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Degradation and Removal Methods for Perfluoroalkyl and Polyfluoroalkyl Substances in Water

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Abstract

Several perfluoroalkyl and polyfluoroalkyl substances (PFASs) have been identified as chemicals of concern in the environment due to their persistence, global ubiquity, and classification as reproductive and developmental toxicants, endocrine disrupters, and possible carcinogens. Multiple PFASs are often found together in the environment due to product manufacturing methods and abiotic and biotic transformations. Treatment methods are needed to effectively sequester or destroy a variety of PFASs from groundwater, drinking water, and wastewater. This review presents a comprehensive summary of several categories of treatment approaches: (1) sorption using activated carbon, ion exchange, or other sorbents, (2) advanced oxidation processes, including electrochemical oxidation, photolysis, and photocatalysis, (3) advanced reduction processes using aqueous iodide or dithionite and sulfite, (4) thermal and nonthermal destruction, including incineration, sonochemical degradation, sub- or supercritical treatment, microwave-hydrothermal treatment, and high-voltage electric discharge, (5) microbial treatment, and (6) other treatment processes, including ozonation under alkaline conditions, permanganate oxidation, vitamin-B_{12} and Ti(III) citrate reductive defluorination, and ball milling. Discussion of each treatment technology, including background, mechanisms, advances, and effectiveness, will inform the development of cost-effective PFAS remediation strategies based on environmental parameters and applicable methodologies. Further optimization of current technologies to analyze and remove or destroy PFASs below regulatory guidelines is needed. Due to the stability of PFASs, a combination of multiple treatment technologies will likely be required to effectively address real-world complexities of PFAS mixtures and cocontaminants present in environmental matrices.

Keywords: destruction; fluorotelomer alcohol (FTOH); perfluorinated compounds; perfluorooctane sulfonic acid (PFOS); perfluorooctanoic acid (PFOA); polyfluorinated compounds; processes; remediation; transformation; treatment

Introduction

PERFLUOROALKYL AND POLYFLUOROALKYL substances (PFASs) are man-made, environmentally persistent contaminants used in many industrial, military, and consumer products, including nonstick coatings, electronics, and aqueous film-forming foams (AFFFs) (Houtz *et al.*, 2013; Wang *et al.*, 2014b, 2014c; Kotthoff *et al.*, 2015). Many studies demonstrate that PFASs are reproductive and developmental toxins, endocrine disrupters, possible carcinogens, and bioaccumulative (Ding and Peijnenburg, 2013; Gorrochategui *et al.*, 2014). PFASs can also be transported globally through several physicochemical and biological processes and found in remote and pristine locations (Paul and Jones, 2009; Butt *et al.*, 2014).

There are several provisional health-based guidelines for PFASs in drinking water. These guidelines range from a lifetime drinking water health advisory of 70 ng/L for combined perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) concentrations and 300–7,000 ng/L for C4–C7 PFASs (Wilhelm *et al.*, 2010; USEPA, 2016). Therefore, industries have shifted toward production of shorter-chain PFASs (Wang *et al.*, 2014b, 2014c).

Removal of PFASs from the environment to below the provisional guidelines is difficult using current treatment methods (Rahman *et al.*, 2014), and many studies are limited by analytical detection methods (up to mg/L). Wastewater

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Table 1. Filtration of Perfluoroalkyl Substances

Compound	*Material*	*Conditions*	*NaCl rejection (%)*	*$[PFAS]_0$ (mg/L)*	*Time (h)*	*PFAS rejection (%)*	*References*
Nanofiltration							
PFOA, PFBA, PFPeA, PFHxA, PFNA, PFDA	NF270	200 L, 18°C, pH 6.7, 0.17–0.97 MPa, feed flow = 1 L/min	>50%	0.001	24	93–99	Appleman *et al.* (2013)
PFOS, PFBS, PFHxS	NF270	200 L, 18°C, pH 6.7, 0.17–0.97 MPa, feed flow = 1 L/min	>50%	0.001	24	95–99	Appleman *et al.* (2013)
PFOS	DK	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	66.4	10	96	90–99	Tang *et al.* (2007)
	NF270	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	56.9	10	96	90–99	Tang *et al.* (2007)
	NF90	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	94.4	10	96	90–99	Tang *et al.* (2007)
Reverse osmosis							
PFOS	SG	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	95.2	10	96	>99	Tang *et al.* (2006, 2007)
	LFC1	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	97.3	10	96	>99	Tang *et al.* (2007)
	LFC3	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	98.5	10	96	>99	Tang *et al.* (2006, 2007)
	BW30	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	97.9	10	96	>99	Tang *et al.* (2006, 2007)
	ESPA3	25°C, pH 4, 1.38 MPa, feed flow = 1.37 L/min	94.9	10	96	>99	Tang *et al.* (2006, 2007)

and drinking water treatment plants do not effectively remove PFASs unless reverse osmosis, nanofiltration, or activated carbon (AC) is utilized, but these need to be frequently renewed or changed (Tang *et al.*, 2006, 2007; Takagi *et al.*, 2008; Shivakoti *et al.*, 2010; Thompson *et al.*, 2011; Appleman *et al.*, 2013; Flores *et al.*, 2013; Table 1).

Other removal methods use extreme conditions that are costly, such as high temperature and pressure. The structures and physicochemical properties (e.g., partitioning constants and solubility) of many PFASs are still uncertain, which pose challenges for their treatment (Rayne and Forest, 2009). Several different treatment methods may need to be applied to cost-efficiently remove PFAS mixtures. Currently, many studies focus on removal of the two most studied PFASs in the environment, PFOS and PFOA. However, most of these methods produce short-chain PFASs that have unknown toxicity.

This review article presents a synopsis of recently described removal methods and discusses the viability and effectiveness of these methods under the following categories (Tables 1–8): (1) sorption, (2) advanced oxidation processes (AOPs), (3) advanced reduction processes (ARPs), (4) thermal and nonthermal destruction, and (5) microbial treatment. All abbreviations, definitions, and equations are listed in Supplementary Tables S1–S6.

Discussion

Sorption processes

Sorption of PFASs has been studied for a wide variety of environmental matrices, mineral surfaces, and other adsorbents (Table 2). In all studies, it was assumed that PFAS molecules formed a monolayer on the adsorbent since PFAS concentration was lower than the critical micelle concentration.

Sorption occurs through two main interactions: (1) electrostatic and (2) hydrophobic. Meng *et al.* (2014) also demonstrated that air bubbles positively affected the sorption of PFOS onto carbonaceous materials, such as carbon nanotubes (CNTs), graphene, and powdered activated carbon (PAC), and the sorption was dependent on the surface polarity of the sorbent. PFOS prefers to exist at the air–water interface, resulting in the C–F chain partitioning into the air bubble, while the polar head group stays in aqueous solution. The effects of air bubble properties on PFASs have also been observed in sonolytic degradation and high-voltage electric discharge reactions.

Electrostatic interaction is a common sorption mechanism for PFASs. Since the pK_a values of PFOA and PFOS are 0.5 (Vierke *et al.*, 2013) and −2.3 (European Food Safety Authority, 2008), respectively, these compounds will likely be in anion form and sorb strongly to positively charged materials due to electrostatic interactions. Thus, pH plays an important role in PFAS sorption processes since it will affect the adsorbent's charge (Zhou *et al.*, 2010a). For example, Johnson *et al.* (2007) found that increasing pH caused two minerals, goethite and kaolinite, to become negatively charged, decreasing PFOS sorption. Electrostatic interaction can also change with monovalent cation concentrations, such as Na^+, due to increasing ionic strength, leading to compression of the electrical double layer (Wang and Shih, 2011; Xiao *et al.*, 2011; Wang *et al.*, 2012).

Hydrophobic interactions also play an important role in PFAS sorption. While previous studies have demonstrated

TABLE 2. SORPTION OF PER- AND POLYFLUOROALKYL SUBSTANCES

Compound	*Sorbent, amt (g)*	*[PFAS]$_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*^{a}	*Best fit isotherm, kinetics*	*References*
Aluminum								
PFOA	Alumina, 0.2	0.1	Pos. charged, $SA_{BET} = 88.6\ m^2/g$, particle size = 87.05 μm	T = 25°C, 20 mL pH 4.3, 150 rpm	48*	0.014	Langmuir, NA	Wang and Shih (2011)
	Boehmite (AlOOH), 0.1	0.2	$SA_{BET} = 299.2\ m^2/g$, particle size = 37.02 μm, $pH_{pzc} = 8.4$	T = 25°C, 20 mL, pH 7, 150 rpm	48*	0.189	Langmuir, NA	Wang *et al.* (2012a)
PFOS	Alumina, 0.2	0.1	Pos. charged $SA_{BET} = 88.6\ m^2/g$ particle size = 87.05 μm	T = 25°C, 20 mL pH 4.3, 150 rpm	48*	0.022	Langmuir, NA	Wang and Shih (2011)
	Boehmite (AlOOH), 0.1	0.2	$SA_{BET} = 299.2\ m^2/g$ particle size = 37.02 μm, $pH_{pzc} = 8.4$	T = 25°C, 20 mL pH 7, 150 rpm	48*	0.262	Langmuir, NA	Wang *et al.* (2012)
	Zeolite NaY80 (80 SiO_2/Al_2O_3), 0.1	15–150	SA = 780 m^2/g	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	114.7	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
Montmorillonite (Mt)–$Na_3(Al,Mg)_2Si_4O_{10}(OH_2)^{\bullet}4H_2O$								
PFOA	Mt, 0.1	0.6	$SA_{BET} = 67.52\ m^2/g$, $pH_{pzc} = 7.2$, CEC = 111 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.108	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFHxA	Mt, 0.1	0.6	$SA_{BET} = 67.52\ m^2/g$, $pH_{pzc} = 7.2$, CEC = 111 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.034	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFOS	Mt, 0.1	0.6	$SA_{BET} = 67.52\ m^2/g$, $pH_{pzc} = 7.2$, CEC = 111 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/LNaN_3	0.33*	0.294	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
	Na-Mt, 4	400	Pos. charged, $pH_{pzc} = 6.6$, $f_{OC} = 0.08\%$	T = 25°C, 25 mL pH 3, 150 rpm, 0.588 g/L $K_2Cr_2O_7$	120**	100^b	NA, NA	Zhou *et al.* (2013a)
	Na-Mt, 10	50–500	Pos. charged, CEC = 106 cmol/kg	T = 25°C, 25 mL pH 6.3, 150 rpm	24*	120	Langmuir, Pseudo-second-order	Zhou *et al.* (2010a)
	AET-Mt, 4	400	Pos. charged, $pH_{pzc} = 2.3$, $f_{OC} = 1.51\%$	T = 25°C, 25 mL pH 3, 150 rpm, 0.588 g/L $K_2Cr_2O_7$	120**	600^b	NA, NA	Zhou *et al.* (2013a)
	AET-HDTMAB-Mt, 4	400	Pos. charged, pH_{pzc} = None, $f_{OC} = 19.24\%$	T = 25°C, 25 mL pH 3, 150 rpm, 0.588 g/L $K_2Cr_2O_7$	120**	900^b	Linear, NA	Zhou *et al.* (2013a)
	HDTMAB-Mt, 4	400	Pos. charged, pH_{pzc} = None, $f_{OC} = 21.34\%$	T = 25°C, 25 mL pH 3, 150 rpm, 0.588 g/L $K_2Cr_2O_7$	120**	1,000^b	Linear, NA	Zhou *et al.* (2013a)
	2.5CEC-Mt, 10	50–500	Pos. charged, CEC = 106 cmol/kg	T = 25°C, 25 mL pH 6.3, 150 rpm	120**	855	Langmuir, Pseudo-second-order	Zhou *et al.* (2010a)

(*continued*)

TABLE 2. (CONTINUED)

