exchange resins (i.e., IRA910

684 and IRA67) for removing the same compound (PFHxA), IRA910 (polystyrene-DVB
685 macroporous-type) exhibited higher adsorption capacity than IRA67 (polyacrylic-DVB
686 gel-type) (Du et al., 2015; Maimaiti et al., 2018). This difference in adsorption capacity
687 for PFHxA may be linked to the resin properties, such as polymer matrix and porosity,
688 and/or to the effect of OM present in the solution. In fact, IRA910 was tested in DI water,
689 whereas IRA67 was employed for PFHxA removal from PFSOF WW (Du et al., 2015;
690 Maimaiti et al., 2018).

691 Moreover, when tested for the same actual WW, IRA67 had higher affinity with PFHpA
692 than BdAC, as demonstrated by the adsorption capacity, which was 3 times higher than
693 that of BdAC (Du et al., 2015). Furthermore, amine groups on adsorbent surface
694 improved the short-chain PFAS removal, due to the establishment of electrostatic
695 interactions, which are the main driving force during short-chain adsorption (Deng et al.,
696 2013; Du et al., 2014; Zhang et al., 2011). Results provided by fixed-bed column tests
697 demonstrated that the adsorption capacity of A532E (strong AE resin) was almost equal
698 to that of GAC (meso-porous bituminous-coal derived AC), when tested for PFBS
699 removal ($4.7 \mu\text{g g}^{-1}$ and $4.1 \mu\text{g g}^{-1}$, respectively) (Zaggia et al., 2016).

700 Results from fixed-bed column experiments carried out by McCleaf et al. (2017) have
701 shown fast breakthrough and low removal percentage for short-chain PFCAs on AE
702 resin (A600, polystyrene-DVB gel-type). The removal efficiencies of short-chained
703 PFCAs tested were ~11% on A600 column after 50,704 BVs. The same anion-
704 exchange resin better removed PFBS (short-chain PFASs tested) than other short-chain
705 compounds, and the removal efficiency of PFBS was ~55% (McCleaf et al., 2017).

706 Results from a pre-equilibrium study carried out by Kothawala et al. (2017)
707 demonstrated that fresh adsorbent materials (i.e., AE resin, Purolite A600, and GAC,
708 Filtrasorb 400) could remove short-chain PFAS from water solutions. Nevertheless,
709 anion-exchange showed the highest percent of removal. In detail, removal percentages
710 of short-chain PFAS (PFBA, PFPeA, PFHxA, PFHpA, PFBS) were about 90% and 50%
711 on A600 and Filtrasorb 400, respectively. However, those results have been obtained
712 from a pre-equilibrium study (contact time was equal to 15 min) with a high dosage of
713 adsorbent materials (Kothawala et al., 2017).

714 Analysis of transmission electron microscopy (TEM) carried out on AE resins allowed a
715 deep understanding of the predominant adsorption mechanism involved during short-
716 chain PFAS removal (Zaggia et al., 2016). Indeed, TEM images showed that the weak
717 hydrophobic interactions between PFBA (or PFBS) and resin surface did not allow the
718 formation of aggregates and, consequently, AE of single molecules was the main
719 mechanism for short-chain removal.

720 Overall, AE resins exhibit a remarkable adsorption capacity for both long- and short-
721 chain PFAS. Nevertheless, the possible release of disinfection by-products (DBP) such
722 as N-Nitrosodimethylamine (NDMA) may represent the main drawback for their full-

723 scale application (SgROI et al., 2018b). Particularly, the amount of NDMA and their
724 precursors released upon usage depends on the resin functional group (such as amine
725 groups) and operational conditions (such as regeneration activities and flow
726 interruptions).

727 **4.4. Comparison between long- and short-chain PFAS** 728 **adsorption**

729 In order to provide a deeper analysis on the long- and short-chain PFAS removal by
730 adsorption, data from Table 2 and Table 3 have been elaborated to build a box-and-
731 whiskers plot (Figure 1) that allows the comparison between the removals of long- and
732 short-chain PFAS by different adsorbents (classified as activated carbon, resin, and
733 synthesized materials). Figure 1 shows that the adsorption capacity of a wide range of
734 adsorbents is lower for short-chain PFAS compared to that observed for long-chain
735 PFAS. This result emphasizes that the removal of short-chain PFAS by adsorption is
736 very challenging and it will need more research in the future. A comparison in
737 adsorption capacity of the different adsorbents employed have shown that resins are
738 more effective than activated carbons for both long- and short-chain PFAS removal.
739 Activated carbon exhibited the lowest adsorption capacity. A significant variation in
740 adsorption capacity of synthesized materials used for long-chain removal was observed.
741 Such variation ranged from the lowest to the highest adsorption capacity observed for
742 long-chain PFAS removal. However, the latter best results were often obtained at acid
743 pH. For instance, the best adsorption capacity (5847 mg g^{-1}) was found by using the
744 PAF-45 adsorbent at pH 3. Therefore, the real operating conditions may affect the
745 removal efficiency. For instance, the effect OM may significantly influence the
746 performance of adsorbents, as discussed in the next section.

747 **5. Effect of organic matter (OM) on PFAS adsorption**

748 **5.1. PFAS adsorption in presence of OM**

749 OM is ubiquitous in the aquatic environment and it can be distinguished between NOM,
750 regarded as OM naturally present in surface water, and EfOM, the OM released from
751 WWTPs (Shon et al., 2006).

752 OM could affect adsorption efficiency of adsorbent materials due to binding and
753 adsorption interactions with PFAS, including long- and short-chain compounds. As
754 illustrated by Du et al. (2014b), NOM could interact with PFAS via electrostatic and/or
755 hydrophobic interactions. Firstly, OM is mostly constituted of anionic species and, after
756 adsorbing on materials, it can lead to repulsive electrostatic interactions. Meanwhile,
757 NOM can also lead to PFAS adsorption via hydrophobic interaction between
758 perfluoroalkyl tail and NOM adsorbed on the adsorbent surfaces. To date, it is not well
759 understood how these effects co-exist and which is predominant.

760 Most of undertaken experimental works investigated adsorption properties of PFAS on
761 different adsorbent materials in DI (Deng et al., 2012; Guo et al., 2018; Maimaiti et al.,
762 2018; Yu et al., 2009). Many researches have selected fulvic and humic acids (FA and
763 HA) as the model DOM in order to evaluate the impact of DOM on PFAS adsorption
764 (Deng et al., 2015a; Du et al., 2016b; Maimaiti et al., 2018; Pramanik et al., 2015; F.
765 Wang et al., 2015; L. Xiao et al., 2017). Actually, NOM consists of a wide range of
766 organic compounds, such as hydrophilic acids, proteins, carboxylic and amino acids in
767 addition to the already mentioned HA and FA. When using a synthetic solution of OM
768 during experimental studies, some interactions between PFAS and water matrix have
769 not been described in detail. Table 4 reports the main experimental works published
770 related to the adsorption of PFAS (both long- and short-chain compounds) on different

771 adsorbent materials in the presence of OM. However, some of those studies lacked a
772 detailed discussion on the effect of OM on PFAS adsorption.

773 Most of these experimental studies (Appleman et al., 2013; Du et al., 2016b, 2016a; Y.
774 Wang et al., 2015; L. Xiao et al., 2017) have investigated the effect of OM on PFAS
775 adsorption through comparison between synthetic OM solution and OM free solution
776 (Table 4). Only few experimental works compared the effect of different DOM sources
777 on PFAS adsorption and tested water had different dissolved organic carbon (DOC)
778 concentrations (Ateia et al., 2018; Inyang and Dickenson, 2017; Kothawala et al., 2017;
779 Sgroi et al., 2018a; Wang et al., 2018) . Furthermore, the simultaneous effect of ionic
780 strength and OM on the PFAS adsorption is very complex. For instance, it was
781 demonstrated that the presence of monovalent and divalent cations increases PFAS
782 adsorption on different materials, i.e. sediments (Higgins and Luthy, 2006), activated
783 carbon (Du et al., 2015), synthesized material (PAF-45) (Luo et al. 2016), resin (Du et
784 al., 2015) and kaolinite (Xiao et al., 2011). Particularly, the adsorption of short-chain
785 PFAS (i.e., PFHxA and PFBA) on kaolinite was more thermodynamically favorable at a
786 higher Na^+ concentration due to the compression of the electrical double layer and
787 ensuing reduced electrostatic repulsion between PFAS and kaolinite surface (Xiao et al.,
788 2011). In addition, divalent cations (such as Ca^{2+} and Mg^{2+}) could also improve PFAS
789 removal by divalent cation-bridging effect (Du et al., 2015, 2014). On the contrary, the
790 presence of inorganic anion negatively affects the adsorption of PFAS due to the
791 competition for adsorption sites (Deng et al., 2010). Whereas, it was observed that the
792 presence of ionic OM (i.e., HA, SDS, FA) compete with both long- (F. Wang et al., 2015;
793 Zhao et al., 2011) and short-chain PFAS (Maimaiti et al., 2018; Zhao et al., 2011).

794 Specifically, such effect seems more relevant for short-chain PFAS due to the
795 competition for active sites (Maimaiti et al., 2018; McCleaf et al., 2017; Zhao et al.,
796 2011). On the other hand, hydrophobic DOM was found to improve the PFAS retention
797 in GAC adsorption, while hydrophilic DOM seems do not affect significantly PFAS
798 adsorption in both GAC and AE adsorption experiments (Kothawala et al., 2017). The
799 positive impact of DOM on the adsorption of long-chain PFAS has been ascribed to the
800 formation of PFAS aggregate or DOM-PFAS complexes (Kothawala et al., 2017;
801 McCleaf et al., 2017) but such bonds have been rarely demonstrated. Consequently,
802 the impact of DOM and ionic strength on the PFAS adsorption need more investigation.
803 A systematic review concerning the effect of OM on PFAS adsorption for different
804 classes of adsorbents is reported in the following sections.

805 **5.2. Activated carbon**

806 Among adsorbent materials, AC has proven to be effective for PFAS removal and its
807 efficiency in presence of OM has been investigated both in batch and column
808 experiments (Table 4). Contrasting trends have been observed in experimental studies
809 because of the influence of co-existing OM. Indeed, the performance of PFAS
810 adsorption onto AC was found negatively impacted, enhanced or not influenced by the
811 presence of OM. The main interactions that could be established between OM
812 molecules and PFAS (both long- and short-chain) during adsorption on GAC column are
813 illustrated in Figure 2. In detail, hydrophobic interactions mainly concern long-chained
814 C-F molecules, while short-chain PFAS interact with OM molecules through pore
815 blockage and electrostatic repulsions.

816 In many cases, activated carbon performance was negatively influenced by co-existing
817 OM and its efficiency decreased with the increase of DOC concentrations (Du et al.,

818 2016a, 2015; Hansen et al., 2010; Kothawala et al., 2017; Pramanik et al., 2017, 2015;
819 Y. Wang et al., 2015; Yu et al., 2012; Zhao et al., 2011). On the contrary, adsorption
820 capacity on GAC did not significantly decrease in the presence of GW NOM probably
821 due to the high concentration of PFAS tested (50 mg L^{-1}) and to the effect of ionic
822 strength (Zhao et al., 2011). Results from the previously mentioned study demonstrated
823 that PFBA removal was affected by OM since the results from adsorption equilibrium
824 suggested a reduction of 40% (Zhao et al., 2011). Results from adsorption kinetic
825 experiments provided by Yu et al. (2012) suggested that adsorption equilibrium for
826 PFOA and PFOS on PAC was reached at 4 h and 24 h in the absence and in the
827 presence of EfOM, respectively (Yu et al., 2012). The competition between PFAS and
828 DOM for AC adsorption sites has also been investigated in RSSCTs (Appleman et al.,
829 2013; McCleaf et al., 2017; Pramanik et al., 2017, 2015; Sgroi et al., 2018a). Results
830 obtained from the latter mentioned experimental works are in disagreement about the
831 effect of OM. This discrepancy may be related to the different experimental setups, such
832 as PFAS initial concentration, source and concentration of DOM, empty bed contact
833 time (EBCT), and to the kind of AC tested. As suggested by Pramanik et al (2015),
834 when DOC concentration increased from 5 to 25 mg L^{-1} the removal efficiency of PFOA
835 and PFOS on PAC decreased by 50% and 35%, respectively. On the contrary, the
836 performance of bituminous coal-based GAC was less influenced by variation of DOC
837 concentration (Pramanik et al., 2015). Similar results have asserted that proteins
838 resulted in a low retention of PFOA and PFOS through hydrophobic and electrostatic
839 interactions (Pramanik et al., 2017). Moreover, breakthrough $> 20\%$ has been reached
840 at 125,000 BVs during DOM-free experiments, and at $\sim 11,000$ BVs when DOC

841 concentration was equal to 1.7 mg L^{-1} . This observation highlights that faster
842 breakthrough of PFAS takes place during RSSCT experiments in presence of DOM
843 (Appleman et al., 2013). Nevertheless, Sgroi et al. (2018a) found that breakthrough
844 curves of some long-chain PFAS (i.e., PFOS, PFOA and PFDA) were similar in column
845 beds packed tests by using Norit Darco 12x40. In those experiments, waters with
846 different DOM concentrations and compositions fed the packed columns (Table 4). The
847 observed behaviour for the investigated long-chain compounds may be ascribed to the
848 hydrophobic interactions occurring between long-chain PFAS and GAC. Indeed,
849 hydrophobic interactions are the main adsorption mechanisms for long-chain PFAS, and
850 the hydrophobic interactions may be only slightly influenced by OM presence.

851 Results of pre-equilibrium study by Kothawala et al. (2017) are in contrast with other
852 findings (Du et al., 2016a; Wang et al., 2016; Yu et al., 2012; Zhao et al., 2011), since
853 the increase in DOC concentration seems to enhance long-chain PFAS removal due to
854 the formation of DOM-PFAS complex and, consequently, their co-adsorption. In detail,
855 removal efficiency of PFAS with a fluorocarbon-chain length of 8 C atoms increased by
856 50% when DOC concentration of a hydrophobic DOM passed from 0 to 8 mg L^{-1} using
857 GAC (Kothawala et al., 2017). It should be highlighted that these results were obtained
858 in pre-equilibrium conditions (contact time about 15 min), at PFAS initial concentration
859 of about $2.5 \text{ } \mu\text{g L}^{-1}$ and at batch dose of GAC of about 10 g L^{-1} , which is much higher
860 than doses typically used in full-scale treatment plants (Ziska et al., 2016). Performance
861 of BdAC for PFCAs removal from PFSOF WWW has been investigated in the presence
862 of high total organic carbon (TOC) concentration. Co-existing OM competed with
863 PFCAs for adsorption sites on the BdAC (Du et al., 2015). The same achievement was

864 obtained using R-CAC for PFOS and its alternative compound, F-53B, removal from
865 electrochemical WWTP effluent (Du et al., 2016a). The high concentration of TOC (78.3
866 mg L⁻¹) was linked to the high amount of hydrocarbon surfactants which were
867 unfavourable for the adsorption of PFOS and F-53B on R-CAC, since adsorption sites
868 were mainly occupied by co-existing organic compounds (Table 4). Depending on OM
869 concentration and source, dose and kind of AC employed (powdered or granular, and
870 its porosity), and PFAS tested (long- or short-chain), the effect of OM is different, due to
871 adsorption mechanisms established. The presence of OM can lead to the enhancement
872 of PFAS adsorption through the formation of agglomerations between long-chain PFAS
873 and OM. Otherwise, OM can negatively influence the adsorption efficiency, due to the
874 reducing of accessible binding sites and pore spaces (i.e., pore blockage) (Figure 2).