Compound	*Sorbent, amt (g)*	*[PFAS]$_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*^{a}	*Best fit isotherm, kinetics*	*References*
PFHxS	Mt, 0.1	0.6	$SA_{BET}=67.52$, $pH_{pzc}=7.2$, CEC = 111 cmol/kg	250 mL, shaking, 0.2 g/L NaN_3	0.33*	0.344	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
Activated carbon								
PFOA	GAC F400, 0.1	15–150	Effective size = 0.55–0.75 mm	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	112.1	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
	GAC, 1.0477	53	NA	pH 4.4, diluted wastewater	168**	5	NA, NA	Lampert *et al.* (2007)
	PAC, 0.25	20	Particle size ≥100, $SA_{BET}=1{,}000\ m^2/g$	T = 25°C, 100 mL, pH 6.1, 120 rpm	1*	16.502	Langmuir, Pseudo-second-order	Qu *et al.* (2009)
	PAC Membrane Bioreactor, 0.48	0.2	Particle size ≤40 μm, $SA_{BET}=763.8\ m^2/g$	T = 28°C, 16 L, 36 L/h air, pH 6.8–7.5, wastewater	720**	4.511	Freundlich, NA	Yu *et al.* (2014)
	PAC-PACl, 5	5–50	$SA_{BET}=880\ m^2/g$, pore volume = $0.365\ cm^3/g$, micropore volume = $0.154\ cm^3/g$	Humic and fulvic acid, fast (300 rpm) and slow (40 rpm) mixing, settling, pH 4–7	NA	NA	NA	Pramanik *et al.* (2015)
PFBS	GAC F400, 0.1	15–150	Effective size = 0.55–0.75 mm	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	98.7	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
PFOS	GAC F300, 0.1	15–150	Effective size = 0.8–1.0 mm	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	196.2	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
	GAC F400, 0.1	15–150	Effective size = 0.55–0.75 mm	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	236.4	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
	GAC F400, 0.1	0.1	SA = 900–1,100 m^2/g	T = 25°C,75 mL, pH 6.4, 150 rpm	<20*^{b}	0.00018	Freundlich, NA	Senevirathna *et al.* (2010)
	GAC URV-MOD1, 0.1	15–150	Effective size = 1.0–1.18 mm	T = 30°C, 100 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	211.6	Freundlich, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
	GAC, 1.0477	16.8	NA	pH 4.4, diluted wastewater	168**	1.6	NA, NA	Lampert *et al.* (2007)
	PAC, 0.01	50	Size <0.1 mm, $SA_{BET}=812\ m^2/g$, $pH_{pzc}=7.5$	T = 25°C, 100 mL, pH 3, 150 rpm, 0.24 g/L NaH_2PO_4	4**	520	Freundlich, Pseudo-second-order	Yu *et al.* (2009)
	PAC Membrane Bioreactor, 0.48	0.2	Particle size ≤40 μm, $SA_{BET}=763.8\ m^2/g$	T = 28°C, 16 L, 36 L/h air, pH 6.8–7.5, Wastewater	720**	5.157	Freundlich, NA	Yu *et al.* (2014)

(*continued*)

Table 2. (Continued)

Compound	*Sorbent, amt (g)*	*[PFAS]$_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*[a]	*Best fit isotherm, kinetics*	*References*
	PAC-PACl, 5	NA	$SA_{BET}=880\ m^2/g$, pore volume = $0.365\ cm^3/g$, micropore volume = $0.154\ cm^3/g$	Humic and fulvic acid, fast (300 rpm) and slow (40 rpm) mixing, settling	NA	NA	NA	Pramanik *et al.* (2015)
	AC43767, 0.1	100	$SA_{BET}=1{,}553\ m^2/g$, microporous and mesoporous	T = 25°C, 200 mL, f = 250/min, 9.81 g/L H_2SO_4	10*[a]	1250[b]	NA, NA	Schuricht *et al.* (2014)
Several PFAAs	GAC Filtrasorb 300 (Calgon F300)	0.001	Coal based, apparent density = 0.48 g/mL	T = 18°C, 150 L feed flow = 1 L/min, pH 6.7				Appleman *et al.* (2013)
	GAC Calgon F600	0.001	Coal based, apparent density = 0.62–0.65 g/mL	DI water and Clear Creek surface water	192*	<10% PFAAs remaining in effluent		Appleman *et al.* (2013)
	GAC AquaCarb 1240C	0.001	Coconut based, Apparent density = 0.46–0.52 g/mL	DI water and Clear Creek surface water	792**			Appleman *et al.* (2013)
Iron								
PFOA	70% Hematite, 0.1	0.6	$SA_{BET}=9.9$, $pH_{pzc}=5.9$, CEC = 78 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.112	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFHxA	70% Hematite, 0.1	0.6	$SA_{BET}=9.9$, $pH_{pzc}=5.9$, CEC = 78 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.059	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFOS	Iron sand, 0.01–0.08	8	Pos. and Neg. charged, SA = $6\ m^2/g$	Up to 15 mL, 1–10 g/L KNO_3	24*	0.096[b]	Langmuir, NA	Johnson *et al.* (2007)
	Goethite, 0.01–0.08	8	Pos. charged, SA = $58\ m^2/g$, $pH_{pzc}=7.5–9.5$	up to 15 mL, 1–10 g/L KNO_3	24*	0.070[b]	Langmuir, NA	Johnson *et al.* (2007)
	Goethite, 0.417	0.005–1	Pos. charged, $SA_{BET}=76.1\ m^2/g$, $pH_{pzc}=9.4$	40 mL, pH 7, 0.584 g/L NaCl	<3*	0.180	Langmuir, NA	Tang *et al.* (2010)
	70% Hematite, 0.1	0.6	$SA_{BET}=9.9$, $pH_{pzc}=5.9$, CEC = 78 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.312	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFHxS	70% Hematite, 0.1	0.6	$SA_{BET}=9.9$, $pH_{pzc}=5.9$, CEC = 78 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.370	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFOS and PFOA	Hematite, 0.01–0.08	207	Pos. charged, $SA_{BET}=69.3\ m^2/g$, particle size = 10–20 nm	pH 6, 0.584 g/L NaCl	6**	5.19	NA, Pseudo-first-order	Gao and Chorover (2012)
Ion-exchange polymers								
PFOA	Amberlite IRA 400, 0.005	50	Exchange capacity = 3–3.5 eq/kg, size = 0.3–1.2 mm	T = 25°C, 100 mL, pH 3, 150 rpm, 0.24 g/L NaH_2PO_4	168*	1,403	Freundlich, Pseudo-first-order	Yu *et al.* (2009)
	US Filter A-714	4,320	NA	Wastewater	25**	686	NA, NA	Lampert *et al.* (2007)
	Amberlite XAD 7HP	0.414	Particle size 20–60 mesh, SA ≥380 m^2/g	T = 22.2°C, 40 mL, pH 7.8, 0.084 g/L $NaHCO_3$ 0.058 g/L NaCl	60**	NA	Freundlich, NA	Xiao *et al.* (2012)

(*continued*)

TABLE 2. (CONTINUED)

Compound	*Sorbent, amt (g)*	*[PFAS]$_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*^{a}	*Best fit isotherm, kinetics*	*References*
PFOS	Amberlite IRA 458, 10	175	Anion exchange with quarternary ammonium ion groups	T = 22°C, 6 L pH 4.4–5.8, 200 rpm, 1.22 g/L $NaClO_4$	4*	1,000^b	NA, NA	Carter and Farrell (2010)
	Amberlite IRA 400, 0.1	0.0001	Exchange capacity = 3–3.5 eq/kg, diameter = 0.8 mm	T = 25°C, 100 mL, pH 6.4, 150 rpm	<20*^{b}	0.0009	Freundlich, NA	Senevirathna *et al.* (2010)
	DowMarathon A, 0.1	0.0001	Exchange capacity = 1.3 eq/L, diameter = 0.57 mm	T = 25°C, 100 mL, pH 6.4, 150 rpm	<20*^{b}	0.00201	Freundlich, NA	Senevirathna *et al.* (2010)
	Amberlite XAD 7HP	0.5	Particle size 20–60 mesh, SA ≥380 m^2/g	T = 22.2°C, 40 mL, pH 7.8, 0.084 g/L $NaHCO_3$ 0.058 g/L NaCl	6**	NA	Freundlich, NA	Xiao *et al.* (2012)
	US Filter A-714	950	NA	500 mL, stirred, Wastewater, pH 2.5	25**	151	NA, NA	Lampert *et al.* (2007)
	AS-F860, 0.1	<25^b	Acrylate, trialkylammonium mesoporous	T = 25°C, 200 mL, 350 rpm, 9.81 g/L H_2SO_4	48**	1,250^b	NA, NA	Schuricht *et al.* (2014)
	AS-F500 and AW-F100, 0.1	100	Styrene/DVB mesoporous	T = 25°C, 200 mL, 350 rpm, 9.81 g/L H_2SO_4	16*^{b}	1,250^b	NA, NA	Schuricht *et al.* (2014)
PFBS	Amberlite IRA 458, 10	165	Anion exchange with quarternary ammonium ion groups	T = 22°C, 6 L pH 4.4–5.8, 200 rpm, 1.22 g/L $NaClO_4$	4*	300^b	NA, NA	Carter and Farrell (2010)
Nonion-exchange polymers								
PFOS	DowL493, 0.1	0.0001	SA = 1,100 m^2/g, diameter = 0.8 mm	T = 25°C, 100 mL, pH 6.4, 150 rpm	100*^{b}	0.00793	Freundlich, Pseudo-second-order	Senevirathna *et al.* (2010)
	Amb XAD 4, 0.1	0.0001	SA >750 m^2/g, diameter = 0.35–1.18 mm	T = 25°C, 100 mL, pH 6.4, 150 rpm	20*^{b}	0.00195	Freundlich, Pseudo-second-order	Senevirathna *et al.* (2010)
	DowV493, 0.1	0.0001	SA = 1,025 m^2/g, diameter = 0.8 mm	T = 25°C, 100 mL, pH 6.4, 150 rpm	100*^{b}	0.00929	Freundlich, Pseudo-second-order	Senevirathna *et al.* (2010)
Kaolinite–$Al_4Si(OH)_4$								
PFDA, PFNA, PFOS, PFUnDA, PFOA	Kaolinite, 0.02	0.513 PFDA, 0.463 PFNA, 0.499 PFOS, 0.563 PFUnDA, 0.413 PFOA	Neg. charged, SA = 10 m^2/g, pH_{pzc} = 5.1, CEC = 3.3 meq/100 g	T = 22°C, 40 mL, pH 7.5, 0.084 g/L $NaHCO_3$ 0.058 g/L NaCl	48*	NA	Linear, NA	Xiao *et al.* (2011)
PFOA	Kaolinite, 0.1	0.6	SA_{BET} = 23.11, pH_{pzc} = 3.6, CEC = 34 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.104	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFHxA	Kaolinite, 0.1	0.6	SA_{BET} = 23.11, pH_{pzc} = 3.6, CEC = 34 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.036	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)

(*continued*)

TABLE 2. (CONTINUED)

Compound	Sorbent, amt (g)	$[PFAS]_0$ (mg/L)	Sorbent properties	Conditions	Equilibration* or Exp.** time (h)	Adsorption capacity^a	Best fit isotherm, kinetics	References
PFOS	Kga-2, 0.01–0.08	8	Neg. charged, SA = 10 m^2/g, $pH_{pzc} = 4.6$, $f_{OC} = 0.02\%$	up to 15 mL, 1–10 g/L KNO_3	24*	0.050^b	Langmuir, NA	Johnson *et al.* (2007)
PFOS	Kaolinite, 0.1	0.6	$SA_{BET} = 23.11$, $pH_{pzc} = 3.6$, CEC = 34 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.303	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
PFHxS	Kaolinite, 0.1	0.6	$SA_{BET} = 23.11$, $pH_{pzc} = 3.6$, CEC = 34 cmol/kg	250 mL, shaking, 0.056 g/L $CaCl_2$, 0.2 g/L NaN_3	0.33*	0.312	Langmuir, Pseudo-second-order	Zhao *et al.* (2014)
Ottawa sand								
PFOS	Ottawa sand, 0.01–0.08	8	Neg. charged, SA = 0.002 m^2/g, $pH_{pzc} = 2.5$	up to 15 mL, 1–10 g/L KNO_3	24*	0.024^b	Langmuir, NA	Johnson *et al.* (2007)
Surfactants								
PFOS	CTAB ($C_{19}H_{42}BrN$), 0.0016	2	Pos. charged	T = 25°C, 45 mL pH 7, 250 rpm, 3 g sediment, 0.056 g/L $CaCl_2$	24**	0.06^b	Freundlich, NA	Pan *et al.* (2009)
	SDBS ($C_{18}H_{29}NaO_3S$), 0.0019	2	Neg. charged	T = 25°C, 45 mL pH 7, 250 rpm, 3 g sediment, 0.056 g/L $CaCl_2$	24**	0.01^b	Freundlich, NA	Pan *et al.* (2009)
Silica								
PFOS	Nanoparticles, 0.5	0.2	Neg. charged, $SA_{BET} = 63.4$ m^2/g, $pH_{pzc} < 3.0$, particle size ~15 nm	40 mL, pH 7, 0.584 g/L NaCl	96**	0.38	NA, NA	Tang *et al.* (2010)
Other sorbents								
PFOA	Electrochemical-assisted adsorption (MWCNT), 0.001	0.1	$SA_{BET} = 519.7$ m^2/g, $pH_{pzc} = 7.3$	T = 25°C, 20 mL pH 6.5, 0.6 V, 0.142 g/L Na_2SO_4	2^{*b}	406	Langmuir, Pseudo-second-order	Li *et al.* (2011)
	Electrocoagulation with Zinc, 200 cm^2	0.207	Zinc hydroxide flocs	400 mL, pH 5, 800 rpm, DC 0.584 g/L NaCl	<0.17*	2,376	Freundlich, Pseudo-second-order	Lin *et al.* (2015)
	MMCN, 150	280	$SA_{BET} = 404$ m^2/g, pore volume = 0.44 cm^3/g, pore size = 3.7 nm, $pH_{pzc} = 4.62$	T = 25°C, 250 mL, pH 3.25, stirred	3*	370.37	Langmuir, Pseudo-second-order	Yan *et al.* (2014)
	Hexagonal mesoporous silica (HMSc), 0.025	6.5	$SA_{BET} = 972$ m^2/g, pore volume ~0.95 cm^3/g	T = 25°C, ~25 mL, stirred	2*	5.13	Langmuir, Pseudo-second-order	Nassi *et al.* (2014)
	Hexagonal mesoporous silica (HMSe), 0.025	6.5	$SA_{BET} = 1{,}019$ m^2/g, pore volume ~0.95 cm^3/g	T = 25°C, ~25 mL, stirred	2*	6.12	Langmuir, Pseudo-second-order	Nassi *et al.* (2014)

(*continued*)

Table 2. (Continued)

Compound	*Sorbent, amt (g)*	*[PFAS]$_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*^{a}	*Best fit isotherm, kinetics*	*References*
	Mesoporous molecular sieve MCM-41, 0.025	6.5	$SA_{BET} = 850\ m^2/g$, pore volume = $0.70\ cm^3/g$	T = 25°C, ∼25 mL, stirred	2*	4.29	Langmuire, Pseudo-second-order	Nassi *et al.* (2014)
PFOS	MIP Chitosan cross-linked to ECH, 0.01	50	NA	T = 25°C, 100 mL, pH 5, 130 rpm, 0–29.22 g/L NaCl	32*	280	Freundlich, Pseudo-second-order	Yu *et al.* (2008)
	NIP Chitosan cross-linked to ECH, 0.0152–0.0153	186	Bead size = 2–3 mm, $SA_{specific} = 14.1\ m^2/g$, pore volume = $9.11\ cm^3/g$	T = 25°C, 200 mL, pH 3, 150 rpm, 0.174 g/L K_2SO_4 or 0.294 g/L $K_2Cr_2O_7$	60*^{b}	2,250^b	NA, double-exponential model	Zhang *et al.* (2011)
	NIP Chitosan cross-linked to ECH, 0.01	50	NA	T = 25°C, 100 mL, pH 5, 130 rpm, 0–29.22 g/L NaCl	16*	129	Langmuir, Pseudo-second-order	Yu *et al.* (2008)
	MIP with TRIA, cross-linker, 0.02	200	NA	T = 25°C, 20 mL, pH 5.1, 120 rpm	<0.5*^{b}	225^b	NA, NA	Deng *et al.* (2009)
	Electrochemical-assisted adsorption (MWCNT), 0.001	0.1	SA = $519.7\ m^2/g$	T = 25°C, 20 mL, pH 6.5, 0.6 V, 0.142 g/L Na_2SO_4	1*^{b}	470	Langmuir, Pseudo-second-order	Li *et al.* (2011)
	MWCNTs-ENFMs, 0.05–0.55	0.1	SA = $13.2\ m^2/g$	T = 25°C, 50 mL, pH 6, 150 rpm	40*	0.750	Freundlich, Pseudo-second-order	Dai *et al.* (2013)
	Electrocoagulation with Zinc, 200 cm^2	0.250	Zinc hydroxide flocs	400 mL, pH 5, DC 0.548 g/L NaCl	<0.17*	3,845	Freundlich, Pseudo-second-order	Lin *et al.* (2015)
	EFMs, 0.05	0.1	Polyvinyl alcohol (PVA)	T = 25°C, 50 mL, 150 rpm	2*	0.03057	Langmuir, Pseudo-first-order	Xu *et al.* (2013)
	EFMs, 0.05	0.1	Poly(D,L-lactide-co-glycolid) (PLGA)	T = 25°C, 50 mL, 150 rpm	2*	0.03098	Freundlich, Pseudo-first-order)	Xu *et al.* (2013)
	PCMAs), 0.01	1	TPODAC and TMB^c, 10–300 μm particle size	T = 25°C, 20 mL, 150 rpm	0.08*	1.75	Freundlich, NA	Wang *et al.* (2014a)
	MMCN, 150	280	$SA_{BET} = 404\ m^2/g$, pore volume = $0.44\ cm^3/g$, pore size = 3.7 nm, $pH_{pzc} = 4.62$	T = 25°C, 250 mL, pH 3.25, stirred	3*	454.55	Langmuir, Pseudo-second-order	Yan *et al.* (2014)
	PA-F2600, 0.1	100	$SA_{BET} = 826\ m^2/g$, pore size = 6–20 nm	T = 25°C, 200 mL, 350 rpm, 9.81 g/L H_2SO_4	8*^{b}	750^b	NA, NA	Schuricht *et al.* (2014)
PFBS, PFHxS	PCMAs, 0.01	1	TPODAC and TMB^c, 10–300 μm particle size	T = 25°C, 20 mL, 150 rpm	0.08*	1.25 PFBS, 1.45 PFHxS	Freundlich, NA	Wang *et al.* (2014a)

(*continued*)

Table 2. (Continued)

Compound	*Sorbent, amt (g)*	*$[PFAS]_0$ (mg/L)*	*Sorbent properties*	*Conditions*	*Equilibration* or Exp.** time (h)*	*Adsorption capacity*[a]	*Best fit isotherm, kinetics*	*References*
Environmental matrices								
PFOA	Anaerobic sludge, 2 g wet	0.26	Moisture content = 70.5%, VSS 10.1%	T = 25°C, 200 mL pH 5.5–5.8, 150 rpm	11*	0.041[b]	NA, Pseudo-second-order	Zhou *et al.* (2010b)
PFOS	Anaerobic sludge, 2 g wet	0.315	Moisture content = 70.5%, VSS 10.1%	T = 25°C, 200 mL pH 5.5–5.8, 150 rpm	11*	0.082	NA, Pseudo-second-order	Zhou *et al.* (2010b)
	Anaerobic digested sewage sludge[d], 0.024–0.352 g VSS	15–150	TSS = 17.20%, VSS = 11.76%	T = 30°C, 200 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	6.14	Langmuir, NA	Ochoa-Herrera and Sierra-Alvarez (2008)
	Anaerobic granular sludge[d], 0.024–0.352 g VSS	15–150	TSS = 7.45%, VSS = 6.98%	T = 30°C, 200 mL, pH 7.2, 150 rpm, 3 mM PO_4^{3-}	48**	120[b]	Freundlich, NA	Ochoa-Herrera and Sierra-Alvarez (2008)

[a] mg-PFASs/g-sorbent.

[b] Estimated from figure in article.

[c] [3-(Trymethoxysily)propyl]-octadecyldimethylammonium chloride (TPODAC); 1,3,5-trimethyl benzene (TMB).

[d] Anaerobic digested sewage sludge from Ina Road municipal WWTP (Tuscon, AZ). Anaerobic granular sludge from industrial reactor treating wastewater from alcohol distillery (Nedako, the Netherlands).

AC, activated carbon; EFMs, electrospun fiber membranes; MMCN, magnetic mesoporous carbon nitride; NA, not available (not mentioned in study); PCMA, permanently confined micelle array; Pos., positively; Neg., negatively.

TABLE 3. ELECTROCHEMICAL DEGRADATION OF PERFLUOROALKYL COMPOUNDS

Compound	*Anode/ cathode*	*Working/counter/ reference electrode*	*Working area (cm^2)/plate distance (cm)*	*Conditions*	*Time (h)*	*$[PFAS]_0$ (mg/L)*	*% Removal*	*% F^-*	*Kinetics*	*References*
PFOA	BDD/Pt-deposited Ti plate	BDD/Pt wire/Ag/ AgCl	0.26/NA	300 mL, 0.2 L/min, 1.22 g/L $NaClO_4$ 0.15 mA/cm^2	8	3,312	60[a]	22[a]	Pseudo first-order	Ochiai *et al.* (2011a)
	BDD/Pt foil	BDD/NA/NA	27/1	500 mL, 1.5 wt% P25 TiO_2, 254 nm, 0.2 L/min, 12.2 g/L $NaClO_4$, 0.6 mA/cm^2	3	1,035	35[a]	NA	Pseudo first-order	Ochiai *et al.* (2011c)
	BDD/Nb/tungsten	BDD/Nb/NA/NA	42/0.8	T = 20°C, 6.66 L/min, 5 g/L Na_2SO_4, 20 mA/cm^2	6	100	>90[a]	36[a]	First-order	Urtiaga *et al.* (2015)
	Nanocrystalline PbO_2 film/Ti	Nanocrystalline PbO_2 film/Pt wire/SCE	97.5/1	T = 25°C, 100 mL, 1.22 g/L $NaClO_4$, 20 mA/cm^2	1.5	100	96.7	81.7	Pseudo first-order	Niu *et al.* (2012)
	Si/BDD	Si/BDD/Ti sheet/ SCE	8.5/3	T = 32°C, 40 mL, pH 3, 1,500 rpm, 1.4 g/L $NaClO_4$, 23.24 mA/cm^2	2	50	>90[a]	60[a]	Pseudo first-order	Zhuo *et al.* (2012)
	Ti/SnO_2–Sb/Ti	Ti/SnO_2–Sb/Pt wire/SCE	NA/1	T = 25°C, 100 mL, pH 5, 1.22 g/L $NaClO_4$, 10 mA/cm^2	1.5	100	90.3	72.9	Pseudo first-order	Lin *et al.* (2012b)
	Ti/SnO_2–Sb/ PbO_2/Ti	Ti/SnO_2–Sb/PbO_2/ Pt wire/SCE	NA/1	T = 25°C, 100 mL, pH 5, 1.22 g/L $NaClO_4$, 10 mA/cm^2	1.5	100	91.1	77.4	Pseudo first-order	Lin *et al.* (2012b)
	Ti/SnO_2–Sb/ MnO_2/Ti	Ti/SnO_2–Sb/MnO_2/ Pt wire/SCE	NA/1	T = 25°C, 100 mL, pH 5, 1.22 g/L $NaClO_4$, 10 mA/cm^2	1.5	100	31.7	45.6	Pseudo first-order	Lin *et al.* (2012b)
	Ti/SnO_2–F[b]/Ti sheet	Ti/SnO_2–F/Pt sheet/ SCE	NA/1	T = 25°C, 50 mL, 1.22 g/L $NaClO_4$, 20 mA/cm^2	0.5	100	>99	80[a]	Pseudo first-order	Yang *et al.* (2015)
	Ti/RuO_2/stainless steel	Ti/RuO_2/NA/NA	100/1.6	Room temperature, AFFF-impacted groundwater, 500 mg/L Na_2SO_4, 1.5 mL/ min, 200 W, 10 mA/cm^2	8	90	90[a]	58	First-order	Schaefer *et al.* (2015)
PFBA, PFPeA, PFHxA, PFHpA	Nanocrystalline PbO_2 film/Ti	Nanocrystalline PbO_2 film/Pt wire/SCE	97.5/1	T = 25°C, 100 mL, 1.22 g/L $NaClO_4$, 20 mA/cm^2	1.5	100	31.8 PFBA, 41.4 PFPeA, 78.2 PFHxA, 97.9 PFHpA	14.9 PFBA, 27.5 PFPeA, 62.5 PFHxA, 86.2 PFHpA	Pseudo first-order	Niu *et al.* (2012)
PFBA, PFHxA, PFDA	Si/BDD	Si/BDD/Ti sheet/ SCE	8.5/3	T = 32°C, 40 mL, pH 3, 1,500 rpm, 1.4 g/L $NaClO_4$, 23.24 mA/cm^2	2	24.4 PFBA, 35.8 PFHxA, 58.6 PFDA	95[a] PFBA, 95[a] PFHxA, 98[a]PFDA	80[a] PFBA, 70[a] PFHxA, 50[a] PFDA	Pseudo first-order	Zhuo *et al.* (2012)
PFOS	Si/BDD	Si/BDD/Ti sheet/ SCE	8.5/3	T = 32°C, 40 mL, pH 3, 1,500 rpm, 1.4 g/L $NaClO_4$, 23.24 mA/cm^2	2	57	95[a]	70[a]	Pseudo first-order	Zhuo *et al.* (2012)

(*continued*)

Table 3. (Continued)