875 **5.3. Anion-exchange resin**

876 Main interactions occurring between OM and PFAS molecules during AE process are
877 reported in Figure 3. As previously asserted about the influence of OM on AC
878 performance, the effect of co-existing OM on AE resins for PFAS removal depends on
879 several factors, including OM composition and concentration, kind of resin and PFAS
880 concentration tested.

881 To date, the competition between OM and PFAS on AE resins is not fully understood.
882 This evidence is confirmed by several experimental studies carried out both on batch
883 and column setups (Du et al., 2015; Kothawala et al., 2017; Maimaiti et al., 2018;
884 McCleaf et al., 2017; Wang et al., 2016; Zaggia et al., 2016). Organic compounds, such
885 as HA, tri-chloroethylene (TCE), methylbenzene (MB) and sodium dodecyl sulphate
886 (SDS), have been selected to evaluate their effect on PFHxS removal on AE resin (i.e.,
887 IRA910, a macroporous polystyrene resin with a dimethyl ethanol ammonium functional

888 group) during batch experiments (Maimaiti et al., 2018). Results suggested that PFHxS
889 retention on IRA910 was not affected by non-ionic co-existing organic compounds (i.e.,
890 MB and TCE), regardless of their concentration in the solution. Whereas, IRA910
891 efficiency decreased with the increasing of HA and SDS concentrations, due to the
892 competition for adsorption sites. A slight effect on PFAS retention has been highlighted
893 under pre-equilibrium condition since AE resin (A600, polystyrene-DVB gel-type with
894 quaternary ammonium functional group) performance was independent from DOM
895 concentration and source (Kothawala et al., 2017). In detail, PFAS removal has been
896 reduced by 10% at the highest DOM concentration tested. This result is consistent with
897 data reported by previous experimental works which have demonstrated that OM
898 weakly interferes with PFAS removal during AE process (Du et al., 2015; Wang et al.,
899 2016). Resins gel-type polymer matrixes, such as IRA400 and IRA67, showed a
900 remarkable affinity with PFAS (both long- and short-chain), which was independent of
901 OM present. In an opposed way, the performance of macroporous resins (i.e., IRA910)
902 could be affected by background DOM levels.

903 **5.4. Other adsorbents (biochar, multi-walled carbon nanotubes,** 904 **mineral materials, nanoparticles)**

905 Effect of co-existing HA on boehmite surface was investigated during PFOS and PFBS
906 batch adsorption experiments (F. Wang et al., 2015). Due to the electrostatic
907 interactions between PFAS and boehmite, strong competition between PFOS (or PFBS)
908 and HA occurred. Electrostatic repulsions increased with the increasing of HA
909 accumulation on boehmite surface (F. Wang et al., 2015). The competition between
910 PFAS and synthetic OM (HA, phenol, 1-naphthol and benzoic acids) has also been
911 investigated on MWCNTs (Deng et al., 2015a). The pronounced influence on PFOS

912 removal was provided by HA, whereas phenol had a lesser influence. Moreover, PFBS
913 removal was the worst in the presence of HA or phenol (Deng et al., 2015a).

914 As demonstrated by Inyang and Dickenson (2017), HWC and PWC biochar
915 performances were affected by OM, since adsorption capacities decreased with the
916 increase of DOC concentration. In particular, OM molecules could lead to pore blockage
917 and may occupy high-energy pore sites. The adsorption capacities were about 41.3 and
918 27.7 mg g⁻¹ when LW (DOC = 2 mg L⁻¹) and tertiary treated WW effluent (DOC = 4.9 mg
919 L⁻¹) were tested, respectively (Inyang and Dickenson, 2017). Results from pilot-scale
920 tests have demonstrated that the breakthrough of some PFAS (i.e., PFPeA, PFHxA,
921 PFOA and PFOS) was faster on biochar HWC filter than on GAC column, since OM
922 adsorbed on biochar surface caused changes on biochar surface charge.
923 Consequently, the electrostatic repulsions between PFAS and biochar surface
924 increased. Adsorption kinetic tests were performed for both PFBA and PFOS, and the
925 observed slow kinetics could have been caused by pre-loaded OM on biochar (Inyang
926 and Dickenson, 2017).

927 The efficiency of novel magnetic adsorbent, 2-MNPs@FG, has been investigated for
928 PFOA and PFOS removal from different water matrixes (Wang et al., 2018). Its
929 performance was independent of OM content also at the highest DOC concentration
930 tested (1.7 mg L⁻¹). Indeed, the removal efficiencies were always found around 99% for
931 both PFOA and PFOS in all performed experiments. However, it should be noted that
932 those efficiencies have probably been obtained because of a high dosage of adsorbent
933 employed (400 mg L⁻¹). The competition between NOM (both SeaW and RW) and PFAS
934 (both long- and short-chain compounds) has also been investigated during PFAS

935 adsorption onto HDPE, PS and PS-COOH particles (Llorca et al., 2018). Results
936 asserted that aromatic rings of PS contrasted NOM competition and, consequently, they
937 favoured PFAS adsorption. By contrast, NOM influenced PFAS adsorption on HDPE
938 and its effect was higher in SeaW than in RW (Llorca et al., 2018). Sun et al. (2018)
939 investigated the effect of co-existing organic compounds on GAC biofilter. They
940 reported that more active sites were available for PFAS adsorption in the pre-ozonated
941 regenerated GAC biofilter, due to the oxidation of competitive DOM (Sun et al., 2018).
942 Recently, the performance of new synthesized adsorbent (PEI-f-CMC) for PFAS removal
943 (both long- and short-chain) was evaluated using three different NOM solutions (Ateia et
944 al., 2018). Firstly, experiments were carried out in LW (Hartwell Lake, South Carolina)
945 and PFAS removal efficiency has been compared with that resulted from DOC free
946 solution. PFOA and PFOS removal efficiency was almost equal in both LW and DI.
947 PFAS with longer chain than PFOA and PFOS ($C_{12} - C_{13}$ PFCAs, $C_9 - C_{10}$ PFSA) were
948 removed better from LW than from DI using PEI-f-CMC. This result can be
949 explained by the co-removal of long-chain PFAS aggregated with OM (Du et al., 2014;
950 McCleaf et al., 2017). By contrast, short-chain PFAS removal was higher in DI than in
951 LW. Indeed, OM competed with short-chain PFAS during adsorption process (Ateia et
952 al., 2018). Overall, lower removal efficiency has been obtained for short-chain PFAS
953 compared with that observed for long-chain ones (removal $<20\%$ and $>70\%$,
954 respectively). The effect of different types of NOM (with different hydrophobicity) was
955 also investigated in order to assert the performance of PEI-f-CMC for PFAS removal.
956 The two NOM solutions tested had the same DOC concentration (2 mg L^{-1}) but a
957 different value of specific ultra-violet absorbance at 254 nm ($SUVA_{254}$). Results have

958 demonstrated that short-chain removal was lower than 30% from both NOM solutions
959 and the competition between short-chain PFAS and hydrophilic NOM ($SUVA_{254} = 1.7 \text{ L}$
960 $\text{mg}^{-1} \text{ m}^{-1}$) was higher than that with hydrophobic NOM ($SUVA_{254} = 4.9 \text{ L mg}^{-1} \text{ m}^{-1}$). The
961 hydrophobicity of NOM does not seem to influence the adsorption of long-chain PFAS
962 on PEI-f-CMC and their removal efficiency was higher than 70% for both NOM
963 solutions.

964 **6. Regeneration of adsorbents**

965 Despite the widespread use of adsorption and anion-exchange processes for PFAS
966 removal, the main drawback of their application at full-scale is the regeneration of spent
967 materials when their adsorption capacity has been exhausted. Regeneration must allow
968 the removal of adsorbed PFAS in order to restore the adsorption capacity of spent
969 materials by means of technologies, which have economic viability and environmental
970 security.

971 For instance, chemical regeneration of saturated adsorbents is usually performed in situ
972 at full scale treatment processes. However, it is not always feasible because of the
973 related costs or environmental issues, as discussed in the next section. On the other
974 hand, the thermal regeneration is often performed off-site at the adsorbent
975 manufacturing establishment. Therefore, a direct comparison between the above-
976 mentioned techniques is not easy. Overall, in-situ regeneration should be pursued to
977 avoid the transportation, treatment and/or disposal of exhausted materials.

978 To date, regeneration and reuse of materials saturated with PFAS is challenging, as
979 discussed in the following sections.

6.1. Chemical regeneration

980 PFAS anionic head could be desorbed using a solution of sodium salts (such as sodium
981 chloride, NaCl, and sodium hydroxide, NaOH), whereas a solution of organic alcohol is
982 required to desorb the hydrophobic C-F chain (Wang et al., 2014; Woodard et al.,
983 2017). Figure 4 depicts a schematic view of PFAS-saturated adsorbent which is
984 regenerated using a chemical solution made of sodium salts and organic alcohol. In
985 Figure 3 the desorption mechanisms related to the use of sodium ions and organic
986 solvents are illustrated as well.

987 Table 5 reports a compilation of data on chemical regeneration of PFAS-exhausted
988 adsorbents with particular attention to chemical solution employed and regeneration
989 percentage achieved. Due to the lower solubility of PFAS on ionic strength media,
990 conventional solutions of sodium salts were ineffective in regenerating adsorbents
991 exhausted by both long- and short-chain PFAS (Conte et al., 2015; Deng et al., 2015b).
992 Regeneration percentages were lower than 10% when regeneration agents, including
993 NaCl and NaOH, were adopted for desorbing PFBS and PFOS from acrylic resins
994 (Carter and Farrell, 2010; Deng et al., 2010). The deprotonation of amine groups on
995 acrylic gel-type resins allowed the desorption of PFOS at a low concentration of NaOH
996 solutions, although a regeneration percentage of about 90% was achieved when a
997 solution of sodium hydroxide and methanol was tested (Table 5).

998 Mixture of sodium salts and organic solvents (such as CH₃OH, C₂H₅OH and C₃H₆O)
999 have been widely used in order to involve the desorption of PFAS (Deng et al., 2015b;
1000 Yu et al., 2008; Zaggia et al., 2016). Moreover, the efficiency of regenerating solution
1001 depends on resin properties and, in particular, on the interactions established between
1002 PFAS and resin functional groups (Deng et al., 2010). In fact, data from regeneration
1003

1004 experiments carried out using different mixtures of inorganic salts with or without
1005 organic alcohol demonstrated that the amount of PFAS desorbed from three different
1006 types of strong AE resins depends on resin hydrophobicity and, consequently, on the
1007 interactions between PFAS and the alkyl chains of the functional exchange group
1008 (Zaggia et al., 2016). Solution of ammonium salts (such as NH_4OH and NH_4Cl) showed
1009 a good efficiency for the desorption of PFBA, PFBS, PFOA and PFOS from AE resins
1010 with trimethyl quaternary amine functional group (i.e., Purolite A600E and A520E).
1011 Whereas, the same regenerating solution was totally ineffective in reactivating highly
1012 hydrophobic resin (i.e., Purolite A532E), since the solvation of fluorine chain was
1013 needed in order to remove PFAS molecules from the active sites of the resin (Table 5).
1014 In that case, the organic solvent could slacken the hydrophobic interactions between
1015 PFAS and resin, then the inorganic anions of ammonium salts (OH^- or Cl^-) could replace
1016 PFAS (Figure 4), achieving the recovery of the exchange capacity of the resin.

1017 The effect of different temperatures of DI water as regenerating solution on PFOS
1018 desorption from BdAC was also investigated (Deng et al., 2015b). Results from
1019 regeneration experiments demonstrated that a higher temperature of DI water (about 80
1020 °C) enhanced the PFOS desorption and the regeneration percentage was about 53%
1021 (Table 5). Furthermore, PFOS desorption from PACFs was about 5% when DI water
1022 was employed as regenerating solution (Chen et al., 2017). By contrast, the
1023 regeneration percentage increased with the increase of ethanol concentration in the
1024 regeneration solution (Chen et al., 2017; Punyapalakul et al., 2013; Wang et al., 2014).
1025 Nevertheless, regenerating solution made of organic solvent is not suitable for the
1026 treatment of drinking water on full-scale applications, and consequently on-site

1027 regeneration is not feasible. It is noteworthy that the effect of OM co-adsorbed on
1028 materials may affect regeneration efficiency. After five successive adsorption-
1029 regeneration cycles, the efficiency of BdAC for PFCAs removal (PFHxA, PFHpA and
1030 PFOA) decreased, while AE resin (IRA67) performance was relatively stable (Du et al.,
1031 2015). Co-adsorbed organic pollutants on surface or pores of AC could interfere with
1032 the regeneration process. It was the case of the R-CAC, which was regenerated
1033 through different regenerating solutions (made of 70% ethanol, 90% methanol or 90%
1034 acetone). After the regeneration, R-CAC was reused for PFOS and F-53B removal from
1035 industrial wastewater (Du et al., 2016a). Due to the presence of non-desorbed OM, the
1036 adsorption efficiency of regenerated R-CAC decreased after each regeneration cycle.
1037 For this reason, persulfate oxidation was tested as alternative regeneration process to
1038 the conventional elution with organic solvents. Regeneration results demonstrated that
1039 sulphate free radicals were able to diffuse into pores of activated carbon and they
1040 allowed the degradation of both PFAS and other organic compounds. However,
1041 sulphate free radicals could cause the release of fluorine and short-chained PFAS due
1042 to break-down of long-chained PFAS (Du et al., 2016a). Consequently, further
1043 investigations should be addressed in order to better understand the effect of sulphate
1044 and other free radicals on degradation of PFOS.

1045 Overall, the need of organic solvents for the chemical regeneration of PFAS saturated
1046 adsorbents is a relevant limitation of this technique.