Compound	*Anode/ cathode*	*Working/counter/ reference electrode*	*Working area (cm^2)/plate distance (cm)*	*Conditions*	*Time (h)*	*$[PFAS]_0$ (mg/L)*	*% Removal*	*% F^-*	*Kinetics*	*References*
	Si/BDD	(RDE) Si/BDD/Pt wire/Hg/Hg_2SO_4	NA/NA	T = 22°C, 15 mL, 3,000 rpm, 0.915 g/L $NaClO_4$, 5 mA/cm^2	20	200	50[a]	NA	Zeroth-order	Carter and Farrell (2008)
	Monopolar electrodes	(Flow through) Si/ BDD/Pt wire/ none	25/0.3	T = 22°C, 2 L, pH 4, 0.1 L/min, 1.22 g/L $NaClO_4$, 20 mA/ cm^2	30	200	>99[a]	NA	Pseudo first-order	Carter and Farrell (2008)
	Ti/RuO_2/stainless steel	Ti/RuO_2/NA/NA	100/1.6	Room temperature, AFFF-impacted groundwater, 500 mg/L Na_2SO_4, recirculation 1.5 mL/min, 200 W	8	90	>90[a]	98	First-order	Schaefer *et al.* (2015)
PFOS, PFBS, PFHxS	Nb/BDD	Nb/BDD/NA/NA	35/2	T = 22–26°C, 1,000 mL, 300 rpm, synthetic groundwater, 14.2 g/L Na_2SO_4, 2.28 mA/cm^2	43	15 PFOS, 2.9 PFBS, 11 PFHxS	98 PFOS, 45 PFBS, 91 PFHxS	66 (total)	Pseudo first-order	Trautmann *et al.* (2015)
PFBS, PFHxS	Si/BDD	Si/BDD/Ti sheet/ SCE	8.5/3	T = 32°C, 40 mL, pH 3, 1,500 rpm, 1.4 g/L $NaClO_4$, 23.24 mA/cm^2	1.67	34.2 PFBS, 45.6 PFHxS	67[a] PFBS, 85[a] PFHxS	50[a] PFBS, 65[a] PFHxS	Pseudo first-order	Zhuo *et al.* (2012)
PFBS	Si/BDD/NA	(RDE) Si/BDD/Pt wire/Hg/Hg_2SO_4	NA/NA	T = 22°C, 350 or 600 mL, pH 5, 3,000 rpm, 1.22 g/L $NaClO_4$, 10 mA/cm^2	1.5	120	>97[a]	91[a]	Zeroth-order	Liao and Farrell (2009)

[a] Estimated from figure in article.

[b] Also tested Ti/SnO_2–Cl, Ti/SnO_2–Br, Ti/SnO_2–I, and Ti/SnO_2–Sb.

BDD, boron-doped diamond; RDE, rotating disk electrode; SCE, saturated calomel electrode.

TABLE 4. DIRECT PHOTOLYSIS OF PER- AND POLYFLUOROALKYL SUBSTANCES

Compound	*λ (nm)*	*Gas*	*Conditions*	*Time (h)*	*[PFAS]₀ (mg/L)*	*% Removal*	*% F⁻*	*Kinetics*	*Energy, W (light intensity, mW/cm²)*	*References*
Direct photolysis of perfluoroalkyl compounds										
PFOA	254 and 185	N_2	T = 23°C, 1.4 L pH 5.998	3	1	87.3	20.5	Pseudo-first-order	20 (4.9)	Giri *et al.* (2012)
	254 and 185	N_2	T = 23°C, 300 rpm pH 5.52–5.71	4	0.5	98.6	28[a]	First-order	20 (NA)	Giri *et al.* (2011)
	254 and 185	Air	NA	2	4.14	87	25	Pseudo-first-order	23 (62–69)	Cao *et al.* (2010)
	185	O_2	T = 25°C, 1,000 mL, 0.407 g/L $K_2S_2O_8$	2	25	87.4	40[a]	Pseudo-first-order	23 (NA)	Chen and Zhang (2006)
	185	N_2	T = 40°C, 800 mL, pH 3.7	2	41.4	61.7	17.1	Pseudo-first-order	15 (NA)	Chen *et al.* (2007)
PFOS	254	N_2	T = 36–46°C, 750 mL	240	20	68	71	Pseudo-first-order	32 (3.73)	Yamamoto *et al.* (2007)
	254 and 185	N_2	T = 25°C, 400 mL, pH 12.5	3	10	NA	32.8	Pseudo-first-order	23 (0.24, 185 nm) (1.65, 254 nm)	Jin and Zhang (2015)
Branched	254 and 185	N_2	T = 25°C, 400 mL, pH 12.5	3	3.25	>99[a]	95.4	Pseudo-first-order	23 (0.24, 185 nm) (1.65, 254 nm)	Jin and Zhang (2015)
Linear	254 and 185	N_2	T = 25°C, 400 mL, pH 12.5	72	NA	70[a]	NA	First-order	23 (0.24, 185 nm) (1.65, 254 nm)	Jin and Zhang (2015)
PFPeA, PFPrA, PFBA	220–460	O_2	T = 25°C, 22 mL, pH 1.5, 0.49 MPa	24	17770 PFPeA, 11039 PFPrA, 14405 PFBA	24.3 PFPeA, 16.1 PFPrA, 16.3 PFBA	12.1 PFPeA, 9.55 PFPrA, 9.66 PFBA	Pseudo-first-order	200 (15.2)	Hori *et al.* (2007b)
Direct photolysis of polyfluoroalkyl compounds										
4:2 FTUCA	220–460	O_2	T = 25°C, 20 mL, pH 2.0–3.5, 0.49 MPa	3	175	92	11.4	Pseudo-first-order	200 (41)	Hori *et al.* (2007a)
Environmental matrices										
PFOA	254 and 185	N_2	T = 23°C, pH 7.82, Neya River water	3	1	37.0	3.1	Pseudo-first-order	20 (4.9)	Giri *et al.* (2012)
			T = 23°C, pH 7.76, Tap water	3	1	56.9	10.6	Pseudo-first-order	20 (4.9)	Giri *et al.* (2012)

[a] Estimated from figure in article.

TABLE 5. PHOTOCATALYSIS AND OXIDATION OF PER- AND POLYFLUOROALKYL SUBSTANCES

Compound	Catalyst (g/L)	λ (nm)	Gas	Conditions	Time (h)	$[PFAS]_0$ (mg/L)	% Removal	% F^-	Kinetics	Energy, W (light intensity, mW/cm^2)	References
Photocatalysis of perfluoroalkyl compounds											
PFNA	TiO_2 RdH (0.66)	254	O_2	T = 25°C, pH 0.9–1.1, 15 g/L $HClO_4$	7	50	99	NA	Pseudo-first-order	16 (0.45)	Panchangam *et al.* (2009a)
PFOA	TiO_2 RdH (0.66)	254	O_2	T = 25°C, pH 1.5–1.3, 7.5 g/L $HClO_4$	7	50	86	NA	Pseudo-first-order	16 (0.45)	Panchangam *et al.* (2009a)
	P25 TiO_2 nanoparticles (1.5 wt%)	254	NA	T = 25°C, 100 mL	4	2,070	nd[a]	21[a]	Pseudo-first-order	NA (600)	Ochiai *et al.* (2011b)
	P25 TiO_2 (0.66)	310–400	NA	T = 30°C	6	1,656	30	22	Pseudo-first-order	500 (7.5)	Sansotera *et al.* (2014)
	P25 TiO_2 (0.66)	315–400	NA	T = 30°C	9	1,656	32	29	Pseudo-first-order	500 (9.5)	Gatto *et al.* (2015)
	TiO_2/Ni-Cu (Sheet)	254	N_2	250 mL, 14.2 Na_2SO_4	6	25	nd	85.6	NA	23 (NA)	Chen *et al.* (2006)
	10:1 TiO_2:MWCNT (1.6)	365	Air	T = 23°C, 250 mL, pH 2.0, 5.0, 11.0[b]	8	30	nd, 94, 58[b]	NA	NA	300 (NA)	Song *et al.* (2012)
	Fe-TiO_2 (0.5)	254	NA	T = 25°C, pH 5.0	12	50	69	9	Pseudo-first-order	400 (120,000 lux)	Chen *et al.* (2015)
	Cu-TiO_2 (0.5)	254	NA	T = 25°C, pH 5.0	12	50	91	19	Pseudo-first-order	400 (120,000 lux)	Chen *et al.* (2015)
	β-Ga_2O_3 (0.5)	254	N_2	T = 23°C, 100 mL, pH 4.8, 9.5 g/L $S_2O_3^{2-c}$	3	31	98.8	31.6	First-order	15 (NA)	Zhao *et al.* (2012)
	β-Ga_2O_3 (0.5, sheaf-like)	254	O_2	T = 25°C, 150 mL, pH 4.7	3	0.5	nd	61	Pseudo-first-order	14 (NA)	Shao *et al.* (2013)
	Fe(III) [0.02 $Fe_2(SO_4)_3$]	254	O_2	T = 20–25°C, 500 mL, pH 3.5–4.0	4	20	78.9	38.7	First-order	23 (NA)	Wang *et al.* (2008)
	Fe(III) [0.02 $Fe_2(SO_4)_3$]	254	N_2	T = 20–25°C, 500 mL, pH 3.5–4.0	4	20	40	13.4	First-order	23 (NA)	Wang *et al.* (2008)
	Fe(III) [0.02 $Fe_2(SO_4)_3$]	254	Air	T = 20–25°C, 500 mL, pH 3.5–4.0	4	20	73.6	35.1	First-order	23 (NA)	Wang *et al.* (2008)
	UV-Fenton [0.112 Fe(II)]	254	Air	200 mL, pH 3.0, 1.02 g/L H_2O_2	5	8.28	98[a]	52[a]	2 stage	9 (NA)	Tang *et al.* (2012)
	UV-Fenton [0.224 Fe(III)]	254	Air	200 mL, pH 3.0, 1.36 g/L H_2O_2	24	8.28	NA	26	2 stage	9 (NA)	Tang *et al.* (2012)
	Catalyzed H_2O_2 propagations [0.2 Fe(III)]	None	NA	T = 20°C, 40 mL, pH 3.5, 34 g/L H_2O_2	2.5	0.1	89	NA	NA	None	Mitchell *et al.* (2014)
	VUV-Fe(III) [0.005 $FeCl_3 \cdot 6H_2O$]	254 and 185	None	Room temp., 500 mL, pH 3.41,	4	14.9	51.21	50	Pseudo-first-order	12 (NA)	Cheng *et al.* (2014)
	Indium oxide (In_2O_3, 0.5)	254	O_2	T = 25°C, 400 mL, pH 3.1–4.2	4	41.4	83.1	33.7	Pseudo-first-order	23 (NA)	Li *et al.* (2012b)
	Persulfate (0.407 $K_2S_2O_8$)	254 and 185	O_2	T = 25°C, 1,000 mL	2	25	92.6	68.2	Pseudo-first-order	23 (NA)	Chen and Zhang (2006)

(*continued*)

Table 5. (Continued)