1047 **6.2. Thermal regeneration**

1048 Thermal regeneration in N₂ gas stream of GAC saturated with long-chain PFAS (PFOS,
1049 PFHxS and PFOA) has been investigated in order to evaluate the release of volatile
1050 organic fluorine (VOF) or other compounds such as short-chain compounds (Watanabe

1051 et al., 2016). Thermal regeneration may be optimized when GAC is maintained at 700
1052 °C and later off-gases will be kept at 1000 °C. Nevertheless, the high temperature
1053 needed for thermally regenerating AC may cause the decreasing of their adsorption
1054 properties and a change in AC morphology (Watanabe et al., 2018). These described
1055 aspect of thermal regeneration of activated carbon deserve a deep investigation.
1056 Calcination at 600 °C in air for 20 min has been investigated for the reusability of h-BNs
1057 saturated with PFDA and PFOS (Feng et al., 2016). A temperature of 600 °C was
1058 selected in order to reach the complete decomposition of the investigated PFAS
1059 compounds, since PFOS and PFDA are decomposed at about 480 °C and 200 °C,
1060 respectively, as inferred from differential scanning calorimetry curves (Feng et al.,
1061 2016). Moreover, calcium hydroxide ($\text{Ca}(\text{OH})_2$) was employed as additive to avoid the
1062 formation of short-chain fluorinated gases during calcination process. Results about
1063 regeneration experiments of h-BNs via calcining demonstrated that adsorption capacity
1064 was maintained approximately constant during different adsorption-regeneration cycles
1065 (the amount of PFOS or PFDA on the surface of h-BNs at equilibrium ranged about 0.35
1066 – 0.45 mg m^{-2}) (Feng et al., 2016).

1067 Recently, microwave (MW) irradiation became an alternative process to conventional
1068 thermal regeneration due to its advantages, such as rapidity and selectivity heating
1069 (Falciglia et al., 2018). This technique includes the conversion of MW electric field
1070 energy adsorbed by the solid medium (i.e., activated carbon) into heat at the molecular
1071 level (Falciglia et al., 2017). The dielectric properties of activated carbon combined with
1072 PFAS properties (such as their volatility) could allow the regeneration of PFAS-

1073 exhausted AC by means of the interactions between delocalized π -electrons of AC and
1074 the MWs. MW regeneration of PFAS-exhausted AC deserve further investigation.

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1075 **7. Conclusions and future research recommended**

1076 **7.1. Conclusions**

1077 Poly- and perfluoroalkyl substances (PFAS) are a large group of manmade chemicals
1078 characterized by high stability and persistence on environmental compartments. Among
1079 interim response measures, adsorption technologies (i.e., by activated carbon or anion-
1080 exchange resin) are currently employed for treatment of PFAS-impacted water.

1081 The present critical review of data on PFAS adsorption suggests that:

- 1082 ▪ To date, most of the experimental works published have been carried out under
1083 unrealistic operating conditions, such as high dosage of adsorbent materials,
1084 long EBCT, acid pH, and high concentrations of PFAS spiked in DI water. Such
1085 conditions are not representative of either full-scale treatment plants or actual
1086 environmental contaminations.
- 1087 ▪ Long-chain PFAS adsorption can involve the formation of aggregates on surface
1088 materials that may improve the adsorption efficiency by hydrophobic interactions.
1089 Furthermore, when different long-chain PFAS co-exist in solution, the longest
1090 PFAS outcompete the shorter due to a stronger hydrophobic interaction between
1091 the longest PFAS and the adsorbent surface.
- 1092 ▪ Meso- and macro-porous adsorbents (both AC and resin) showed high
1093 adsorption capacity because long-chain PFAS can easily access.
- 1094 ▪ Anion-exchange resins are more effective than activated carbon for both long-
1095 and short-chain PFAS removal.
- 1096 ▪ Regardless of the adsorbent used, the adsorption capacity of short-chain PFAS
1097 is lower than that observed for long-chain PFAS. Therefore, short-chain PFAS
1098 removal is more challenging.

- 1099 ▪ Adsorption of short-chain PFAS seems related to the availability of active pore
1100 sites because the formation of micelles (multi-layer adsorption) does not occur.
- 1101 ▪ Studies focused on the effect of OM during adsorption process report contrasting
1102 results. Overall, co-existing OM negatively affects short-chain PFAS adsorption
1103 due to active site competition, whereas the effect of OM seems less relevant
1104 during long-chain PFAS adsorption.
- 1105 ▪ Due to pore blockage and electrostatic repulsions, negative effect of OM on other
1106 adsorbent materials (such as biochar, mineral materials and magnetic-
1107 nanoparticles) is also reported.
- 1108 ▪ Effect of co-existing OM should be taken into account during both adsorption and
1109 regeneration processes.
- 1110 ▪ Chemical regeneration of PFAS saturated adsorbents requires the use of organic
1111 solvents, which are harmful. Nevertheless, thermal regeneration at high
1112 temperature of exhausted activated carbon causes the decline of adsorption
1113 capacity and may release dangerous short-chain fluorinated gases. Overall, the
1114 in-situ implementation of either chemical or thermal regeneration is not feasible.

1115 **7.2. Research needs**

1116 The following specific research needs can be drawn based on the observed gaps in
1117 PFAS removal by adsorption:

- 1118 • The investigation of PFAS removal by adsorption (especially short-chain PFAS)
1119 under typical water treatment conditions (i.e., adsorbent dose or EBCT, pH,
1120 PFAS concentrations) is strongly advised.
- 1121 • An advanced characterization of OM before and after the adsorption processes
1122 is also advised. In particular, fractionation of OM measuring hydrophobic,

1123 transphilic and hydrophilic fractions as well as acid, neutral and base fractions is
1124 relevant to better understand the adsorption mechanism and the effect of OM.
1125 Size exclusion chromatography (SEC) is also suggested to ascertain the effect
1126 of molecular weight of OM.

1127 • Characterization of adsorbent morphology by means of transmission electron
1128 microscopy, scanning electron microscopy and Fourier transform infrared
1129 spectroscopy before and after PFAS uptake is also useful.

1130 • The simultaneous effect of ionic strength and OM should be considered during
1131 PFAS adsorption and further investigations are required in order to better
1132 understand the contribution of ionic OM and inorganic ions (i.e., monovalent and
1133 divalent cations) on PFAS removal. Further experimental works should be
1134 performed at different OM fractions but at the same ionic strength in order
1135 minimize the possible effect of inorganic ions.

1136 • Economic assessment of PFAS removal techniques should also address the
1137 adsorbent disposal/regeneration. For instance, the management of the eluate
1138 produced by chemical regeneration of the resins should be considered.

1139 • Other unconventional techniques, such as microwave irradiation, may help to
1140 overcome issues concerning the regeneration of PFAS-saturated adsorbents.

1141 • A novel treatment train, which allows the combination of separation and
1142 destructive technologies is expected for the sustainable removal of PFAS.

1143

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1477 **Tables**

1478 **Table 1** Physic-chemical properties of selected PFAS.

1479 **Table 2** Batch adsorption experiments of long-chain PFAS fitted by Langmuir model.

1480 **Table 3** Batch adsorption experiments of short-chain PFAS fitted by Langmuir model.

1481 **Table 4** Experimental studies (both batch and column test) of PFAS adsorption performed in the
1482 presence of organic matter.

1483 **Table 5** Chemical regeneration of PFAS-saturated materials.

1484 **Figures**

1485 **Figure 1** Box-and-whiskers plot concerning the adsorption capacity of long- and short-chain
1486 PFAS on different adsorbent materials (AC: activated carbon; Resin; SM: Synthetized
1487 materials).

1488 **Figure 2** The main interactions established between OM and PFAS molecules during GAC
1489 adsorption. A: pore blockage; B: hydrophobic interaction; C: electrostatic repulsion.

1490 **Figure 3** The main interactions established between OM and PFAS molecules during anion-
1491 exchange process. B: Hydrophobic interaction; C: Electrostatic repulsion.

1492 **Figure 4** Chemical regeneration of PFAS-exhausted **adsorbents** using a solution of sodium salt
1493 and organic alcohol.

Table 1 Physico-chemical properties of selected PFAS.

Compound name (CAS No.)	Acronym	Formula	MW g mol ⁻¹	Log K _{ow}	Solubility in water (mg L ⁻¹)	Vapor pressure	pKa	Boiling point (°C)	Melting point (°C)	Density (g cm ⁻³)
Perfluoroalkyl carboxylic acids (PFCAs)										
<i>Short-chain (C₄-C₇)</i>										
Perfluorobutanoic acid (375-22-4)	PFBA	C ₃ F ₇ COOH	214.039 ^a	2.31 ^b	2.14 · 10 ⁵ (at 25 °C) ^c	6.38 mm Hg (25 °C) ^c	1.07 ^b	121 ^c	-17.5 ^c	1.651 ^c
Perfluoropentanoic acid (2706-90-3)	PFPeA	C ₄ F ₉ COOH	264.047 ^a	3.01 ^b	n.a.	n.a.	0.34 ^b	n.a.	n.a.	n.a.
Perfluorohexanoic acid (307-24-4)	PFHxA	C ₅ F ₁₁ COOH	314.054 ^a	3.48 ^d	15700 (at 25 °C) ^d	198 mm Hg (at 25 °C) ^d	-0.16 ^d	157 ^d	n.a.	
Perfluoroheptanoic acid (375-85-9)	PFHpA	C ₆ F ₁₃ COOH	364.062 ^a	4.15 ^d	3.65 (at 25 °C) ^d	0.133 mm Hg (at 25 °C) ^d	-2.29 ^d	175 ^d	30 ^d	1.792 (at 20 °C) ^d
<i>Long-chain (C₈-C₁₄)</i>										
Perfluorooctanoic acid (335-67-1)	PFOA	C ₇ F ₁₅ COOH	414.07 ^a	4.81 ^d	2290 (at 24 °C)	3.16 · 10 ⁻² mm Hg (at 25 °C) ^d	-0.5 – 4.2 ^d	192 ^d	54.3 ^d	3300 g mL ⁻¹ (at 24 °C) ^d
Perfluorononanoic acid (375-95-1)	PFNA	C ₈ F ₁₇ COOH	464.078 ^a	5.48 ^d	n.a.	8.3 · 10 ⁻² mm Hg (at 25 °C) ^d	-0.21 ^d	n.a.	n.a.	n.a.
Perfluorodecanoic acid (335-76-2)	PFDA	C ₉ F ₁₉ COOH	514.086 ^a	6.51 ^b	n.a.	n.a.	-5.2 ^b	217 (at 740 mmHg) ^a	77 – 78 ^a	1.707 (at 68 °F) ^a
Perfluoroundecanoic acid (2058-94-8)	PFUnDA	C ₁₀ F ₂₁ COOH	564.093 ^a	7.21 ^b	n.a.	n.a.	-5.2 ^b	n.a.	n.a.	n.a.
Perfluorododecanoic acid (307-55-1)	PFDODA	C ₁₁ F ₂₃ COOH	614.101 ^a	7.92 ^b	n.a.	n.a.	-5.2 ^b	n.a.	n.a.	n.a.
Perfluorotridecanoic acid (72629-94-8)	PFTTrDA	C ₁₂ F ₂₅ COOH	664.109 ^a	8.62 ^b	n.a.	n.a.	-5.2 ^b	n.a.	n.a.	n.a.
Perfluorotetradecanoic acid (376-06-7)	PFTeDA	C ₁₃ F ₂₇ COOH	717.117 ^a	9.32 ^b	n.a.	n.a.	-5.2 ^b	n.a.	n.a.	n.a.
Perfluoroalkyl sulfonic acids (PFSAs)										
<i>Short-chain (C₄-C₅)</i>										
Perfluorobutane sulfonic acid (375-73-5)	PFBS	C ₄ F ₉ SO ₃ H	300.095 ^a	1.82 ^d	344 (at 25 °C) ^d	2.68 · 10 ⁻² mm Hg (at 25 °C) ^d	-3.31 ^d	210 – 212 ^d	n.a.	1.811 g mL ⁻¹ (at 25 °C) ^d
Perfluoropentane sulfonic acid (2706-91-4)	PFPeS	C ₅ F ₁₁ SO ₃ H	350.102 ^a	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
<i>Long-chain (C₆-C₁₄)</i>										
Perfluorohexane sulfonic acid (355-46-4)	PFHxS	C ₆ F ₁₃ SO ₃ H	400.11 ^a	3.16 ^d	6.2 (at 25 °C) ^d	4.6 · 10 ⁻³ mm Hg (at 25 °C) ^d	0.14 ^d	238 - 239 ^d	n.a.	1.841 g L ⁻¹ ^d
Perfluoroheptane sulfonic acid (375-92-8)	PFHpS	C ₇ F ₁₅ SO ₃ H	450.118 ^a	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Perfluorooctane sulfonic acid (1763-23-1)	PFOS	C ₈ F ₁₇ SO ₃ H	500.126 ^a	4.49 ^d	3.2 · 10 ⁻³ (at 25 °C) ^d	2 · 10 ⁻³ mm Hg (at 25 °C) ^d	< 1.0 ^d	249 ^d	n.a.	n.a.
Perfluorononane sulfonic acid (474511-07-4)	PFNS	C ₉ F ₁₉ SO ₃ H	549.126 ^a	6.13 ^b	n.a.	n.a.	-3.24 ^b	n.a.	n.a.	n.a.

Table 1 Physico-chemical properties of selected PFAS (continued).

Compound name (CAS No.)	Acronym	Formula	MW g mol ⁻¹	Log K _{ow}	Solubility in water (mg L ⁻¹)	Vapor pressure	pKa	Boiling point (°C)	Melting point (°C)	Density (g cm ⁻³)
Perfluoroalkyl sulfonic acids (PFASs)										
<i>Long-chain (C₆-C₁₄)</i>										
Perfluorodecane sulfonic acid (335-77-3)	PFDS	C ₁₀ F ₂₁ SO ₃ H	600.141 ^b	6.83 ^b	n.a.	n.a.	-3.24 ^b	n.a.	n.a.	n.a.
Perfluoroundecane sulfonic acid (749786-16-1)	PFUnDS	C ₁₁ F ₂₃ SO ₃ H	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Perfluorododecane sulfonic acid (79780-39-5)	PFDoDS	C ₁₂ F ₂₅ SO ₃ H	700.157 ^a	8.23 ^b	n.a.	n.a.	-3.24 ^b	n.a.	n.a.	n.a.
Perfluorotridecane sulfonic acid (n.a.)	PFTriDS	C ₁₃ F ₂₇ SO ₃ H	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Perfluorotetradecane sulfonic acid (n.a.)	PFTeDS	C ₁₄ F ₂₉ SO ₃ H	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
Perfluoroalkane sulfonamides (FASAs)										
Perfluorooctane sulfonamide (754-91-6)	FOSA	C ₈ H ₂ F ₁₇ NO ₂ S	499.142 ^a	5.8 ^d	8.04 · 10 ⁻³ (at 25°C) ^d	0.31 mm Hg (at 25°C) ^d	3.37 ^b	n.a.	n.a.	n.a.
Potassium salt of 6:2 chlorinated polyfluorinated ether sulfonate (73606-19-6)	F-53B	6:2 Cl-PFAES	570.67 ^f	4.84 ^e	n.a.	n.a.	< 1 ^e	n.a.	n.a.	n.a.
n.a. not available, ^a PubChem (URL: https://pubchem.ncbi.nlm.nih.gov/); ^b Chemicalize (URL: https://chemicalize.com/#/); ^c Lide (2007); ^d Hazardous Substances Data Bank (HSDB) (URL: https://toxnet.nlm.nih.gov/); ^e predicted by software SPARC (Xiao, 2017); ^f (Gao et al., 2017).										

Table 2 Batch adsorption experiments of long-chain PFAS fitted by Langmuir model.

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹ , except stated otherwise)				
Activated carbon							
PFOA	15 – 150 mg L ⁻¹	GAC Calgon F400	1000	DI	T = 30 °C, pH 7.2,	112.1	(Ochoa-Herrera and Sierra-Alvarez, 2008)
	1 – 1000 mg L ⁻¹	PAC	1000	DI	T = 25 °C, 140 rpm, 48 h	426.49 (1.03 mmol g ⁻¹)	(Rattanaoudom et al., 2012)
	120 mg L ⁻¹	BdAC S _{BET} = 2450 m ² g ⁻¹ Effective size = 0.6–0.85 mm	200	Actual WW (PFOSE WWW)	T=25 °C, 170 rpm, 48 h, pH 4.0	372.6 (0.9 mmol g ⁻¹)	(Du et al., 2015)
	0.01 – 100,000 µg L ⁻¹	Biochar HWC S _{BET} = 453 m ² g ⁻¹ pore volume= 0.11 cm ³ g ⁻¹	ratio of biochar:PFOA 1:3 mg mL ⁻¹	LW (DOC=2 mg L ⁻¹) WW (DOC=4.9 mg L ⁻¹)	T = 22 °C, 30 d, pH 7.2	41.2	(Inyang and Dickenson, 2017)
						31.7	
						41.3	
	3.8 – 259 mg L ⁻¹	Biochar PWC S _{BET} = 413 m ² /g pore volume= 0.10 cm ³ /g	ratio of biochar:PFOA 1:3 mg mL ⁻¹	LW (DOC=2 mg L ⁻¹) WW (DOC=4.9 mg L ⁻¹)	Room temperature, 3 d, pH 6	27.7	(Wang et al., 2016)
						78.67 ± 8.28 (190 ± 20 mmol kg ⁻¹)	
	20 – 250 mg L ⁻¹	SWNT Single walled carbon nanotubes S _{BET} = 468 m ² g ⁻¹ pore volume = 0.52 cm ³ g ⁻¹ AC Calgon , microporous S _{BET} = 825 m ² g ⁻¹ pore volume = 0.54 cm ³ g ⁻¹	250	DI	T = 25 °C, 150 rpm, 168 h (12 h for PAC)	194.6 ± 4.14 (470 ± 10 mmol kg ⁻¹)	(Yu et al., 2009)
						277.42 (0.67 mmol g ⁻¹)	
0.05 - 10 mg L ⁻¹	PAC S _{BET} = 812 m ² g ⁻¹ micropore area = 466 m ² g ⁻¹ GAC S _{BET} = 712 m ² g ⁻¹ micropore area = 313 m ² g ⁻¹	100	DI	T = 25 °C, pH 6.5	161.48 (0.39 mmol g ⁻¹)	(Li et al., 2011)	
					2.69 ± 0.29 (0.0065 ± 0.0007 mmol g ⁻¹)		
20 – 300 mg L ⁻¹	MWNT S _{BET} = 519.7 m ² g ⁻¹ PAC S _{BET} = 1227.19 m ² g ⁻¹ GAC S _{BET} = 815.34 m ² g ⁻¹	100	DI	T = 25 °C, 180 rpm, pH 5	202.9 (0.49 mmol g ⁻¹)	(Chen et al., 2017)	
					178.05 (0.43 mmol g ⁻¹)		
PFOS	15 – 150 mg L ⁻¹	GAC Calgon F400	1000	DI	T = 30 °C, pH 7.2	236.4	(Ochoa-Herrera and Sierra-Alvarez, 2008)
		GAC Calgon F300				196.2	
		GAC Calgon URV-MOD1				211.6	
	1 – 1000 mg L ⁻¹	PAC	1000	DI	T = 25 °C, 140 rpm, 48 h	440.11 (0.88 mmol g ⁻¹)	(Rattanaoudom et al., 2012)
44.1 µg L ⁻¹	R-CAC S _{BET} = 1125 m ² g ⁻¹ pore volume = 0.90 cm ³ g ⁻¹	50 - 250	Effluent EWTP	T = 25 °C, 170 rpm, 48 h	0.345 mg g ⁻¹ (345 µg g ⁻¹)	(Du et al., 2016a)	

Table 2 Batch adsorption experiments of long-chain PFAS fitted by Langmuir model (continued).

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹ , except stated otherwise)				
Activated carbon							
PFOS	20 – 250 mg L ⁻¹	PAC S _{BET} = 812 m ² g ⁻¹ micropore area = 466 m ² g ⁻¹	100	DI	T = 25 °C, 150 rpm, 168 h (12 h for PAC)	520.13 (1.04 mmol g ⁻¹)	(Yu et al., 2009)
		GAC S _{BET} = 712 m ² g ⁻¹ micropore area = 313 m ² g ⁻¹				185.04 (0.37 mmol g ⁻¹)	
	20 – 300 mg L ⁻¹	PAC S _{BET} = 1227.19 m ² g ⁻¹	100	DI	T = 25 °C, 180 rpm, pH 5	535.13 (1.07 mmol g ⁻¹)	(Chen et al., 2017)
		GAC S _{BET} = 815.34 m ² g ⁻¹				390.1 (0.78 mmol g ⁻¹)	
	0.5 – 10 mg L ⁻¹	Corn-derived-biochar (T=250 °C) S _{BET} = 2.5 m ² g ⁻¹ total pore volume = 0.013 cm ³ g ⁻¹	200	DI + 0.01 mol L ⁻¹ of CaCl ₂ + 200 mg L ⁻¹ of NaN ₃	T = 25 ± 1 °C, 150 rpm, 48 h, pH 7	135.53	(Guo et al., 2017)
		Corn-derived-biochar (T=400 °C) S _{BET} = 3.75 m ² g ⁻¹ total pore volume = 0.017 cm ³ g ⁻¹				146.52	
		Corn-derived-biochar (T=550 °C) S _{BET} = 41.10 m ² g ⁻¹ total pore volume = 0.043 cm ³ g ⁻¹				166.42	
		Corn-derived-biochar (T=700 °C) S _{BET} = 297.58 m ² /g total pore volume = 0.199 cm ³ /g				169.90	
	59.9 – 415 mg L ⁻¹	SWNT S _{BET} = 468 m ² g ⁻¹ pore volume = 0.52 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	560.14 ± 40.01 (1120 ± 80 mmol kg ⁻¹)	(Wang et al., 2016)
	1 – 500 mg L ⁻¹	SWNT S _{BET} = 547.2 m ² g ⁻¹	200 - 1200	DI	T = 25 °C, 150 rpm, 2 h, pH 7	712	(Chen et al., 2011)
		MWNT10 S _{BET} = 324.9 m ² g ⁻¹				656	
		MWNT50 S _{BET} = 97.2 m ² g ⁻¹				514	
59.9 – 415 mg L ⁻¹	AC Calgon microporous S _{BET} = 825 m ² g ⁻¹ pore volume = 0.54 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	480.12 ± 10 (960 ± 20 mmol kg ⁻¹)	(Wang et al., 2016)	
0.05 - 10 mg L ⁻¹	MWNT S _{BET} = 519.7 m ² g ⁻¹	50	DI	T = 25 °C, pH 6.5	5.00 ± 0.5 (0.010 ± 0.001 mmol g ⁻¹)	(Li et al., 2011)	
30 – 200 mg L ⁻¹ (30 – 200 ppm)	PAC S _{BET} = 1521.85 m ² g ⁻¹	133.3	DI	T = 25 °C, 170 rpm	360.1 (0.72 mmol g ⁻¹)	(Liang et al., 2011)	
	40% Ni-PAC S _{BET} = 947.98 m ² g ⁻¹				355.1 (0.71 mmol g ⁻¹)		
F-53B	102.9 µg L ⁻¹	R-CAC S _{BET} = 1750 m ² g ⁻¹ pore volume = 1.36 cm ³ g ⁻¹	50 - 250	Effluent EWTP	T = 25 °C, 170 rpm, 48 h	1.059 (1059.4 µg g ⁻¹)	(Du et al., 2016a)

Table 2 Batch adsorption experiments of long-chain PFAS fitted by Langmuir model (continued).

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹)				
Molecularly imprinted polymers							
PFOA	1-12 mg L ⁻¹	DFB-CDP S _{BET} < 10 m ² g ⁻¹	100	DI	T= 23 °C, 400 rpm, 2 h	34	(L. Xiao et al., 2017)
PFOS	20 – 550 mg L ⁻¹	Chitosan-based MIP	100	DI	T = 25 °C, 36 h, pH 5	1455.52 (2910.3 μmol g ⁻¹)	(Yu et al., 2008)
		Chitosan-based NIP				1203.51 (2406.4 μmol g ⁻¹)	
	10 - 80 mg L ⁻¹	MIP-CMS	250	DI	T= 25 °C, 150 rpm, 2 h, pH 3	75.99	(Guo et al., 2018)
		NIP-CMS	250			43.94	
Anion-exchange resin							
F-53B	25-400 mg L ⁻¹	IRA67 Polyacrylic-DVB gel-type Polyamine Exchange capacity = 1.6 meq mL ⁻¹ Effective size = 0.3–1.2 mm	50	Actual WW (chrome plating WW)	T=25 °C, 150 rpm, 48 h, pH 3	2396.84 (4.2 mmol g ⁻¹)	(Gao et al., 2017)
PFOA	120 mg L ⁻¹	IRA67 Polyacrylic-DVB gel-type Polyamine Exchange capacity = 1.6 meq mL ⁻¹ Effective size = 0.3 – 1.2 mm	100	Actual WW (PFOSF WWW)	T=25 °C, 170 rpm, 48 h, pH 4.0	1167.67 (2.82 mmol g ⁻¹)	(Du et al., 2015)
	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	1436.82 (3.47 mmol g ⁻¹)	(Maimaiti et al., 2018)
	20 – 250 mg L ⁻¹	IRA400 Styrene-DVB gel-type Quaternary ammonium Exchange capacity = 1.4 meq mL ⁻¹	100	DI	T = 25 °C, 150 rpm, 168 h	1209.08 (2.92 mmol g ⁻¹)	(Yu et al., 2009)
	3.8 – 259 mg L ⁻¹	IRA400 Quaternary ammonium Exchange capacity = 1.4 meq mL ⁻¹	375	DI	Room temperature, 3 d, pH 6	331.25 ± 8.28 (800 ± 20 mmol kg ⁻¹)	(Wang et al., 2016)
PFHxS	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	1364.37 3.41 mmol g ⁻¹	(Maimaiti et al., 2018)
PFOS	25-400 mg L ⁻¹	IRA67 Polyacrylic-DVB gel-type Polyamine Exchange capacity = 1.6 meq mL ⁻¹	0.05	Actual WW (chrome plating WW)	T=25 °C, 150 rpm, 48 h, pH 3	2750.71 (5.5 mmol g ⁻¹)	(Gao et al., 2017)
	400 mg L ⁻¹	Exchange capacity = 1.6 meq mL ⁻¹ Particle size = 16 – 50 mesh	35 - 500	DI		2000.52 – 2500.65 (4 – 5 mmol g ⁻¹)	(Deng et al., 2010)

Table 2 Batch adsorption experiments of long-chain PFAS fitted by Langmuir model (continued).

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹)				
Anion-exchange resin							
PFOS	400 mg L ⁻¹	IRA958 Polyacrylic macroporous-type Quaternary ammonium Exchange capacity = 3.7 meq g ⁻¹ Effective size = 0.3 – 1.2 mm	35 - 500	DI	T=25 °C, 150 rpm, 48 h, pH 3	2000.52 – 2500.65 (4 – 5 mmol g ⁻¹)	(Deng et al., 2010)
	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	1395.36 2.79 mmol g ⁻¹	(Maimaiti et al., 2018)
	20 – 250 mg L ⁻¹	IRA400 Styrene-DVB gel-type Quaternary ammonium Exchange capacity = 1.4 meq mL ⁻¹	100	DI	T = 25 °C, 150 rpm, 168 h	210.05 (0.42 mmol g ⁻¹)	(Yu et al., 2009)
	59.9 – 415 mg L ⁻¹		375	DI	Room temperature, 3 d, pH 6	165.04 ± 20 (330 ± 40 mmol kg ⁻¹)	(Wang et al., 2016)
Other materials							
PFOS	15 – 150 mg L ⁻¹	13X zeolite	1000	DI	T = 30 °C, pH 7.2	12.0	(Ochoa-Herrera and Sierra-Alvarez, 2008)
		NaY80 zeolite				114.7	
	1 – 500 mg L ⁻¹	Chars from maize straw pyrolysis S _{BET} = 11.6 m ² g ⁻¹	200 - 1200	DI	T= 25 °C, 150 rpm, 384 h, pH 7	164	(Chen et al., 2011)
		Chars from willow sawdust pyrolysis S _{BET} = 7.21 m ² g ⁻¹				91.6	
	MA S _{BET} = 38.3 m ² g ⁻¹				811		
Synthesized materials							
PFDA	0.4 – 50 mg L ⁻¹	h-BNs S _{BET} = 125.5 m ² g ⁻¹ pore volume = 0.915 cm ³ g ⁻¹	200	DI	T= 23 ± 2 °C, 150 rpm, 5 mM ammonium acetate	0.16 mg m ⁻²	(Feng et al., 2016)
PFHxS	19.2 – 180 mg L ⁻¹	CTF S _{BET} = 1270 m ² g ⁻¹ pore volume = 0.63 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	224.06 ± 28 (560 ± 70 mmol kg ⁻¹)	(Wang et al., 2016)
PFOS	46.51 – 372.1 mg L ⁻¹	Crosslinked chitosan beads S _{BET} = 14.1 m ² g ⁻¹ Particle diameter = 4 – 5 mm	60.8	DI	T= 25 °C, 150 rpm, 150 h, pH 3	2605.67 (5.21 mmol g ⁻¹)	(Zhang et al., 2011)
	50 – 200 mg L ⁻¹	PAF-45 S _{BET} = 875.38 m ² g ⁻¹ pore volume = 0.40 mL g ⁻¹	12.5	DI	T= 25 °C, 180 rpm, 48 h, pH 3	5847.39	(Luo et al., 2016)

Table 2 Batch adsorption experiments of long-chain PFAS fitted by Langmuir model (continued).