Compound	Catalyst (g/L)	λ (nm)	Gas	Conditions	Time (h)	$[PFAS]_0$ (mg/L)	% Removal	% F^-	Kinetics	Energy, W (light intensity, mW/cm^2)	References
	Persulfate (13.5 $K_2S_2O_8$)	220–460	O_2	T = 25°C, 22 mL, pH 3.0–3.1, 0.48 MPa	4	559	nd	59.1	NA	200 (NA)	Hori *et al.* (2005)
	Aqueous periodate (0.11 $NaIO_4$)	254	Air	NA	2	4.14	70	17	Pseudo-first-order	23 (62–69)	Cao *et al.* (2010)
	Aqueous periodate (0.11 $NaIO_4$)	254 and 185	Air	NA	2	4.14	60	13	Pseudo-first-order	23 (62–69)	Cao *et al.* (2010)
	Aqueous carbonate (3.36 $NaHCO_3$)	254	NA	T = 25°C, pH 8.96–8.3, 0.075% H_2O_2	12	50	nd	82.3	Pseudo-first-order	400 (120,000 lux)	Phan Thi *et al.* (2013)
	Tungstic heteropolyacid (20)	220–460	O_2	T = 20–25°C, 22 mL, 0.48 MPa	24	559	99.6	70.7	NA	200 (NA)	Hori *et al.* (2004)
PFDA	Persulfate (0.027 $K_2S_2O_8$)	254 and 185	O_2	T = 30°C, 500 mL	6	51.4	nd	30	NA	23 (62–69)	Wang *et al.* (2010)
	Na_2S (0.001)	254 and 185	N_2	T = 30°C, 500 mL	6	51.4	nd	45[a]	NA	23 (62–69)	Wang *et al.* (2010)
	TiO_2 RdH (0.66)	254	O_2	T = 25°C, pH 0.9–1.0, 15 g/L $HClO_4$	7	50	nd	NA	Pseudo-first-order	16 (0.45)	Panchangam *et al.* (2009a)
PFPeA, PFPrA, PFBA	Fe^{3+} (2.62 $Fe_2(SO_4)_3 \cdot 7.5H_2O$)	220–460	O_2	T = 25°C, 22 mL, pH 1.5, 0.49 MPa	24	PFPeA 17770 PFPrA 11039 PFBA 14405	PFPeA 64.5 PFPrA 61.3 PFBA 49.9	PFPeA 69.2 PFPrA 58.4 PFBA 46.5	Pseudo-first-order	200 (15.2)	Hori *et al.* (2007b)
PFOS	Alkaline 2-propanol (3.63)	254	N_2	T = 38–50°C, 750 mL	240	20	92	NA	Pseudo-first-order	32 (3.73)	Yamamoto *et al.* (2007)
Photocatalysis of polyfluoroalkyl compounds											
4:2 FTUCA	Persulfate (3.38 $K_2S_2O_8$)	220–460	O_2	T = 25°C, 20 mL, pH 2.0–3.5, 0.49 MPa	3	175	nd	54.8	NA	200 (41)	Hori *et al.* (2007a)
	WO_3 (0.136) and $K_2S_2O_8$ (9.14)	385–470	Ar	T = 25°C, 22 mL	70	1,431	49[a]	7[a]	Pseudo-first-order	500 (0.79)	Hori *et al.* (2013a)
6:2 FTS	H_2O_2 (30%)	NA	NA	T = 20°C, 250 mL	1.3	45	95.8	30[a]	Pseudo-first-order	300 (NA)	Yang *et al.* (2014)
Environmental matrices											
PFOA	In_2O_3 (0.5)	254	O_2	T = 25°C, pH 4.0, secondary effluent, 8 mg/L–h O_3	4	41.4	80[a]	NA	NA	23 (NA)	Li *et al.* (2012b)
	β-Ga_2O_3 (0.5 Sheaf-like)	254 and 185	O_2	T = 25°C, 150 mL, pH 4.3, secondary effluent	3	0.5	nd	66	Pseudo-first-order	14 (NA)	Shao *et al.* (2013)

[a] Estimated from figure in article.
[b] % Removal matches order of pH values.
[c] Also tested other reductive reagents: CH_3OH and $C_2O_4^{2-}$.
nd, not detected (below detection limit).

Table 6. Advanced Reduction Processes for Destruction of Perfluoroalkyl Substances

Compound	*Catalyst (g/L)*	*λ (nm)*	*Gas*	*Conditions*	*Time (h)*	*$[PFAS]_0$ (mg/L)*	*% Removal*	*% F^-*	*Kinetics*	*Energy, W (light intensity, $mW \cdot cm^{-2}$)*	*References*
PFOA	Sodium sulfite (2.5 SO_3^{2-})	254	N_2	T = 25°C, 200 mL, pH 10.3	6	8.28	NA	68.6	NA	10 (NA)	Song *et al.* (2013)
	Sodium sulfite (NA SO_3^{2-})	254 or 311	N_2	NA	20	NA	0–10	NA	NA	NA	Vellanki *et al.* (2013)
	Dithionite (NA $S_2O_4^{2-}$)	254 or 311	N_2	NA	20	NA	0–10	NA	NA	NA	Vellanki *et al.* (2013)
	Aqueous iodide (1.66 KI)	254	Ar	pH 6.0–8.0	2.5	10	15[a]	1.4	Pseudo-first-order	8 (NA)	Park *et al.* (2009)
	Aqueous iodide (0.05 KI)	254	N_2	T = room temp. 740 mL, pH 9.0	6	10.35	93.9	76.8	First-order	15 (NA)	Qu *et al.* (2010)
	Aqueous iodide (0.04 KI)	254	N_2	T = room temp. pH 10.0	6	10.35	nd[a]	69.8	Pseudo-first-order	15 (NA)	Qu *et al.* (2014)
	Aqueous iodide (0.1 KI)	254	N_2	T = 40°C, 740 mL, pH 9, 0.15 g/L NaCl	6	8.28	nd[a]	80.91	NA	15 (NA)	Zhang *et al.* (2014)
PFHxA, PFBA, PFOS, PFHxS, PFBS	Aqueous iodide (1.66 KI)	254	Ar	T = room temp. pH 6.0–8.0	2.5	10	5[a], 15[a], 35[a], 15[a], 5[a]	2.9, 3.1, 17, 7.1, 1.9	Pseudo-first-order	8 (NA)	Park *et al.* (2009)
Environmental matrices											
PFOA, PFHpA, PFPeA, PFHxA, PFBA, PFPrA	Aqueous iodide (0.05 KI)	254	N_2	Wastewater from fluorochemical plant in Jiangsu Province, China	12	6, 0.4, 0.05, 0.04, 0.02, 0.04	95.9, 62.0, 49.7, 41.5, 44.3, 45.7	NA	NA	15 (NA)	Qu *et al.* (2010)

[a]Estimated from figure in article.

TABLE 7. THERMAL AND NONTHERMAL DEGRADATION OF PER- AND POLYFLUOROALKYL SUBSTANCES

Compound	*Temp (°C)*	*Additive (g/L)*	*Conditions*	*[PFAS]$_0$ (mg/L)*	*Time (h)*	*% Removal*	*% F$^-$*	*Kinetics*	*References*
High-voltage electric discharge									
PFOA	20	NA	10 mL, DC current = 7 mA, O_2 gas (0.8 mL/s)	240	1.7	NA	95	NA	Yasuoka *et al.* (2010)
	22	NA	20 mL, DC current = 10 mA, O_2 gas (1.8 mL/s)	65	2.5	nd[a]	NA	First-order	Takeuchi *et al.* (2014)
	80	27 $K_2S_2O_8$	200 mL, DC current = 10 mA, O_2 gas (1.8 mL/s)	39	3	nd[a]	NA	First-order	Takeuchi *et al.* (2014)
	NA	NA	20 mL, DC current = 10 mA, O_2 gas (1.8 mL/s)	41.4	3	98	94.5	NA	Hayashi *et al.* (2015)
PFOS	25	NA	50 mL, DC current = 7 mA, O_2 gas (1.8 mL/s)	50	4	NA	55	NA	Yasuoka *et al.* (2011)
	NA	NA	20 mL, DC current = 10 mA, O_2 gas (1.8 mL/s)	60	8	nd	70	NA	Hayashi *et al.* (2015)
Forafac 1110	NA	3 TiO_2 (rutile, $SA_{specific}$ >50 m^2/g)	NTP, 180 mL, pH 2.5–6.9, energy = 100 W	200	1	97	21	Pseudo-first-order	Marouf-Khelifa *et al.* (2008)
	NA	0.2 TiO_2 (anatase, $SA_{specific}$ >10 m^2/g)	NTP, 180 mL, pH 2.4–4.8, energy = 100 W	200	1	97.5	28	Pseudo-first-order	Marouf-Khelifa *et al.* (2008)
	NA	0	NTP, 180 mL, pH 1.4–3.87, energy = 100 W	200	6	96.7	49	Pseudo-first-order	Marouf-Khelifa *et al.* (2008)
Incineration									
APFO (FC-143)	196, 206, 215, 224, 234	NA	^{19}F NMR with 10 mm probe	67 mg	0.66, 0.32, 0.22, 0.08, 0.07	50	NA	First-order	Krusic and Roe (2004)
Thermal chemical									
PFOA	85	2.4 $Na_2S_2O_8$	40 mL, pH 7.1	0.2	30	93.5	43.6	Pseudo-first-order	Liu *et al.* (2012a)
	40	48 $Na_2S_2O_8$	1,000 mL, pH 2.5	100	72	nd[a]	67.1	Pseudo-first-order	Lee *et al.* (2012b)
PFNA, PFOA, PFHpA, PFHxA, PFPeA	80	13.5 $K_2S_2O_8$	10 mL, 0.78 MPa synthetic air	141, 155, 136, 113, 99	6	nd, nd, nd, nd, nd	88.9, 77.5, 75.7, 82.8, 89.7	Pseudo-first-order	Hori *et al.* (2008a)
PFOS	900	$Ca(OH)_2$	Heat rate 20°C /min	8.65 mg	0.5	NA	90	NA	Wang and Shih (2011)
Microwave-hydrothermal									
PFOA	90	1.19 $Na_2S_2O_8$	18 psi, 50 mL, pH 2, energy = 70 W	105	4	79.1	70[a]	Pseudo-first-order	Lee *et al.* (2009)
	90	1.19 $Na_2S_2O_8$, 0.2 zero-valent iron	40 psi, 50 mL	100	2	67.6[a]	22.5[a]	Pseudo-first-order	Lee *et al.* (2010)
	90	2.7 $Na_2S_2O_8$	40 psi, 50 mL, pH 2.3, energy = 70 W	105	4	85.7	32[a]	Pseudo-first-order	Lee *et al.* (2012a)

(*continued*)

TABLE 7. (CONTINUED)

Compound	*Temp (°C)*	*Additive (g/L)*	*Conditions*	*$[PFAS]_0$ (mg/L)*	*Time (h)*	*% Removal*	*% F^-*	*Kinetics*	*References*
Subcritical									
PFOS	350	54 Fe powder	23.3 MPa, 10 mL, Argon-saturated solution	186	6	>99	51.4	Pseudo-first-order	Hori *et al.* (2006)
	350	62 Zn powder	23.3 MPa, 10 mL, Argon-saturated solution	186	6	76.9	18.5	NA	Hori *et al.* (2006)
	350	61 Cu powder	23.3 MPa, 10 mL, Argon-saturated solution	186	6	15.3	6.78	NA	Hori *et al.* (2006)
	350	25 Al powder	23.3 MPa, 10 mL, Argon-saturated solution	186	6	6.4	0.05	NA	Hori *et al.* (2006)
	350	Antireflective coating agent	23.3 MPa, 10 mL, Argon-saturated solution	55.9	6	NA	46.7	NA	Hori *et al.* (2006)
PFHxS	350	54 Fe powder	23.3 MPa, 10 mL, Argon-saturated solution	146	6	95.33	52.9	NA	Hori *et al.* (2006)
	350	54 Fe powder	22 MPa, 8 mL, pH 4.3–6.1, Argon-saturated solution	150.4	6	84.7	30.2	NA	Hori *et al.* (2008b)
PFBS	350	54 Fe powder	23.3 MPa, 10 mL, Argon-saturated solution	110	6	96.34	62.3	NA	Hori *et al.* (2006)
Nafion NRE-212	350	54 Fe powder	17.6 MPa, 10 mL, Argon-saturated solution	2,980	6	nd	63.9	NA	Hori *et al.* (2010)
$[(C_4F_9SO_2)_2N]^-$	344	54 Fe powder	16.5 MPa, 10 mL, Argon-saturated solution	454	6	98.65	69	NA	Hori *et al.* (2013b)
$^-O_3SC_3F_6SO_3^-$	350	54 Zerovalent iron	17.4 MPa, 10 mL, Argon-saturated solution	456	6	nd	70	NA	Hori *et al.* (2015)
Supercritical									
PFOA	50	1008 Nitric acid	20.3 MPa, 12 mL, 0.5–1 mL/min CO_2	0.83	0.67	80	NA	NA	Chen *et al.* (2012)
PFOS	50	1008 Nitric acid and 20% Methanol	20.3 MPa, 12 mL, 0.5–1 mL/min CO_2	0.83	0.67	55	NA	NA	Chen *et al.* (2012)
PFHxS	380	54 Fe powder	26.4 MPa, 8 mL, pH 4.3–6.1, argon-saturated solution	296.5	6	94.77	47.1	NA	Hori *et al.* (2008b)
$^-O_3SC_3F_6SO_3^-$	380	70 FeO	0.6 MPa, 10 mL, Argon-saturated solution	0.46	6	nd	89	NA	Hori *et al.* (2015)
Environmental matrices									
PFOS	400	0.3 g $Ca(OH)_2$	0.7 g WWTP Dried Sludge (Hong Kong)	0.016–0.256 g	0.25	NA	70	NA	Wang *et al.* (2013)
FTOH-based acrylic polymer	1,000	NA	1.88 cm^3, residence time = 2 s, air flow = 0.94 mL/s	1.6 mg	NA	99.9	NA	NA	Yamada *et al.* (2005)

[a] Estimated from figure in article.