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹)				
Synthesized materials							
PFOS	0.1 – 250 mg L ⁻¹	ARH	100	DI	T= 25 °C, 150 rpm, 24 h, pH 5	1325.34 (2.65 mmol g ⁻¹)	(Deng et al., 2013)
	0.4 – 50 mg L ⁻¹	h-BNs S _{BET} = 125.5 m ² g ⁻¹ pore volume = 0.915 cm ³ g ⁻¹	200	DI	T= 25 °C, 180 rpm, 5 mM ammonium acetate	0.04 mg m ⁻²	(Feng et al., 2016)
	59.9 – 415 mg L ⁻¹	CTF S _{BET} = 1270 m ² g ⁻¹ pore volume = 0.63 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	665.17 ± 40.01 (1330 ± 80 mmol kg ⁻¹)	(Wang et al., 2016)
	95.02 – 460.12 mg L ⁻¹	Quaternized cotton	100	DI	T= 25 °C, 150 rpm, 24 h, pH 5	1650.43 (3.3 mmol g ⁻¹)	(Deng et al., 2012)
	0.5 – 40 mg L ⁻¹	2-MNPs@FG S _{BET} = 169.85 - 225.42 m ² g ⁻¹	250	DI	220 rpm, 30 min	17.2	(Wang et al., 2018)
	20 – 300 mg L ⁻¹	PACFs S _{BET} = 1782 m ² g ⁻¹	100	DI	T= 25 °C, 180 rpm, pH 5	760.2 (1.52 mmol g ⁻¹)	(Chen et al., 2017)
PFOA	3.8 – 259 mg L ⁻¹	CTF S _{BET} = 1270 m ² g ⁻¹ pore volume= 0.63 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	269.14 (650 ± 30 mmol kg ⁻¹)	(Wang et al., 2016)
	78.67 – 381 mg L ⁻¹	Quaternized cotton	100	DI	T= 25 °C, 150 rpm, 24 h, pH 5	1283.61 (3.1 mmol g ⁻¹)	(Deng et al., 2012)
	0.1 – 207.03 mg L ⁻¹	ARH	100	DI	T= 25 °C, 150 rpm, 24 h, pH 5	1031.03 (2.49 mmol g ⁻¹)	(Deng et al., 2013)
	0.5 – 40 mg L ⁻¹	2-MNPs@FG S _{BET} = 169.85 - 225.42 m ² g ⁻¹	250	DI	220 rpm, 30 min	50.4	(Wang et al., 2018)
	20 – 300 mg L ⁻¹	PACFs S _{BET} = 1782 m ² g ⁻¹	100	DI	T= 25 °C, 180 rpm, pH 5	302.27 (0.73 mmol g ⁻¹)	(Chen et al., 2017)
	2 – 50 µg L ⁻¹	PEI-f-CMC S _{BET} = 7.8 m ² g ⁻¹	10	DI	T= 25±1 °C, 150 rpm, 24 h	2.32	(Ateia et al., 2018)
<p>S_{BET}: BET surface area; DI: Deionized water; PFSOF WWW: Perfluorooctanesulfonyl fluoride washing wastewater; LW: Lake water; EWTP: Electroplating wastewater treatment plant; GAC: Granular activated carbon; PAC: Powdered activated carbon BdAC: Bamboo-derived activated carbon; 40% Ni-PAC: Ni-compounded PAC with Ni loading an amount of ~40 wt%; R-CAC: reactivated coconut shell-based GAC; HWC: Harwood; PWC: Pinewood; SWNT: Single-walled carbon nanotubes; MWNT: Multi-walled carbon nanotubes; DFB-CDP: cross-linked polymer network where β-CD substitutes decafluorobiphenyl (DFB); MIP-CMS: molecularly imprinted carbon microsphere; NIP-CMS: non-imprinted carbon microsphere; MA: Ash from burning maize straw on stainless steel plate; CTF: Covalent triazine-based framework; h-BNs: porous hexagonal boron nitride nanosheets; PAF-45: porous aromatic framework constructed from benzene rings; ARH: Aminated rice husk; 2-MNPs@FG: Magnetic nano-particles attached into fluorographene (mass ratio of MNPs and FG is 3:5); WW: wastewater; PACFs: Polyacrylonitrile fiber (PANF)-derived activated carbon fibers; PEI-f-CMC: Poly(ethylenimine)-functionalized cellulose microcrystals.</p>							

Table 3 Batch adsorption experiments of short-chain PFAS fitted by Langmuir model.

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹)				
Activated carbon							
PFBA	6.5 – 204 mg L ⁻¹	AC Calgon microporous S _{BET} = 825 m ² g ⁻¹ pore volume = 0.54 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	51.36 ± 4.28 (240 ± 20 mmol kg ⁻¹)	(Wang et al., 2016)
PFHxA	31.4 mg L ⁻¹	BdAC S _{BET} = 2450 m ² g ⁻¹ Effective size = 0.6–0.85 mm	200	Actual WW (PFOSF WWW)	T=25 °C, 170 rpm, 48 h, pH 4	18.84 (0.06 mmol g ⁻¹)	(Du et al., 2015)
	7.2 – 217 mg L ⁻¹	AC Calgon microporous S _{BET} = 825 m ² g ⁻¹ pore volume = 0.54 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	235.54 ± 72.23 (750 ± 230 mmol kg ⁻¹)	(Wang et al., 2016)
PFHpA	40.04 mg L ⁻¹	BdAC S _{BET} = 2450 m ² g ⁻¹ Effective size = 0.6–0.85 mm	200	Actual WW (PFOSF WWW)	T=25 °C, 170 rpm, 48 h, pH 4	65.53 (0.18 mmol g ⁻¹)	(Du et al., 2015)
PFBS	6 – 247 mg L ⁻¹	AC Calgon microporous S _{BET} = 825 m ² g ⁻¹ pore volume = 0.54 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	51.01 ± 3 (170 ± 10 mmol kg ⁻¹)	(Wang et al., 2016)
	15 – 150 mg L ⁻¹	GAC Calgon F400	1000	DI	T = 30 °C, pH 7.2	98.7	(Ochoa-Herrera and Sierra-Alvarez, 2008)
Anion exchange resin							
PFBA	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	635.69 (2.97 mmol g ⁻¹)	(Maimaiti et al., 2018)
PFHxA	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	1089.76 (3.47 mmol g ⁻¹)	(Maimaiti et al., 2018)
	31.4 mg L ⁻¹	IRA67 Polyacrylic-DVB gel-type Polyamine Exchange capacity = 1.6 meq mL ⁻¹ Effective size = 0.3–1.2 mm	100	Actual WW (PFOSF WWW)	T=25 °C, 170 rpm, 48 h, pH 4	37.68 (0.12 mmol g ⁻¹)	(Du et al., 2015)
PFHpA	40.04 mg L ⁻¹	IRA67 Polyacrylic-DVB gel-type Polyamine Exchange capacity = 1.6 meq mL ⁻¹ Effective size = 0.3–1.2 mm	100	Actual WW (PFOSF WWW)	T=25 °C, 170 rpm, 48 h, pH 4	192.95 (0.53 mmol g ⁻¹)	(Du et al., 2015)
PFBS	50 – 400 mg L ⁻¹	IRA910 Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium Exchange capacity = 1.0 meq mL ⁻¹	100	DI	T= 25 °C, 160 rpm, 240 h, pH 6	1023.32 (3.41 mmol g ⁻¹)	(Maimaiti et al., 2018)

Table 3 Batch adsorption experiments of short-chain PFAS fitted by Langmuir model (continued).

Adsorbate		Adsorbent		Water matrix	Experimental setup	Adsorption capacity (mg-PFAS/g-adsorbent)	References
Compound	[PFAS] ₀	Type and properties	Batch dose (mg L ⁻¹)				
Synthesized materials							
PFBA	0.1 – 107.02 mg L ⁻¹	ARH	100	DI	T= 25 °C, 150 rpm, 24 h, pH 5	363.86 (1.70 mmol g ⁻¹)	(Deng et al., 2013)
	6.5 – 204 mg L ⁻¹	CTF S _{BET} = 1270 m ² g ⁻¹ pore volume = 0.63 cm ³ g ⁻¹	250	DI	Room temperature, 3 d, pH 6	92.03 ± 4.28 (430 ± 20 mmol kg ⁻¹)	(Wang et al., 2016)
PFHxA	7.2 – 217 mg L ⁻¹					376.86 ± 94.21 (1200 ± 300 mmol kg ⁻¹)	
PFBS	6 – 247 mg L ⁻¹					141.04 ± 12 (470 ± 40 mmol kg ⁻¹)	
AC: activated carbon; S _{BET} : BET surface area; DI: deionized water; PFOSF WWW: perfluorooctanesulfonyl fluoride washing wastewater; GAC: granular activated carbon; BdAC: bamboo-derived activated carbon; ARH: aminated rice husk; CTF: covalent triazine-based framework; WW: wastewater							

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter.

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
PFOS PFOA	200 $\mu\text{g L}^{-1}$ Spiking (Single solute)	PAC	10 mg L^{-1}	EfOM (WWTP effluent)	DOC = 7.3 mg L^{-1}	Batch • PFOA or PFOS + EfOM • PFOA or PFOS + PAC loaded of EfOM	PFOA (or PFOS) needs much contact time to reach the equilibrium in the presence of EfOM, in particular onto EfOM-preloaded PAC; EfOM (especially low-molecular-weight compounds) impacts negatively adsorption rate and capacity.	(Yu et al., 2012)
PFBA PFPeA PFHxA PFOA PFNA PFDA PFBS PFHxS PFOS	1 $\mu\text{g L}^{-1}$ of each PFAS Spiking (Mix)	GAC (F300, F600, 1240C)	0.38 min	DOM (SW) Clear Creek (Golden, CO)	DOC = 1.7 mg L^{-1}	Column RSSCT experiment	The presence of DOM leads to faster breakthrough of PFAS, in particular shortest chain compounds. Breakthrough of >20% for all PFAS tested has been reached at 11,000 BVs and 125,000 BVs, respectively in the presence and absence of DOM. F300 is less effective with PFBA and PFPeA.	(Appleman et al., 2013)
PFOS PFOA PFBS PFHxS	250.06 $\mu\text{g L}^{-1}$ 207.03 $\mu\text{g L}^{-1}$ 150.04 $\mu\text{g L}^{-1}$ 200.05 $\mu\text{g L}^{-1}$ Spiking (Single solute)	MWCNTs (MWCNTs-Pri MWCNTs-COOH MWCNTs-OH)	20 mg L^{-1}	Synthetic OM (HA, 1-naphthol, phenol, benzoic acid) (Sinopharm Chemical Regent) in DI	Range of tested concentration of HA, 1-naphthol, phenol and benzoic acid 0 - 2.5 mg L^{-1}	Batch • PFOS + HA (or 1-naphthol, phenol, benzoic acid) • PFBS (or PFHxS, PFOA) + HA or Phenol	Competition between PFAS and organic compounds on the adsorption sites of the MWCNTs; Effect of co-existing organic compounds on PFOS removal: HA > 1-naphthol > benzoic acid > phenol; % removal in the presence of HA or phenol: PFOS > PFOA > PFHxS > PFBS.	(Deng et al., 2015a)
PFOS F-53B	44.1 $\mu\text{g L}^{-1}$ 102.9 $\mu\text{g L}^{-1}$ Real (Mix)	R-CAC	100 mg L^{-1}	EfOM (EWTP effluent)	TOC = 78.3 mg L^{-1}	Batch adsorption experiments of actual WW and synthetic PFAS solution (OM free)	EWTP effluent contains high amount of hydrocarbon surfactants (high concentration of TOC) which are in competition with PFOS and F-53B for active sites on R-CAC; F-53B is better adsorbed on R-CAC than PFOS due to its more hydrophobic chain and its higher concentration in the actual wastewater.	(Du et al., 2016a)
PFOS PFOA	280 ng L^{-1} 550 ng L^{-1} (read from graph)	PAC (BET 880 $\text{m}^2 \text{g}^{-1}$) GAC (BET 800 $\text{m}^2 \text{g}^{-1}$)	10 min	Synthetic HA and FA (Sigma-Aldrich) in DW	Range of tested concentration of HA and FA 5 – 25 mg L^{-1}	Column	PAC and GAC efficiencies decrease with the increasing of DOC concentration.	(Pramanik et al., 2015)

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
C₄ – C₁₂, C₁₄ PFCA s (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDODA, PFTeDA) C₄, C₆, C₈ PFSA s (PFBS, PFHxS, PFOS) FOSA	0.1 µg L ⁻¹ Spiking (Mix)	GAC (F400)	6.1 min	DOM (DW)	DOC = 1.8 mg L ⁻¹	Column RSSCT experiment	DOC removal is not linked to PFAS removal from AE or GAC column. DOC loading seems to favour the agglomeration of PFAS on the adsorbent surface.	(McCleaf et al., 2017)
		AE resin (A-600, Polystyrene-DVB gel-type Quaternary ammonium)	4.9 min					
PFBS PFHxS PFOS PFHxA PFHpA PFOA PFNA PFDA	0.073 ± 0.01 µg L ⁻¹ 0.47 ± 0.08 µg L ⁻¹ 1.4 ± 0.2 µg L ⁻¹ 0.28 ± 0.04 µg L ⁻¹ 0.32 ± 0.04 µg L ⁻¹ 1.4 ± 0.13 µg L ⁻¹ 0.067 ± 0.005 µg L ⁻¹ 0.04 ± 0.02 µg L ⁻¹ Real (Mix)	GAC (coal-based, Silcarbon)	125 mg L ⁻¹	DOM (GW) (well at 9.5 m depth, downstream a water-resistant clothing manufacture)	DOC = 5.27 ± 0.12 ng L ⁻¹	Batch adsorption isotherm for each adsorbent have been measured for the undiluted PFAS contaminated GW	Effect of OM has not been highlighted.	(Hansen et al., 2010)
		PAC (anthracite coal, Silcarbon TH90 extra)	25 mg L ⁻¹					
C₄ – C₁₂ PFCA s (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDODA) C₄, C₆, C₈ PFSA s (PFBS, PFHxS, PFOS) FOSA	2.5 µg L ⁻¹ of each PFAS Spiking (Mix)	AE resin (A600, Polystyrene-DVB gel-type Quaternary ammonium)	10 g L ⁻¹	DOM (LW) <ul style="list-style-type: none"> •Görvåln (autochtonous DOM, hydrophobic) • Bolmen (terrestrial, algal and bacterial-derived DOM, hydrophilic) 	<ul style="list-style-type: none"> •DOC = 8.3 mg L⁻¹ •DOC = 10 mg L⁻¹ 	Batch Pre-equilibration condition (contact time about 15 min) 4 discrete DOM levels (8, 4, 2, 0 mg DOC L ⁻¹) for the two lake waters + PFAS mix	Effect of DOM is weak during PFAS removal by AE, in fact at the highest DOM concentration PFAS percentual removal has been reduced by 10% in comparison with DOC free solution;	(Kothawala et al., 2017)
		GAC (Filtrisorb 400, Calgon)					GAC performance is affected by DOC source and concentration. Most hydrophobic long chain PFAS are better removed at highest DOC concentration, in detail the increase of hydrophobic DOM (Görvåln) concentration enhances progressively PFAS removal. Whereas, only the absence or presence of hydrophilic DOM (Bolmen) seems to influence PFAS adsorption.	