Table 8. Sonolytic Degradation of Per- and Polyfluoroalkyl Substances

Compound	*Frequency (kHz)*	*Conditions*	*Time (h)*	*[PFAS]$_0$ (mg/L)*	*% Removal*	*Kinetics*	*Half life (min)*	*Power density (W/L)*	*References*
PFOA	200	T = 20°C, 60 mL, pH 3.5–4.8, Argon	1	10	85	Pseudo-first-order	22	NA	Moriwaki *et al.* (2005)
	354	T = 10°C, 600 mL, pH 7.0–8.0, Argon	2	0.1	NA	Pseudo-first-order	16.9	250	Vecitis *et al.* (2008b)
	358	T = 10°C, 600 mL, Argon	3	0.01–82.8[b]	nd–70[a,b]	Pseudo-first-order (0.01–2.7 mg/L); Zero-order (14.5–82.8 mg/L)	NA	250	Vecitis *et al.* (2008a)
	612	T = 10°C, 600 mL pH 7.9, Argon	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2010)
	40	T = 25°C, 1,000 mL, pH 8.65, N_2 gas, 2.52 g/L $NaHCO_3$	4	50	nd	Pseudo-first-order	NA	150	Phan Thi *et al.* (2014)
PFOS	200	T = 20°C, 60 mL, pH 3.5–4.8, Argon	1	10	60	Pseudo-first-order	43	NA	Moriwaki *et al.* (2005)
	354	T = 10°C, 600 mL, pH 7.0–8.0, Argon	2	0.1	NA	Pseudo-first-order	25.7	250	Vecitis *et al.* (2008b)
	358	T = 10°C, 600 mL, Argon	3	0.01–100[b]	98–70[a,b]	Pseudo-first-order (0.01–7 mg/L); Zero-order (19.5–100 mg/L)	NA	250	Vecitis *et al.* (2008a)
	600	T = 10°C, 400 mL, pH 6.2, VUV (185 nm, 27 mW/cm^2), Air	4	10	nd[a]	Pseudo-first-order	NA	250	Yang *et al.* (2013)
	612	T = 10°C, 600 mL, pH 7.9, Argon	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2010)
AFFF FC-600 PFOS	505	T = 10°C, 400 mL, Argon	2	1.10	95[a]	Exponential decay	NA	187.5	Vecitis *et al.* (2010)
PFHxA	358	T = 10°C, 600 mL, pH 7, Argon	2	0.10	90	Pseudo-first-order	16.8	250	Campbell *et al.* (2009)
PFHxS	358	T = 10°C, 600 mL, pH 7, Argon	2	0.10	86	Pseudo-first-order	23.2	250	Campbell *et al.* (2009)
AFFF FC-600 PFHxS	505	T = 10°C, 400 mL, Argon	2	0.21	40[a]	NA	NA	187.5	Vecitis *et al.* (2010)
PFBA	358	T = 10°C, 600 mL, pH 7, Argon	2	0.10	57	Pseudo-first-order	57.2	250	Campbell *et al.* (2009)
PFBS	358	T = 10°C, 600 mL, pH 7, Argon	2	0.10	66	Pseudo-first-order	42.3	250	Campbell *et al.* (2009)
AFFF FC-600 PFBS	505	T = 10°C, 400 mL, Argon	2	0.08	40[a]	NA	NA	187.5	Vecitis *et al.* (2010)
NFDOHA	28	T = 28°C, 15 L/min, 6 L, 2.7 g/L $K_2S_2O_8$, argon	24	15	98	Pseudo-first-order	NA	25	Hori *et al.* (2012)

(*continued*)

Table 8. (Continued)

Compound	*Frequency (kHz)*	*Conditions*	*Time (h)*	*[PFAS]$_0$ (mg/L)*	*% Removal*	*Kinetics*	*Half life (min)*	*Power density (W/L)*	*References*
Environmental matrices									
PFOA									
Groundwater (Oakdale, MN)	612	T = 11.7°C, pH 7.9, 600 mL, Argon, TOC 1.5 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2010)
Landfill Groundwater (Oakdale, MN)	354	T = 10°C, pH 6.9–7.9, 600 mL, Argon, TOC 20 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2008)
Landfill Groundwater (Oakdale, MN)	354	T = 10°C, pH 6.9–7.9, 600 mL, 2.5% v/v O_3, Argon, TOC 20 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2008)
PFOS									
Groundwater (Oakdale, MN)	612	T = 11.7°C, pH 7.9, 600 mL, Argon, TOC 1.5 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2010)
Landfill Groundwater (Oakdale, MN)	354	T = 10°C, pH 6.9–7.9, 600 mL, Argon, TOC 20 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2008)
Landfill Groundwater (Oakdale, MN)	354	T = 10°C, pH 6.9–7.9, 600 mL, 2.5% v/v O_3, Argon, TOC 20 mg/L	2	0.10	NA	Pseudo-first-order	NA	250	Cheng *et al.* (2008)

[a] Estimated from figure in article.

[b] Min–max conditions; ranges for decomposition and defluorination ratio correspond to the minimum and maximum, respectively.

the importance of electrostatic interactions, ~90% PFOS removal was observed on negatively charged silica, regardless of changes in pH, ionic strength, and Ca^{2+} concentration (Tang *et al.*, 2010). Hydrophobic interactions likely occur between the perfluoroalkyl tail and the hydrophobic surfaces of the sorbent, especially for longer C–F chain lengths (Higgins and Luthy, 2006; Zhou *et al.*, 2010a; Zhang *et al.*, 2011, 2013a; Du *et al.*, 2015). The addition of each extra CF_2 moiety increases the hydrophobicity of PFASs, resulting in increased sorption of longer-chain PFASs. Longer chains may also outcompete shorter chains in sorption processes (Xiao *et al.*, 2011; Du *et al.*, 2015). In addition, perfluorosulfonic acids (PFSAs), such as PFOS, contain one more C–F bond compared with the corresponding perfluorocarboxylic acid (PFCA), such as PFOA, resulting in stronger hydrophobic properties and increased sorption of PFSAs compared with PFCAs (Zhou *et al.*, 2010b).

Sorption processes: activated carbon

Granular activated carbon (GAC) and PAC are two of the most studied adsorbents for PFASs. Activated carbons (ACs) have a porous structure with strong heterogeneous surfaces and has been used to sorb various compounds due to its low cost and versatility (Marsh and Reinoso, 2006). ACs are produced from almost any carbonaceous materials, and current research has analyzed PACs, BioNuchar, activated carbon fibers (ACF15, ACF20, ACF25), Ambersorb563, bamboo-derived AC (BAC), and several commercial GACs, including Filtrasorb (F) 300, F400, F600, URV-MOD1, 1240C, 43765, and 43767 (Ochoa-Herrera and Sierra-Alvarez, 2008; Qu *et al.*, 2009; Yu *et al.*, 2009; Appleman *et al.*, 2013; Du *et al.*, 2014, 2015; Schuricht *et al.*, 2014; Pramanik *et al.*, 2015; Zhi and Liu, 2015).

GACs were most effective at sorbing longer alkyl chain lengths with more C–F bonds (e.g., PFOS > PFBS and PFOS > PFOA) (Ochoa-Herrera and Sierra-Alvarez, 2008; Carter and Farrell, 2010; Senevirathna *et al.*, 2010). Furthermore, GACs with higher surface areas and larger micropores to facilitate diffusion of PFASs, such as BAC, are more advantageous (Du *et al.*, 2015). Similarly, PAC improves PFAS sorption compared with GAC due to its larger surface area and increased sorption sites (Ochoa-Herrera and Sierra-Alvarez, 2008; Qu *et al.*, 2009; Yu *et al.*, 2009; Bao *et al.*, 2014; Pramanik *et al.*, 2015). For example, PAC was combined with a membrane bioreactor (PAC-MBR) consisting of synthetic wastewater and seed sludge from a municipal wastewater treatment plant and effectively removed 94.8% PFOS and 90.6% PFOA only when PAC was added (Yu *et al.*, 2014). In addition, AC absorption has been recommended by the New Jersey Drinking Water Quality Institute Treatment Subcommittee (Cummings *et al.*, 2015) as one of the most effective treatment options for removal of PFNA, PFOA, and PFOS. Municipalities and industrial treatment facilities in the United States and Europe have carried out few AC absorption case studies with successful performance cited by the recommendation.

Sorbent properties strongly influence the sorption of PFASs. Sorbents made of synthetic polymers were observed to be more effective than those made of natural materials and followed the perfluoroalkyl acid (PFAA) sorption efficiency trend of ACF20 (activated carbon fiber) > AquaNuchar > Ambersorb 563 > F400 or 1240C > WVB or BioNC (BioNuchar) (Zhi and Liu, 2015). Sorbent macroscopic size was also the dominant factor controlling adsorption uptake. In addition to size, carbon surface chemistry [e.g., basicity and pH point of zero charge (pH_{pzc})] also affected uptake, but acidity and oxygen content of the sorbents did not.

Sorption processes: ion exchange

Ion exchange can be more efficient than GAC (Lampert *et al.*, 2007; Carter and Farrell, 2010; Senevirathna *et al.*, 2010; Schuricht *et al.*, 2014). Lampert *et al.* (2007) observed greater removal with six different ion-exchange resins compared with four different other methods: AC adsorption, adsorption onto calcium fluoride solids, evaporation, and liquid–liquid extraction. For example, US Filter A-714 removed PFOS to <1 mg/L ($PFOS_0 = 151$ mg/g·resin) and PFOA to 13 mg/L ($PFOA_0 = 686$ mg/g·resin). Shorter removal times have been observed with Amberlite IRA-458 (Carter and Farrell, 2010), Amberlite IRA-400 (Yu *et al.*, 2009; Senevirathna *et al.*, 2010), Dow Marathon A (Senevirathna *et al.*, 2010), Amberlite XAD-7HP (Xiao *et al.*, 2012), AS-F860 (Schuricht *et al.*, 2014), AS-F500 (Schuricht *et al.*, 2014), and AW-F100 (Schuricht *et al.*, 2014).

Ion-exchange resins need to be regenerated. This can be done with a small percentage of NaCl or NaOH and a large percentage of methanol (Senevirathna *et al.*, 2010; Xiao *et al.*, 2012; Du *et al.*, 2015). For example, Du *et al.* (2015) used 1% NaCl in 70% methanol mixture and reused Amberlite IRA 67 for at least five cycles with very little decrease in removal effectiveness for PFOA, perfluoroheptanoic acid (PFHpA), and perfluorohexanoic acid (PFHxA). A larger amount of methanol would be needed to remove longer-chain PFASs (Xiao *et al.*, 2012). To improve removal of PFASs, ion-exchange columns could be run in series with regeneration occurring in every other column. In addition, other factors that need to be considered include the pH_{pzc} of the resin and pH of the solution. For example, Xiao *et al.* (2012) observed increased PFOS removal for Amberlite XAD-7HP ($pH_{pzc} = 6.2$) with decreasing pH (4.8–7.8) due to increased electrostatic attraction.

Sorption processes: other sorbents

PFAS sorption has been studied with 12 other sorbents: molecular imprinted polymers (MIPs) (Yu *et al.*, 2008; Deng *et al.*, 2009; Zhang *et al.*, 2013a), cationic/anionic surfactants (Pan *et al.*, 2009), multiwalled carbon nanotubes (MWCNTs) (Li *et al.*, 2011; Kwadijk *et al.*, 2013), modified organo-montmorillonite (Zhou *et al.*, 2013a), silica-based adsorbents (Zhou *et al.*, 2013b), black carbon (Chen *et al.*, 2009), magnetic mesoporous carbon nitride (MMCN) (Yan *et al.*, 2013, 2014), polymeric adsorbents (Schuricht *et al.*, 2014), mesoporous molecular sieves (Nassi *et al.*, 2014), metal-organic frameworks (Liu *et al.*, 2015), electrocoagulation (Lin *et al.*, 2015), and permanently confined micelle arrays (PCMAs) (Wang *et al.*, 2014a).

Only a few of these sorbents are promising, such as MWCNTs, MIPs, MMCNs, and sorption with electrochemical assistance. MWCNTs need to be combined with electrochemical assistance (Li *et al.*, 2011) or electrospun nanofibrous membranes (Dai *et al.*, 2013) to have efficient removal, but regeneration of MWCNTs requires 90°C to achieve only 85% PFOA and PFOS release. Similarly, MIPs require 40°C and NaOH/acetone (Yu *et al.*, 2008) and can only be used for targeted PFASs due to polymer-specific manufacturing.

The most favorable sorption technology for *Other Sorbents* is electrochemical assistance combined with zinc sheet (anode) and stainless steel (cathode) in an electrocoagulation reactor (Lin *et al.*, 2015). PFOA and PFOS were removed to below detection limits within 20 min, and sorption equilibrium was reached within 10 min due to production of zinc hydroxide flocs. This method may be a cost-effective and safe adsorption removal technology for PFASs since it is currently being utilized at wastewater treatment facilities and only needs simple easy-to-operate equipment and low maintenance costs. The energy consumption was 0.18 Wh/L and low concentrations (0.88 mg/L) of residual zinc ions were detected (US EPA drinking water limit for zinc ions is 5 mg/L). However, electrocoagulation can result in formation of chlorinated organic compounds (e.g., trihalomethanes) and bad taste and odor (Mollah *et al.*, 2004).