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
PFOS PFOA PFDA	0.153 $\mu\text{g L}^{-1}$ 0.138 $\mu\text{g L}^{-1}$ 0.282 $\mu\text{g L}^{-1}$ Spiking (Mix)	GAC (Norit Darco 12x40)	5.2 min	Synthetic NOM (Suwanne river, International Humic Substances Society) in DI	DOC = 4.3 mg L^{-1}	Column RSSCT experiment	Similar breakthrough curves of PFAS tested have been obtained despite different DOM composition and concentration. DOM seems not influenced the amount of long-chained PFAS adsorbed since hydrophobic interaction represents the main adsorption mechanism.	(Sgroi et al., 2018a)
	0.153 $\mu\text{g L}^{-1}$ 0.269 $\mu\text{g L}^{-1}$ 0.238 $\mu\text{g L}^{-1}$ Spiking (Mix)			NOM (SW)	DOC = 5.4 mg L^{-1}			
	0.556 $\mu\text{g L}^{-1}$ 0.174 $\mu\text{g L}^{-1}$ Real (Mix)			EfOM (WWTP effluent secondary)	DOC = 7 mg L^{-1}			
PFHxS	200 mg L^{-1} Spiking	AE resin (IRA910, Polystyrene-DVB macroporous-type Dimethyl ethanol ammonium)	0.1 g L^{-1}	Synthetic TCE, MB, SDS (Sinopharm Chemical Reagent) in DI	Range of tested concentration of TCE, MB, SDS 0 – 5 mmol L^{-1}	Batch adsorption experiments are carried out by dissolving PFHxS with different organic compounds at varying concentration	Non-ionic co-existing organic compounds (i.e. MB and TCE) do not impacted PFHxS removal, regardless their concentration. Due to the affinity of AE with anionic species, IRA910 performance decreases with the increase of HA and SDS concentrations. In detail, HA and SDS occupies adsorption sites on IRA910 and the latest could form aggregates in competition with PFHxS.	(Maimaiti et al., 2018)
				Synthetic HA (Sinopharm Chemical Reagent) in DI	Range of tested concentration of HA 0 – 200 mg L^{-1}			
PFOA	100 mg L^{-1}	ACF	800 mg L^{-1}	Synthetic FA (Sinopharm Chemical Regent) in DI	Range of tested concentration of FA 0 – 500 mg L^{-1}	Batch adsorption experiments at varying FA concentrations are compared with blank experiments (FA free solution)	FA concentration of 100 mg L^{-1} causes a decrease of PFOA adsorbed (about 75.4% lower than that of FA free solution).	(Y. Wang et al., 2015)
PFHxA PFHpA PFOA	31.4 mg L^{-1} 40.04 mg L^{-1} 120.08 mg L^{-1} Real (Mix)	BdAC	0.2 g L^{-1}	DOM (PFOSF WWW)	TOC = 63.2 mg L^{-1}	Batch adsorption experiments of simulated (DI+PFAS) and actual WW	Co-existing organic matter in actual WW has no effect on PFCAs adsorption on IRA67, whereas their presence influences negatively BdAC efficiency.	(Du et al., 2015)
		AE resin (IRA67, Polyacrylic-DVB gel- type Polyamine)	0.1 g L^{-1}					

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
PFOA	1 $\mu\text{g L}^{-1}$ Spiking	DFB-CDP	10 mg L^{-1}	Synthetic HA (n.a.) in DI	20 mg L^{-1}	Batch kinetic experiments are performed by adding HA to the adsorbent suspension together with PFOA solution. Comparison between PFOA removal by DFB-CDP in the presence or absence of HA	DFB-CDP adsorption efficiency is unaffected by HA.	(L. Xiao et al., 2017)
PFOS PFBS	50 – 1000 $\mu\text{g L}^{-1}$ Spiking (Single compound)	Boehmite	5 g L^{-1}	Synthetic HA in DI Fluka Co. (Buchs, Switzerland)	Range of tested concentration of HA 2 – 50 mg L^{-1}	Batch adsorption experiments performed for 72 h for achieving the equilibrium	With the increasing of HA adsorbed, boehmite surface becomes negative and electrostatic repulsions cause the reduction in PFOS and PFBS sorption.	(F. Wang et al., 2015)
PFOA PFOS	340 ng L^{-1} 520 ng L^{-1} Real (Mix)	GAC (BET = 800 $\text{m}^2 \text{g}^{-1}$)	20 min	Commercial organics: BSA, sodium alginate and HA (Sigma-Aldrich) in LW	DOC = 7.88 \pm 0.07 mg L^{-1} Protein = 11.54 \pm 0.22 mg L^{-1} Carbohydrate = 8.92 \pm 0.28 mg L^{-1}	Column	Removal efficiency is the highest at the lower DOC concentration, due to the competition for adsorption sites. Protein has the greater influence on PFOA and PFOS adsorption behaviour by means of hydrophobic and electrostatic interactions.	(Pramanik et al., 2017)
PFOA PFOS	103.5 – 4140.7 $\mu\text{g L}^{-1}$ 125 – 5001.2 $\mu\text{g L}^{-1}$ Spiking (Single compound)	F-MT	10 mg L^{-1}	Synthetic HA (n.a.) in DI	Tested concentration of HA 1 and 10 mg L^{-1}	Batch comparison of PFOA and PFOS adsorbed amounts in absence or in presence of HA	Due to nanoscale interlayers and high hydrophobicity of F-MT, HA macromolecular with hydrophilic groups could not be adsorbed. Consequently, co-existing HA weakly interferes with PFOS and PFOA adsorption (little decrease of PFOS and PFOA adsorption appeared in the presence of 10 mg L^{-1} HA).	(Du et al., 2016b)
PFOS PFOA PFBA PFBS	50 mg L^{-1} Spiking (Mix)	GAC (Filtrisorb 600, Calgon)	400 mg L^{-1}	DOM (Landfill GW)	Range of tested concentration of TOC 5-20 mg L^{-1}	Batch adsorption isotherm experiments are conducted on both DI and landfill GW	Organic compounds in groundwater can competitive with PFAS for GAC adsorption sites as it is demonstrated by the great equilibrium and fast kinetic adsorption in DI instead of in the landfill groundwater.	(Zhao et al., 2011)

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
C₄ – C₁₄, C₁₆, C₁₈ PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDODA, PFTTrDA, PFTeDA, PFHxDA, PFODA) C₄, C₆, C₈, C₁₀ PFASs (PFBS, PFHxS, PFOS, PFDS) FOSA	10 µg L ⁻¹ of each PFAS (ΣPFAS=180 µg L ⁻¹) Spiking (Mix)	HDPE	5 mg L ⁻¹	NOM SeaW (Alfacs Bay) RW (Ebro River)	TOC = 7.735 mg L ⁻¹ TOC = 4.386 mg L ⁻¹	Batch Contact time 0, 4, 7, 50 days	PFAS adsorption on HDPE is influenced by the presence of NOM and this effect is more evidence in seawater where NOM concentration is higher than in river water. Percentages of adsorption on HDPE are below 30% and carboxylic acids are adsorbed better than PFASs (PFOS and PFDS no adsorption). Higher adsorption velocity despite HDPE. Aromatic rings of PS monomer favour PFAS adsorption on PS and they go against organic matter competition. PFAS adsorption on PS-COOH is similar to that on PS, but adsorption rate is lower. C ₁₁ and C ₁₈ are the most adsorbed compounds. Effect of NOM not assessed.	(Llorca et al., 2018)
		PS	2 mg L ⁻¹					
		PS-COOH	2 mg L ⁻¹					
PFOA PFOS	180 µg L ⁻¹ of each compound Spiking (Mix)	MNPs@FG (1-MNPs@FG, 2-MNPs@FG, 3-MNPs@FG)	400 mg L ⁻¹	Synthetic NOM (International Humic Substance Society) in DI	DOC = 2 mg L ⁻¹	Batch adsorption experiments in orbital shaker at room temperature	The performance of MNPs@FG tested is slightly influenced by the presence of synthetic NOM.	(Wang et al., 2018)
	5 µg L ⁻¹ of each compound Spiking (Mix)	MNPs@FG (2-MNPs@FG)	400 mg L ⁻¹	NOM (LW, RW, DW)	DOC = 1.7 mg L ⁻¹ DOC = 0.6 mg L ⁻¹ DOC < 0.1 mg L ⁻¹	Batch, investigation of PFOA and PFOS removal from environmental water	After 11 minutes, 2-MNPs@FG exhibits high removal efficiencies (about 99.2 %) for both PFOA and PFOS in all water matrixes tested. The residual concentration of PFOA and PFOS are 11 and 28 ng L ⁻¹ , 16 and 22 ng L ⁻¹ , 31 and 18 ng L ⁻¹ , in lake, river and drinking waters, respectively. Residual combined concentration of PFOA and PFOS (49 ng L ⁻¹) in DW is lower than health advisory level of 70 ng L ⁻¹ (US EPA, 2016)	

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
C₄ – C₈ PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA) C₄ - C₈ PFSA s (PFBS, PFHxS, PFHpS, PFOS)	$3.7 \pm 0.9 \mu\text{g L}^{-1}$ $5.2 \pm 0.4 \mu\text{g L}^{-1}$ $7.6 \pm 0.5 \mu\text{g L}^{-1}$ $1.5 \pm 0.1 \mu\text{g L}^{-1}$ $10.7 \pm 0.9 \mu\text{g L}^{-1}$	GAC (Filtrisorb 300, Calgon)	100 mg L ⁻¹	DOM (AFFF-impacted GW)	DOC = $46.0 \pm 0.9 \text{ mg L}^{-1}$	Batch adsorption experiments with unfiltered AFFF-impacted GW for 5 days (no equilibrium reached)	Effect of OM has not been highlighted.	(X. Xiao et al., 2017)
	$1.8 \pm 0.0 \mu\text{g L}^{-1}$ $21.0 \pm 0.3 \mu\text{g L}^{-1}$ $1.7 \pm 0.1 \mu\text{g L}^{-1}$ $33.1 \pm 0.6 \mu\text{g L}^{-1}$	Biochar (Mountain Crest Gardens, MCG, pine needle-derived biochar)						
PFOA PFOS	82.8 mg L^{-1} 129.12 mg L^{-1}	CTF	250 mg L ⁻¹	Synthetic HA (soil-extracted) in DI	Range of tested concentration of HA 10 – 40 mg L ⁻¹	Batch single-point adsorption data (equilibrium pH equal to 6) of each chemical testing the different adsorbent materials	HA do not strongly interfere with CTF adsorption since HA molecules are unable to enter in the internal pores of CTF due to size-exclusion mechanism.	(Wang et al., 2016)
	Spiking (Single compound)	AE resin (IRA-400, Amberlite Sinopharm Chemical Regent Co., China)	375 mg L ⁻¹				HA present slightly increases PFOA and PFOS adsorbed rates, maybe due to a weak expansion of the polymer matrix and consequently an increase of PFOA and PFOS access to more adsorption sites.	
		Microporous AC (Calgon Carbon Co., USA)	250 mg L ⁻¹				Direct competition for adsorption sites on AC and SWNT between dissolved HA and PFOA or PFOS. At the highest concentration of HA (40 mg L ⁻¹), the reduction of PFOA and PFOS adsorption	
		SWNT (Nanotech Port Co., China)	250 mg L ⁻¹				rates are 31-48 % and 95% for both AC and SWNT, respectively.	

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
C₄ – C₁₄ PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA) C₄, C₆, C₈ PFSAs (PFBS, PFHxS, PFOS)	212 ng L ⁻¹ 133 ng L ⁻¹ 109 ng L ⁻¹ 24 ng L ⁻¹ 430 ng L ⁻¹ <1 ng L ⁻¹ 1 ng L ⁻¹ <1 ng L ⁻¹ <1 ng L ⁻¹ 171 ng L ⁻¹ 13 ng L ⁻¹ 27 ng L ⁻¹ Real (Mix)	AE resin (A600E Polystyrene-DVB gel-type Trimethyl quaternary amine A520E Polystyrene-DVB macroporous-type Trimethyl quaternary amine A532E Polystyrene-DVB gel-type Bifunctional quaternary amine)	10.1 min	DOM (GW)	n.a.	Column Pilot-scale	Effect of OM has not been assessed.	(Zaggia et al., 2016)
		GAC C (mesh: 8x30, iodine number 900 mg g ⁻¹) J (mesh: 12x40, iodine number 1100 mg g ⁻¹) S (mesh: 12x40, iodine number 1000 mg g ⁻¹)	10 – 11 min			Column Full-scale		
PFBA PFOA	1 µg L ⁻¹ Spiking (Single compound)	Biochar (HWC, PWC)	50 mg in 150 mL 333 mg L ⁻¹ 1:3 mg mL ⁻¹ (biochar:PFAA volume ratio)	EfOM (Tertiary treated WW effluent)	DOC = 4.9 mg L ⁻¹	Batch adsorption kinetic tests, samples are continuously mixed over 30 days on an orbital shaker	Slow kinetics for PFBA and PFOA adsorption may be attributed to pre-loaded EfOM on the biochar surface. At the equilibrium, the amount of PFOA adsorbed is 3-4 times higher than the amount of PFBA sorbed on both HWC and PWC.	(Inyang and Dickenson, 2017)
PFOA	0.01 – 100000 µg L ⁻¹ Spiking	Biochar (HWC, PWC)	1:3 mg mL ⁻¹ (biochar:PFAA volume ratio)	EfOM (Tertiary treated WW effluent) NOM (LW, Lake Mead in Southern Nevada)	DOC = 4.9 mg L ⁻¹ DOC = 2.0 mg L ⁻¹	Batch adsorption isotherm tests using two test waters for evaluating the impact of source water on the adsorption of PFOA on different type of biochar	Effect of OM on PFOA adsorption is more intense in WW in which the higher DOC concentration has been measured. OM molecules occupy high-energy pore sites and consequently they lead to pore blockage during PFOA adsorption. OM adsorbed leads also to a change in surface charge with the increasing of repulsion forces between anionic PFOA molecules and biochar surfaces. PFOA adsorbed in WW is 25% less than that in LW.	

Table 4 Experimental studies (both batch and column test) of PFAS adsorption performed in the presence of organic matter (continued).

Adsorbate		Adsorbent		Organic matter		Experimental setup	Remarks	References
Target PFAS	Level	Type	Batch dose or EBCT	Source (Water matrix)	Concentration			
PFPeA PFHxA PFOA PFOS	17.6 ± 2.4 ng L ⁻¹ 9.5 ± 1.4 ng L ⁻¹ 7.3 ± 0.8 ng L ⁻¹ 4.1 ± 5.6 ng L ⁻¹ Real (Mix)	GAC (Norit 830) Anthracite (Leopold) Biochar (HWC)	8 min	EfOM (Tertiary treated WW effluent)	DOC = 4.9 mg L ⁻¹	Column Pilot-scale	Faster breakthrough for PFAS tested is observed in anthracite and biochar filters than GAC. The latter shows the best PFAS removal efficiency in comparison with anthracite and HWC. Effect of OM has not been highlighted.	(Inyang and Dickenson, 2017)
PFBS PFHpA PFNA PFOA	32 ng L ⁻¹ 27 ng L ⁻¹ 7 ng L ⁻¹ 26 ng L ⁻¹	Biofilter (O ₃ + GAC)	23 min	DOM (water reclamation plant)	DOC = 4.17 mg L ⁻¹	Column Pilot-scale	In the pre-ozonated regenerated GAC biofilter more active sites are accessible for PFAS, due to the oxidation of competitive organic compounds. However, short-chain PFAS adsorption amount is lower than long-chain ones.	(Sun et al., 2018)
C₄ – C₁₃ PFCAs (PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFDoDA, PFTrDA) C₄ – C₁₀ PFSAs (PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFNS, PFDS)	1 µg L ⁻¹ of each compound Spiking (Mix)	PEI-f-CMC	25 mg L ⁻¹	DOM (LW) Isolated NOM (hydrophobic type) Isolated NOM (hydrophilic type)	DOC = 2 mg L ⁻¹ SUVA ₂₅₄ = 2.0 L mg ⁻¹ m ⁻¹ DOC = 2.5 mg L ⁻¹ SUVA ₂₅₄ = 4.9 L mg ⁻¹ m ⁻¹ DOC = 2.5 mg L ⁻¹ SUVA ₂₅₄ = 1.7 L mg ⁻¹ m ⁻¹	Batch	The longest PFAS (PFDoDA, PFTrDA, PFNS, PFDS) are better removed in lake water than in DI (removal efficiency is ~100% and ~85%, respectively). Whereas, the removal efficiency of short PFCAs is higher in DI (<20%). Removal efficiency of others PFAS is almost equal in both LW and DI. The removal efficiency of short-chain PFAS is higher at the highest SUVA ₂₅₄ (hydrophobic NOM). However, short-chain PFAS removal efficiency is lower than 30%. The difference in NOM composition (different SUVA ₂₅₄) does not influence the adsorption of longer PFAS, whose removal efficiency is higher than 70%.	(Ateia et al., 2018)
<p>EBCT: Empty bed contact time; PAC: powdered activated carbon; EfOM: effluent organic matter; DOC: dissolved organic carbon; GAC: granular activated carbon; DOM: dissolved organic matter; SW: Surface water; RSSCT: rapid small-scale column tests; MWCNTs: multi-walled carbon nanotubes; HA: humic acids; R-CAC: reactivated coconut shell based GAC; EWTP: electroplating wastewater treatment plant; TOC: total organic carbon; DW: drinking water; FA: fulvic acids; AE: anion-exchange; GW: groundwater; LW: lake water; NOM: natural organic matter; DI: deionized water; WWTP: wastewater treatment plant; TCE: tri-chloroethylene; MB: methylbenzene; SDS: sodium dodecyl sulphate; ACF: activated carbon fiber; BdAC: bamboo-derived AC; PFSOF WWW: perfluorooctanesulfonyl fluoride washing wastewater; DFB-CDP: cross-linked polymer network where β-CD substitutes decafluorobiphenyl; BSA: bovine serum albumin; F-MT: Fluorinated montmorillonite; HDPE: high-density polyethylene; PS: polystyrene; PS-COOH: poly-styrene carboxylic acid; RW: river water; SeaW: seawater; MNPs@FG: magnetic nano-particles attached into fluorographene; AFFF: Aqueous film-forming foam; CTF: Covalent triazine-based framework; SWNT: single-walled carbon nanotubes; HWC: hardwood biochar; PWC: pinewood biochar; PEI-f-CMC: poly(ethylenimine)-functionalized cellulose microcrystals; SUVA₂₅₄: specific ultra-violet absorbance at 254 nm.</p>								

Table 5 Chemical regeneration of PFAS-saturated materials.

Adsorbate	Adsorbent	Water matrix in adsorption experiments	Ratio mass:solution mixture	Experimental setup	Chemical solution	Regeneration percentage (%)	References
PFOS	PAC hydrophobic surface $S_{BET} = 980 \text{ m}^2 \text{ g}^{-1}$ pore volume = $276 \text{ mm}^3 \text{ g}^{-1}$	DI	100 mg:50 mL	Batch Adsorbed PFOS is extracted by varying ethanol and Milli-Q water ratios $T=25 \text{ }^\circ\text{C}$	100% $\text{C}_2\text{H}_5\text{OH}$	75.41	(Punyapalakul et al., 2013)
					70% $\text{C}_2\text{H}_5\text{OH}$ + 30% DI	87.41	
					50% $\text{C}_2\text{H}_5\text{OH}$ + 50% DI	94.58	
	40% $\text{C}_2\text{H}_5\text{OH}$ + 60% DI				81.46		
	30% $\text{C}_2\text{H}_5\text{OH}$ + 70% DI				80.96		
	20% $\text{C}_2\text{H}_5\text{OH}$ + 80% DI				55.16		
	10% $\text{C}_2\text{H}_5\text{OH}$ + 90% DI				45.22		
	100% $\text{C}_2\text{H}_5\text{OH}$				101.27		
	70% $\text{C}_2\text{H}_5\text{OH}$ + 30% DI				101.14		
	50% $\text{C}_2\text{H}_5\text{OH}$ + 50% DI				100.16		
	40% $\text{C}_2\text{H}_5\text{OH}$ + 60% DI				74.74		
	30% $\text{C}_2\text{H}_5\text{OH}$ + 70% DI				71.50		
	20% $\text{C}_2\text{H}_5\text{OH}$ + 80% DI				68.47		
	10% $\text{C}_2\text{H}_5\text{OH}$ + 90% DI				58.69		
	100% $\text{C}_2\text{H}_5\text{OH}$				101.12		
70% $\text{C}_2\text{H}_5\text{OH}$ + 30% DI	100.72						
50% $\text{C}_2\text{H}_5\text{OH}$ + 50% DI	101.34						
40% $\text{C}_2\text{H}_5\text{OH}$ + 60% DI	61.70						
30% $\text{C}_2\text{H}_5\text{OH}$ + 70% DI	66.07						
20% $\text{C}_2\text{H}_5\text{OH}$ + 80% DI	60.98						
10% $\text{C}_2\text{H}_5\text{OH}$ + 90% DI	57.90						
PFOA	Purosorb PAD 500 Polystyrenic macroporous-type, nonionic Exchange capacity = n.a.	DI	5 g:250 mL	Batch, magnetic stirrer at 250 rpm in a thermostatic oven for 80 h $T=20 \text{ }^\circ\text{C}$	5% NH_4Cl	0	(Conte et al., 2015)
					5% NH_4OH	0	
					3% NH_4Cl + 3% NH_4OH	0	
	30% MeOH				20		
	50% MeOH + 3% NH_4OH				90		
	5% NH_4Cl				0		
	5% NH_4OH				0		
	3% NH_4Cl + 3% NH_4OH				0		
	30% MeOH				20		
	50% MeOH + 3% NH_4OH				90		
	5% NH_4Cl				20		
	5% NH_4OH				30		
	3% NH_4Cl + 3% NH_4OH				85		
	30% MeOH				< 20		
	50% MeOH + 3% NH_4OH				40		
5% NH_4Cl	35						
5% NH_4OH	35						
3% NH_4Cl + 3% NH_4OH	90						
30% MeOH	20						
50% MeOH + 3% NH_4OH	50						
* After 5 Adsorption/regeneration cycles. PAC: powdered activated carbon; S_{BET} : BET surface area; DI: deionized water; HMS: hexagonal mesoporous silica; OD-HMS: N-octyldichlorosilane grafted hexagonal mesoporous silica; AE: anion-exchange;							

Table 5 Chemical regeneration of PFAS-saturated materials (continued).

Adsorbate	Adsorbent	Water matrix in adsorption experiments	Ratio mass:solution mixture	Experimental setup	Chemical solution	Regeneration percentage (%)	References
PFOS	BdAC $S_{BET} = 2450 \text{ m}^2 \text{ g}^{-1}$ Effective size = 0.6–0.85 mm	DI	10 g:100 mL	Batch, orbital shaker at 175 rpm for 24 h $T = 25 \text{ }^\circ\text{C}$	DI	19.8	(Deng et al., 2015b)
					50% CH_3OH	93	
					50% $\text{C}_2\text{H}_5\text{OH}$	98	
				Batch, orbital shaker at 175 rpm for 7 h $T = 80 \text{ }^\circ\text{C}$	DI	53	
					Batch, orbital shaker at 175 rpm for 24 h	0.06% NaCl	
				0.06% NaOH			
				Batch, orbital shaker at 175 rpm for 23 h $T = 45 \text{ }^\circ\text{C}$		40% $\text{C}_2\text{H}_5\text{OH}$	
	10% $\text{C}_2\text{H}_5\text{OH}$	46					
	20% $\text{C}_2\text{H}_5\text{OH}$	71					
	30% $\text{C}_2\text{H}_5\text{OH}$	80					
	Batch, orbital shaker at 175 rpm for 12 h $T = 25 \text{ }^\circ\text{C}$	50% $\text{C}_2\text{H}_5\text{OH}$	70 *				
PFOA	BdAC $S_{BET} = 2450 \text{ m}^2 \text{ g}^{-1}$ Effective size = 0.6–0.85 mm	Actual WW	25 g:30 mL	Batch, spent adsorbents shake at a thermostatic shaker at 170 rpm for 12 h $T = 45 \text{ }^\circ\text{C}$	50% CH_3OH	60 *	(Du et al., 2015)
PFHpA						< 30 *	
PFHxA						< 10 *	
PFOA	AE resin Amberlite IRA-67 Acrylic gel-type, tertiary amine functional groups Exchange capacity = 1.6 meq mL^{-1}	10 g:30 mL	Batch, spent adsorbents shake at a thermostatic shaker at 170 rpm for 12 h $T = 25 \text{ }^\circ\text{C}$	1% NaCl/ CH_3OH	< 80 *		
PFHpA					< 40 *		
PFHxA					< 10 *		
PFBA PFOA PFBS PFOS	AE resin Purolite A600E Polystyrene-DVB gel-type, trimethyl quaternary amine functional groups Exchange capacity = 1.6 eq L^{-1}	DW	1 g:1 L	Batch, flash stirred for 18 h and later at controlled temperature for 60 h $T = 20 \text{ }^\circ\text{C}$	5% NaCl	0	(Zaggia et al., 2016)
					5% KCl		
					5% NH_4Cl	0 - 5	
					2% NaOH		
					2% KOH		
					2% NH_4OH		
	0.5% NaOH + 0.5% NaCl	< 10					
	0.5% KOH + 0.5% KCl	40 - 60					
	0.5% NH_4OH + 0.5% NH_4Cl						
	80% CH_3OH + 1% NH_4Cl	60 - 80					
	80% $\text{CH}_3\text{CH}_2\text{OH}$ + 1% NH_4Cl						
	AE resin Purolite A520E Polystyrene-DVB macroporous-type, trimethyl quaternary amine functional groups Exchange capacity = 0.9 eq L^{-1}	DW	1 g:1 L	Batch, flash stirred for 18 h and later at controlled temperature for 60 h $T = 20 \text{ }^\circ\text{C}$	1% NaCl/ CH_3OH	5% NaCl	
5% KCl							
5% NH_4Cl						< 10	
2% NaOH						0 - 5	
2% KOH							
2% NH_4OH						< 10	

* After 5 adsorption/regeneration cycles.

BdAC: bamboo-derived activated carbon; S_{BET} = BET surface area; AE: anion-exchange; WW: wastewater; DW: drinking water; DI: deionized water.

Table 5 Chemical regeneration of PFAS-saturated materials (continued).

Adsorbate	Adsorbent	Water matrix in adsorption experiments	Ratio mass:solution mixture	Experimental setup	Chemical solution	Regeneration percentage (%)	References
PFOS	Chitosan-based MIP	DI	0.01 g:100 mL	Batch regeneration solution in an orbital shaker for 24 h T = 40 °C	90% NaOH + 10% C ₃ H ₆ O	60 *	(Yu et al., 2008)
PFOS	AE resin Amberlite IRA-458 Acrylic gel-type, quaternary ammonium functional groups Exchange capacity = 1.25 meq mL ⁻¹	DI	n.a.	Column Resin loaded with PFOS or PFBS packed in glass column (diameter of 1 cm, height of 10 cm) Flow rate 20 mL/min for 12 h T=22 °C	NaCl (320 mM)	0.0	(Carter and Farrell, 2010)
					NaOH (320 mM)	0.36	
PFBS					NaOH (320 mM)	4	
PFOS	AE resin Amberlite IRA-958 Acrylic macroreticular-type, quaternary ammonium functional groups Exchange capacity = 0.8 eq L ⁻¹	Chromium planting WW (inorganic anions)	20 mg:100 mL	Batch Resin loaded with PFOS and regeneration solution in an orbital shaker for 24 h at 150 rpm T = 25 °C	5% NaCl	< 10	(Deng et al., 2010)
					5% NaCl + 10% CH ₃ OH		
					1% NaCl + 10% CH ₃ OH		
					1% NaCl + 30% CH ₃ OH		
					100% CH ₃ OH		
					4% NaOH		
	1% NaCl + 50% CH ₃ OH				70		
	1% NaCl + 50% CH ₃ OH				90		
	100% CH ₃ OH				< 5		
	4% NaOH				< 5		
	0.4% NaOH				45		
	0.04% NaOH				50		
	1% NaCl + 30% CH ₃ OH				20		
	1% NaCl + 50% CH ₃ OH				85		
	1% NaCl + 70% CH ₃ OH				95		
1% NaOH + 30% CH ₃ OH	95						
1% NaOH + 50% CH ₃ OH	90						
1% NaOH + 70% CH ₃ OH	90						
	AE resin Amberlite IRA-67 Acrylic gel-type, tertiary amine functional groups Exchange capacity = 1.6 meq mL ⁻¹						

* After 5 adsorption/regeneration cycles.