Sorption processes: summary

Sorption of PFASs has been shown to be an effective removal method, especially when using AC or ion exchange. For more information on PFAS sorption techniques, Du *et al.* (2014) have published a detailed review. While sorption can be cost-effective, treatment processes should consider sorbent regeneration and further destruction of sorbed PFASs. For example, AC can only be moderately regenerated using methanol or ethanol, and subsequent reuse can result in decreased removal percentages (Senevirathna *et al.*, 2010; Punyapalakul *et al.*, 2013; Chularueangaksorn *et al.*, 2014; Du *et al.*, 2015).

More research is needed on (1) sorption of other PFASs and PFAS mixtures and (2) the influence of environmental matrices (e.g., inorganic ion concentration, organic matter content), mixtures of PFASs, and cocontaminants. For example, environmental matrices, such as soil, sediment, and sludge from wastewater treatment plants, can impact PFAS sorption treatment methods due to several factors, including compression of the adsorbent's electrical double layer, reduction in electrostatic repulsion between adsorbent and PFASs, and formation of bridges with cations between negatively charged groups and PFASs (Zhou *et al.*, 2010b; Yu *et al.*, 2012; Kwadijk *et al.*, 2013; Zhang *et al.*, 2013a; Du *et al.*, 2015; Millinovic *et al.*, 2015; Wang *et al.*, 2015). In addition, competitive adsorption between PFOA, PFHxA, and PFHpA has been shown to negatively affect sorption of each PFCA onto BAC and Amberlite IRA 67 (Du *et al.*, 2015).

Advanced oxidation processes

AOPs that have been tested for PFAS removal include electrochemical oxidation, photolysis, and photocatalysis. During these processes, strong, oxidizing, and nonselective radicals are generated (including $^{\bullet}OH$, $O_2^{\bullet-}$, $SO_4^{\bullet-}$, and $CO_3^{\bullet-}$) that can attack a variety of xenobiotics, such as pharmaceuticals (Ikehata *et al.*, 2006), phenols and dyes (Ahmed *et al.*, 2011), and trinitrotoluenes (TNTs) (Ayoub *et al.*, 2010).

AOPs: electrochemical oxidation

Electrochemical oxidation destroys contaminants through two mechanisms: (1) direct anodic or (2) indirect. When contaminants are destroyed by direct anodic oxidation, contaminants will adsorb onto the anode surface and are destroyed by an electron transfer reaction. In indirect oxidation, contaminants are destroyed in solution by oxidation through strong oxidants generated by cathodic electrochemical reactions. This process has been used to treat many different contaminants, including phenols (Cañizares *et al.*, 2005), dyes (Chen *et al.*, 2003), and endocrine-disrupting chemicals (Murugananthan *et al.*, 2007). Electrochemical oxidation can have long life spans and is versatile, energy efficient, automated, and cost-effective (Jüttner *et al.*, 2000). It can also be used on different volumes of gases, liquids, and solids and is relatively easy and inexpensive to construct and operate electrodes. There are a wide variety of electrode materials, including Pt, IrO_2, and RuO_2, but for PFAS removal, researchers have studied boron-doped diamond (BDD) thin film, Ti/SnO_2, Ce/PbO_2, and Ti/RuO_2 (Table 3).

Degradation of PFASs through BDD anodes has been the most studied electrochemical method. PFASs will undergo direct anodic oxidation, resulting in one-carbon removal through decarboxylation pathways [Eqs. (S1)–(S8) in Supplementary Table S6] (Zhuo *et al.*, 2012). This pathway continuously repeats and shorter-chain PFASs, fluoride ions, and sulfate ions (PFSAs only) are produced. Hydroxyl radicals formed from water on the BDD anode can also help mineralize PFASs in solution to elemental or inorganic end products [Eqs. (S2) and (S4) in Supplementary Table S6]. Compared with other electrode materials, BDD has a higher oxygen evolution potential, allowing the formation of more hydroxyl radicals at low background currents (Zhu *et al.*, 2008; Panizza, 2010). In addition, hydroxyl radicals are weakly adsorbed to the BDD electrode and should not interfere with the initial PFAS reaction.

BDD thin film electrodes can effectively degrade PFOA, PFBA, PFHxA, perfluorodecanoic acid (PFDA), PFBS, perfluorohexanesulfonic acid (PFHxS), and PFOS under optimized conditions. For example, PFOA degraded by 97% (60% fluoride yield) within 2 h (Zhuo *et al.*, 2012). For PFCAs, the defluorination ratios increased with decreasing chain lengths, while defluorination ratios for PFSAs increased with increasing chain length. The degradation of PFSAs also led to formation of shorter-chain PFCAs [Eqs. (S4)–(S8) in Supplementary Table S6]. Similar results were observed using groundwater collected from a former fire service training ground (Trautmann *et al.*, 2015). Greater mineralization of PFOA occurred using the ultra-nanocrystalline boron-doped conductive diamond electrode (Urtiaga *et al.*, 2015). Other studies on BDD thin film electrodes were not as successful and took much longer to degrade PFOA (Carter and Farrell, 2008; Liao and Farrell, 2009; Ochiai *et al.*, 2011a, 2011c).

A major limitation to BDD thin film electrodes is the cost and difficulty of building BDD compared with other electrode materials (Panizza and Cerisola, 2009). Other studies have explored the use of Ti/SnO_2 anodes (Lin *et al.*, 2012b; Yang *et al.*, 2015), Ce-doped PbO_2 film electrodes (Niu *et al.*, 2012, 2013), and commercially available Ti/RuO_2 (Schaefer *et al.*, 2015). For example, 90.3% PFOA degraded (72.9%

fluoride yield) to shorter-chain PFCAs and fluoride when using Ti/SnO_2-Sb anode (Lin *et al.*, 2012b). However, the degradation efficiency decreased when pH increased, plate distance increased, or initial PFOA concentration increased. The service life of Ti/SnO_2 anodes is relatively short, but could be improved upon by doping with SnF_4 (F-doped Ti/SnO_2) (Yang *et al.*, 2015).

In general, electrochemical oxidation has some limitations. Production of toxic by-products may occur when treating PFAS-contaminated wastewater mixed with other harmful substances (Trautmann *et al.*, 2015), including chlorine gas, hydrogen fluoride, bromate, perchlorate, and adsorbable organic halides. Future research needs to focus on degradation of different PFASs, including polyfluoroalkyl compounds, over a range of concentrations and determine the degradation pathway. Furthermore, only two studies have observed PFAS destruction when using electrochemical oxidation in the presence of AFFF-impacted or PFAS-contaminated synthetic groundwater (Schaefer *et al.*, 2015; Trautmann *et al.*, 2015). More studies need to be conducted with environmental matrices to determine whether electrochemical oxidation is suitable for PFAS remediation.

AOPs: photolysis and photocatalysis

Photolysis and photocatalysis of PFASs involve the use of vacuum ultraviolet (VUV, 100–200 nm) or ultraviolet (UV, 200–400 nm) light (Tables 4 and 5). With the addition of a photocatalyst, such as titania or indium oxide, the ability to remove PFASs can be enhanced. Heterogeneous photocatalytic decomposition occurs when an energy difference is produced between the valence (VB) and conduction band (CB) with light exposure (Coronado *et al.*, 2013), allowing for oxidation–reduction processes to occur. Common products of photocatalysis are hydroxyl radicals, superoxide radicals, and secondary radicals of organic substrates.

For successful photocatalysis of PFASs, several conditions must be considered. The pH of the solution and the pH_{pzc} of the catalyst with respect to the pKa of the target PFASs are the two main important factors. Generally, the pH_{pzc} of the catalyst must be greater than the solution pH to allow for greater contact with PFASs. Other conditions include light wavelength and intensity, initial catalyst and PFAS concentrations, and water quality (e.g., the turbidity of water, total organic matter content, dissolved oxygen concentration, and natural water scavengers, such as bicarbonate). Current studies on photocatalysis of PFASs mainly utilize UV light, but VUV may also effectively decompose PFOA by stepwise radical formation, decarboxylation, HF elimination, and shorter-chain PFAS formation as described by Equations (S9)–(S13) in Supplementary Table S6. Natural sunlight in combination with iron and H_2O_2 or persulfate could effectively decompose PFOA (Liu *et al.*, 2013a). Future studies should optimize conditions for natural sunlight remediation of PFASs and determine whether VUV in combination with photocatalysts can decompose PFASs more efficiently.

In all photolysis and photocatalysis studies, decomposition of PFASs occurs in a stepwise manner. First, a PFAS radical is activated through exposure to (1) direct photolysis, (2) radicals and highly reactive intermediates, or (3) to a semiconductor material with an energy band gap [Eq. (S9) in Supplementary Table S6] (Chen *et al.*, 2007; Hori *et al.*, 2007b; Wang *et al.*, 2008; Song *et al.*, 2012; Tang *et al.*, 2012; Phan Thi *et al.*, 2013). More specific reactions that may take place with different photocatalysts are described in their respective sections. Products include shorter-chain PFASs, formic acid, fluoride ions, sulfate ions, and hydrogen. With longer reaction times, depending on the method used, PFASs can be degraded to low to nondetectable levels.

AOPs: direct photolysis

Direct photolysis of PFASs, mainly PFOA, has been studied over a wide range of different gases, wavelengths, and initial PFAS concentrations, but generally under ambient temperatures and acidic conditions. For example, more decomposition of PFOA occurred under oxygen gas compared with argon-saturated solution (Hori *et al.*, 2004). Direct photolysis of PFOA can be improved with the addition of VUV light (185 nm), accompanied by changes in gases, pH, and temperature. Based on the photon energy values of UV and VUV light and the average bond energy, the C–C bonds (bond energy = 347.0 kJ/mol) in PFOA are likely to be cleaved by both 254 nm (photon energy = 471.1 kJ/mol) and 185 nm (photon energy = 646.8 kJ/mol), whereas the C–F bonds (bond energy = 552.0 kJ/mol) are only likely to be cleaved by 185 nm (Giri *et al.*, 2011). However, direct photolysis of PFASs tends to have relatively low removal efficiencies and fluoride yields compared with other processes (Chen and Zhang, 2006; Giri *et al.*, 2011; Phan Thi *et al.*, 2013; Cheng *et al.*, 2014), and remediation strategies using direct photolysis must consider additional treatment methods.

Presence of oxygen could also play an important role in the direct photolysis of PFOA (Giri *et al.*, 2012; Jin *et al.*, 2014). Giri *et al.* (2012) tested various dissolved oxygen (0–36.4 mg/L) concentrations with about 90% VUV and 10% UV light. Less PFOA was decomposed with increasing DO levels. Jin *et al.* (2014) observed similar results for PFOS when using light-emitting 10% VUV and 90% UV. When using oxygen and air gas or nitrogen gas with H_2O_2, PFOS was not decomposed and there was insignificant fluoride ion production. These observations could indicate other chemical reactions that coexist with direct photolysis. The impact of dissolved oxygen on PFAS decomposition under direct photolysis could be attributed to the scavenging of hydrated electrons, which are formed during VUV water splitting. A detailed summary is discussed in *ARPs.*

The direct photolysis of four other perfluorinated substances [PFOS, perfluoropentanoic acid (PFPeA), perfluoropropanoic acid (PFPrA), PFBA] (Hori *et al.*, 2007b; Yamamoto *et al.*, 2007) and one polyfluorinated compound (4:2 fluorotelomer unsaturated carboxylic acid, FTUCA) (Hori *et al.*, 2007a) has been examined, mainly as control groups for experiments to analyze heterogeneous photocatalysis. Future studies should analyze whether the optimum conditions for PFOA decomposition could be used to decompose PFAS mixtures efficiently.

AOPs: titania (TiO_2) photocatalysis

Photocatalysis of PFOA using titania has been one of the most studied types of heterogeneous photocatalysis reactions. Current studies on PFOA decomposition utilize TiO_2 in anatase and rutile forms due to high band gap energies (3.2 and

3.0 eV, respectively) (Linsebigler *et al.*, 1995; Fujishima *et al.*, 2000; Carp *et al.*, 2004). Two commercially available TiO_2 materials have been used (RdH and P25), as well as home-made TiO_2 using the sol–gel process. One of the more successful studies used P25 TiO_2 nanoparticles with 254 nm exposure and observed almost complete PFOA degradation within 4 h based on detection capabilities of LC with a variable wavelength detector (Ochiai *et al.*, 2011b). However, higher wavelengths (315–400 nm) decreased PFOA degradation (Gatto *et al.*, 2015).

Other studies improved TiO_2 photocatalysis of PFOA by (1) combining with perchloric acid and an ultrasonic probe (Panchangam *et al.*, 2009a, 2009b) and (2) doping with iron:nobium (Fe:Nb), MWCNTs, Cu^{2+}, or Fe^{3+} (Estrellan *et al.*, 2009, 2010; Panchangam *et al.*, 2009a; Song *et al.*, 2012; Sansotera *et al.*, 2014; Chen *et al.*, 2015; Gatto *et al.*, 2015). Certain doped TiO_2 can be effective photocatalysts for PFOA decomposition due to increased lifetime of electron–hole pairs and adsorption of compounds to the catalyst surface (Hernández-Alonso, 2013). For example, a 10:1 ratio (TiO_2:MWCNT) was most effective for PFOA decomposition compared with other ratios (Song *et al.*, 2012). MWCNTs are stable (e.g., acidic and basic conditions) and can also accept electrons and reduce recombination between electron–hole pairs in TiO_2. Similarly, Cu-TiO_2 decomposed PFOA (91%) within 12 h, resulting in greater degradation compared with Fe^{3+}–TiO_2 and shorter-chain PFCAs (19% defluorination) (Chen *et al.*, 2015). Thus, while doped TiO_2 can greatly enhance PFOA decomposition, the type of dopant needs to be considered to have high removal and fluoride yields. Future studies should optimize current dopants for higher fluoride yields and consider testing other PFASs and varying conditions (e.g., lower energy lamps, different gases, and different wavelengths).

AOPs: other semiconductor material photocatalysis

Gallium oxide (β-Ga_2O_3) (Zhao *et al.*, 2012; Shao *et al.*, 2013) and indium oxide (In_2O_3) (Li *et al.*, 2012b) have also been studied for PFAS decomposition and have more potential than TiO_2. Gallium oxide has a wider bandgap, reduction potential, and pH_{pzc}, but it is more expensive than titania. Compared with TiO_2, indium oxide promotes faster conversion of PFASs to PFAS radical, but slower conversion of H_2O and hydroxy groups to hydroxyl radicals. Both semiconductor materials were able to decompose PFOA much faster than undoped TiO_2. For gallium oxide, within just 45 min, 100% PFOA degradation (based on the detection capabilities of a UPLC-MS/MS) with 61% defluorination yield was observed (Shao *et al.*, 2013). However, special, synthesized gallium oxide nanomaterial containing sheaf-like structures must be prepared since commercial gallium oxide decomposed only 38% PFOA in 3 h (Shao *et al.*, 2013). These results were similar to those described by Zhao *et al.* (2012) for non-nanomaterial gallium oxide. To improve the reaction with non-nanomaterial gallium oxide, a reductive additive at basic pH under N_2 gas is needed, such as $S_2O_3^{2-}$ (Zhao *et al.*, 2012).

AOPs: iron photocatalysis

Iron photolysis is also one of the most studied heterogeneous photocatalysis mechanisms for PFAS decomposition, including PFOA, PFPeA, PFPrA, and PFBA, since iron ions are abundant and relatively cheap. For PFAS decomposition, various iron sources have been used to represent both Fe^{2+} and Fe^{3+} ions and have been tested with 12–15% VUV (185 nm) (Cheng *et al.*, 2014) and H_2O_2 to produce Fenton reaction. Other metal ions were also tested for their ability to decompose PFOA, including Cu^{2+}, Mg^{2+}, Mn^{2+}, and Zn^{2+}, but could only decompose 4.2–7.4% PFOA within 4 h (Wang *et al.*, 2008).

Oxygen plays an important role in PFOA decomposition by iron photocatalysis, similar to direct photolysis, and more PFOA decomposition occurred with oxygen gas compared with air or nitrogen gas. When using Fe^{3+} ions [as $Fe_2(SO_4)_3$] with oxygen gas, 78.9% PFOA decomposed (38.7% defluorination yield) within 4 h compared with air and nitrogen gas (Wang *et al.*, 2008). The importance of oxygen and Fe^{3+} ions for iron photolysis of PFOA can be explained by a likely reaction pathway that involves formation of PFOA and Fe^{3+} complex and oxidation of Fe^{2+} by oxygen [Eqs. (S14)–(S17) in Supplementary Table S6]. Other Fe^{3+} ion sources, such as $Fe_2(SO_4)_3 \cdot 7.5H_2O$, $Fe_2(ClO_4)_3 \cdot 6H_2O$, and $FeCl_3 \cdot 6H_2O$, could affect PFAS degradation efficiencies (Hori *et al.*, 2007b).

Iron photocatalysis was greatly improved with the addition of hydrogen peroxide, producing UV-Fenton reactions. Generally, H_2O_2 will decompose Fe^{2+} ions without a light source present, but the reaction [Eqs. (S18)–(S22) in Supplementary Table S6] increases with UV-Vis irradiation, especially with the regeneration of Fe^{2+} ions (Chong *et al.*, 2010). Within 1 h, 87.9% PFOA was decomposed (25.8% defluorination yield) when Fe^{2+} ions (as $FeSO_4$) were used in conjunction with 1 g/L H_2O_2 (Tang *et al.*, 2012). Fe^{2+} ions were more effective than Fe^{3+} ions under UV-Fenton conditions.

PFAS decomposition through UV-Fenton process is a feasible and applicable AOP. With the addition of H_2O_2, PFASs undergo general photocalytic degradation when H_2O_2 is abundant. Then, the traditional Fenton reaction mechanism mentioned at the beginning of this section starts and continues to decompose PFASs (Tang *et al.*, 2012). Future studies should focus on optimizing pH, Fe^{2+} ion concentration, and H_2O_2 loading for a range of PFAS concentrations, PFAS mixtures, and environmental matrices.

Modified Fenton's reaction, known as catalyzed H_2O_2 propagation (CHP) [Eqs. (S23)–(S27) in Supplementary Table S6], can efficiently decompose PFOA (89% decomposed within 2.5 h) due to superoxide and hydroperoxide species (Mitchell *et al.*, 2014). Instead of UV, this method utilized a high concentration of H_2O_2 with the addition of initiators (e.g., soluble Fe(III), iron chelates, and minerals) to decompose xenobiotics (Watts and Teel, 2005). CHP has been successfully used for *in situ* chemical oxidation (ISCO) for over 10 years and has been used to treat industrial wastewater. In addition to PFOA degradation, Mitchell *et al.* (2014) could only detect F^- as the main degradation product [Eqs. (S23)–(S27) in Supplementary Table S6].

AOPs: activated persulfate ($S_2O_8^{2-}$) oxidation

Persulfate has been used to successfully decompose PFOA, PFDA, and 4:2 FTUCA. This strong oxidant (E° = 2.1 V) is highly soluble and can become activated and generate free sulfate radicals ($SO_4^{\bullet -}$, E° = 2.6 V) when exposed to UV light, transition metals, hydrogen peroxide, heat, etc. (Tsitonaki *et al.*, 2010). Sulfate radicals are considered harmless and can react with organics, water, or hydroxyl groups [Eqs. (S4) and (S28)–(S30) in Supplementary Table S6]. This increases the oxidation capabilities of persulfate ions toward PFASs since both sulfate and hydroxyl radicals will interact with PFASs [Eq. (S33) in Supplementary Table S6]. The reaction will then follow general stepwise decomposition [Eqs. (S9)–(S13) in Supplementary Table S6] (Lee *et al.*, 2009).

Five studies demonstrated that persulfate can decompose PFOA, PFDA, and 4:2 FTUCA to shorter-chain PFCAs and elemental components, such as F^-, under oxygen gas, various wavelengths, ambient temperatures, and acidic pH. For example, persulfate (50 mM) decomposed PFOA to nondetectable levels within 4 h (59.1% defluorination yield), and within 8 h, the fluoride yield increased to 73.8% (Hori *et al.*, 2005). A lamp requiring less energy (23 W vs. 200 W) was later applied successfully (Chen and Zhang, 2006). Persulfate ions could also decompose PFDA when using both UV and VUV light (Wang *et al.*, 2010). Other sources of sulfate radicals may be used, such as sulfide ions (as Na_2S). Sulfide ions under N_2 gas increased the defluorination yield to 45% due to the ability of S_2^- to scavenge species, such as H^+ and OH, and allowing for e_{aq}^- to act on PFDA (Wang *et al.*, 2010). Persulfate can also be used to decompose polyfluoroalkyl compounds such as 4:2 FTUCA within a few minutes (Hori *et al.*, 2007a), but less degradation was observed when using visible light combined with persulfate and tungsten trioxide (WO_3) (Hori *et al.*, 2013a).

Since persulfate has proven to be highly successful in decomposing PFOA, PFDA, and 4:2 FTUCA, future studies should determine if persulfate could be used for environmental matrices, especially as an ISCO oxidant.

AOPs: other UV-induced oxidation

Aqueous solutions (e.g., aqueous periodate and carbonate) with photolysis can be effective at decomposing PFASs. With photolysis, aqueous periodate (as $NaIO_4$) produces products such as IO_4^-, $IO_3^{\bullet}$, $^{\bullet}OH$, and $O^{\bullet -}$ (Cao *et al.*, 2010), while aqueous carbonate (as $NaHCO_3$) produces products such as $CO_3^{2-\bullet}$, HCO_3^{2-}, and $^{\bullet}OH$ (Phan Thi *et al.*, 2013). These radicals can react with PFASs and produce shorter-chain PFASs, F^- ions, CO_2, or SO_4^{2-}.

Under oxidizing conditions, aqueous periodate and carbonate can decompose PFOA, with more efficient decomposition occurring with aqueous periodate (Cao *et al.*, 2010). This difference may be due to scavenging of aqueous periodate by aquated electrons. While decomposition of PFOA by aqueous periodate was fast, higher temperatures were used (40°C). Comparatively, aqueous carbonate decomposed PFOA to nondetectable levels (82.3% defluorination yield) at ambient temperatures and basic pH (8.3–8.96), but at a longer reaction time (12 h) and with the addition of H_2O_2 (Phan Thi *et al.*, 2013).

Other UV-involved oxidation processes have not been as successful for PFAS decomposition, including tungstic heteropolyacid ($H_3PW_{12}O_{40} \cdot 6H_2O$) (Hori *et al.*, 2004) and alkaline 2-propanol (Yamamoto *et al.*, 2007), which required long reaction times (24 h and 10 days, respectively).

AOPs: summary

Several AOPs are successful in degrading PFASs, especially PFOA and PFOS. Electrochemical oxidation of PFASs may be a possible treatment method, but more research is needed on other electrode materials and on the influence of environmental matrices, PFAS mixtures, and cocontaminants. Photolysis, photocatalysis, activated persulfate oxidation, and other UV-induced oxidation are also promising treatment methods, but require similar research as electrochemical oxidation.

Advanced reduction processes

ARPs are a new treatment method that has successfully degraded other xenobiotics, including monochloroacetic acid (Li *et al.*, 2012a), vinyl chloride (Liu *et al.*, 2013c), and 1,2-dichloroethane (Yoon *et al.*, 2014). In contrast to AOPs, ARPs degrade contaminants with highly reactive, nonselective reducing nucleophiles or radicals, such as aqueous electrons (also known as hydrated electrons, e_{aq}^-), $H^{\bullet}$, and $SO_3^{\bullet -}$. Current studies (Table 6) on PFAS degradation have focused on PFCAs and have generated radicals by utilizing dithionite, sulfite, aqueous iodide, and ferrocyanide in combination with UV, laser flash photolysis, ultrasound, microwave, or electron beam (E-beam).

Hydrated electrons are the main nucleophiles that degrade PFCAs (E° = −2.9 V) (Park *et al.*, 2009). Cleavage of the α-position C–F bond, instead of the C–C bond, initiates degradation [Eqs. (S34)–(S37) in Supplementary Table S6] (Qu *et al.*, 2010; Song *et al.*, 2013). This attack results from the inductive effect of the carboxyl head group and the ability of fluorine to withdraw electrons (electron affinity 3.40 eV) (Blondel *et al.*, 1989). In comparison, the carbon atoms in PFCAs are saturated and cannot gain more electrons. After cleavage occurs [Eqs. (S34)–(S37) in Supplementary Table S6], other free radicals are formed as a result of UV irradiation or other activation methods, including $^{\bullet}C_{n-1}F_{2n-1}$, carbene ($:CH_2$), and $^{\bullet}COOH$ (Qu *et al.*, 2010; Song *et al.*, 2013). Carbene can then form $^{\bullet}CH_3$, leading to the formation of CH_3COOH (Qu *et al.*, 2010).

ARPs: aqueous iodide (KI)

KI with UV (254 nm) has been one of the most studied ARPs and can decompose several PFASs. When UV light is present, iodide (I^-) will form a caged complex ($I^{\bullet}$, e^-) in water [Eqs. (S42) and (S43) in Supplementary