MIP: molecularly imprinted; AE: Anion-exchange; WW: wastewater; DW: drinking water; DI: deionized water.

Table 5 Chemical regeneration of PFAS-saturated materials (continued).

Adsorbate	Adsorbent	Water matrix in adsorption experiments	Ratio mass:solution mixture	Experimental setup	Chemical solution	Regeneration percentage (%)	References	
PFBA PFOA PFBS PFOS	AE resin Purolite A520E Polystyrene-DVB macroporous-type, trimethyl quaternary amine functional groups Exchange capacity = 0.9 eq L ⁻¹	DW	1 g:1 L	Batch, flash stirred for 18 h and later at controlled temperature for 60 h T = 20 °C	0.5% NaOH + 0.5% NaCl	10 - 20	(Zaggia et al., 2016)	
					0.5% KOH + 0.5% KCl	50 - 65		
					0.5% NH ₄ OH + 0.5% NH ₄ Cl	70 - 80		
					80% CH ₃ OH + 1% NH ₄ Cl	80 - 90		
					80% CH ₃ CH ₂ OH + 1% NH ₄ Cl			
					5% KCl	0		
	5% NaCl							
	5% KCl							
	5% NH ₄ Cl	0 - 5						
	2% NaOH	0						
	2% KOH							
	PFOS	R-CAC S _{BET} = 1750 m ² g ⁻¹	Actual WW	10mg:20 mL	Batch, shaking at 170 rpm for 12 h T = 50 °C	50% C ₂ H ₅ OH		76.0
70% C ₂ H ₅ OH						89.5		
90% C ₂ H ₅ OH						85.1		
100% C ₂ H ₅ OH						83.1		
50% CH ₃ OH						85.3		
70% CH ₃ OH						89.5		
F-53B		R-CAC S _{BET} = 1750 m ² g ⁻¹	Actual WW	10mg:20 mL	Batch, shaking at 170 rpm for 12 h T = 40 °C	90% CH ₃ OH	95.1	
						100% CH ₃ OH	90.2	
						50% C ₃ H ₆ O	80.6	
						70% C ₃ H ₆ O	85.6	
						90% C ₃ H ₆ O	91.6	
						100% C ₃ H ₆ O	90.0	
F-53B	R-CAC S _{BET} = 1750 m ² g ⁻¹	Actual WW	10mg:20 mL	Batch, shaking at 170 rpm for 12 h T = 50 °C	50% C ₂ H ₅ OH	80.4	(Du et al., 2016a)	
					70% C ₂ H ₅ OH	96.6		
					90% C ₂ H ₅ OH	90.2		
					100% C ₂ H ₅ OH	88.3		
					50% CH ₃ OH	88.3		
					70% CH ₃ OH	93.1		
	F-53B	R-CAC S _{BET} = 1750 m ² g ⁻¹	Actual WW	10mg:20 mL	Batch, shaking at 170 rpm for 12 h T = 40 °C	90% CH ₃ OH		97.7
						100% CH ₃ OH		92.6
						50% C ₃ H ₆ O		87.4
						70% C ₃ H ₆ O		92.3
						90% C ₃ H ₆ O		97.6
						100% C ₃ H ₆ O		93.4

AE: anion-exchange; WW: wastewater; DW: drinking water; DI: deionized water; R-CAC: reactivated coconut shell based GAC; S_{BET} = BET surface area.

Table 5 Chemical regeneration of PFAS-saturated materials (continued).

Adsorbate	Adsorbent	Water matrix in adsorption experiments	Ratio mass:solution mixture	Experimental setup	Chemical solution	Regeneration percentage (%)	References							
PFOS PFHxS PFBS	PCMA	DI	0.5 g/L of PCMA with 50 mg/L of each PFAS	Adsorbed PFAS are extracted with methanol. Regenerated PCMA are reused for subsequent PFAS sorption experiments.	CH ₃ OH	85 - 88 *	(Wang et al., 2014)							
PFOS								PACF	DI	10mg:100 mL	Batch, shaking at 180 rpm for 24 h T = 25 °C	DI	5	(Chen et al., 2017)
												10% C ₂ H ₅ OH	22	
	30% C ₂ H ₅ OH	65												
	50% C ₂ H ₅ OH	85												
					100% C ₂ H ₅ OH	87								
* After 5 adsorption/regeneration cycles DI: deionized water; PCMA: permanently confined micelle arrays; PACF: Polyacrylonitrile fiber (PANF)-derived activated carbon fibers														

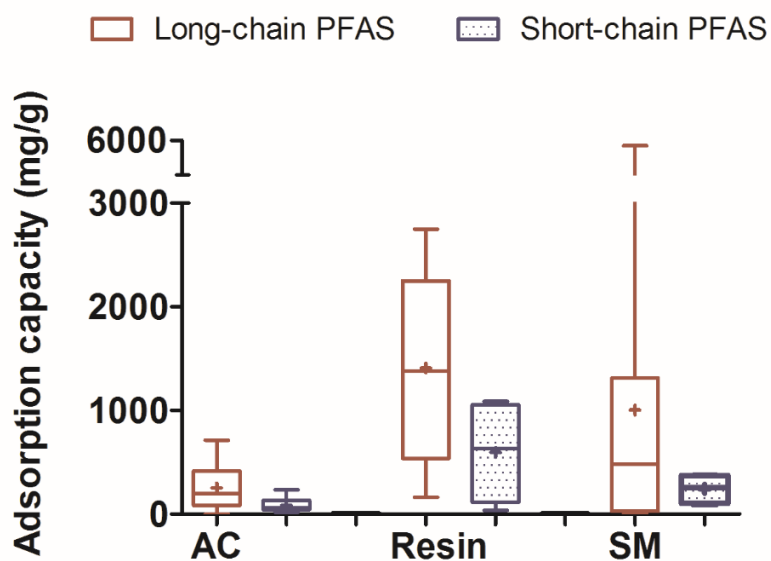


Figure 1 Box-and-whiskers plot concerning the adsorption capacity of long- and short-chain PFAS on different adsorbent materials (AC: activated carbon; Resin; SM: Synthetized materials).

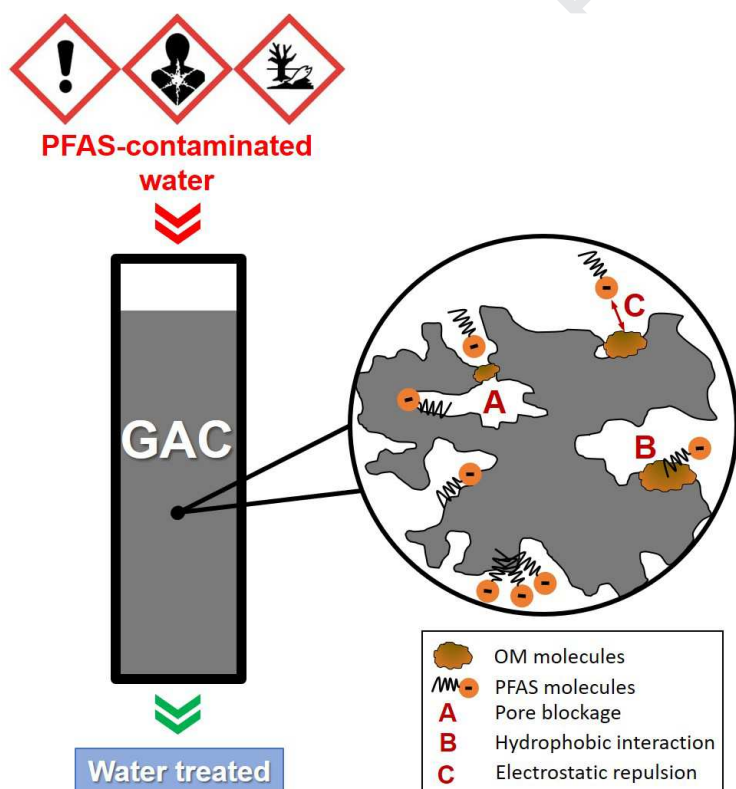


Figure 2 The main interactions established between OM and PFAS molecules during GAC adsorption. A: pore blockage; B: hydrophobic interaction; C: electrostatic repulsion.

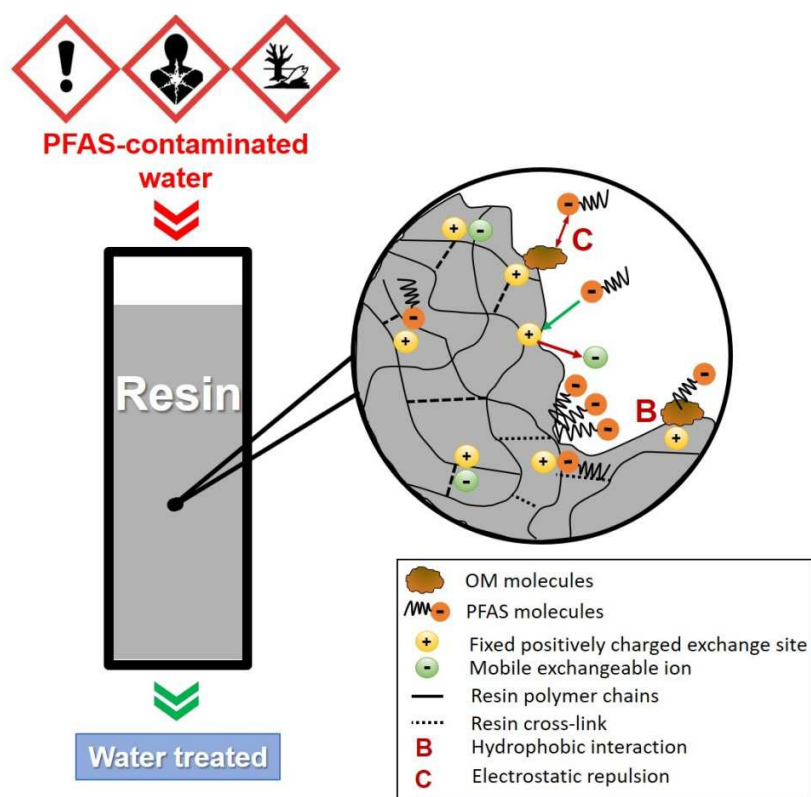


Figure 3 The main interactions established between OM and PFAS molecules during anion-exchange process. B: Hydrophobic interaction; C: Electrostatic repulsion.

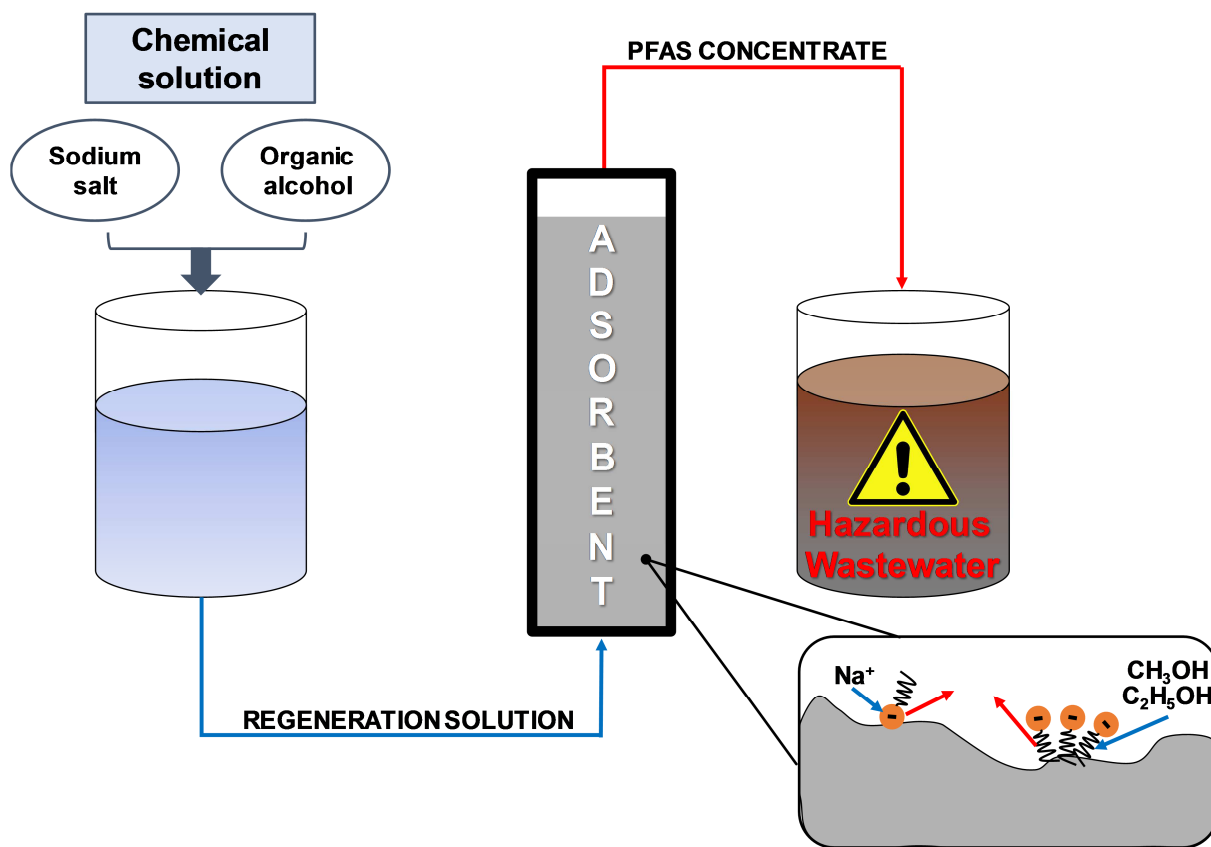


Figure 4 Chemical regeneration of PFAS-exhausted adsorbents using a solution of sodium salt and organic alcohol.

Highlights

- Most PFAS adsorption studies were carried out under unrealistic conditions.
- Organic matter often improves long-chain PFAS adsorption by aggregates formation.
- Co-existing OM reduces the short-chain PFAS adsorption efficiency.
- Short-chain PFAS are better removed by anion-exchange mechanism.
- Regeneration of PFAS-saturated adsorbents often needs harmful organic solvents.

Journal Pre-proof

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: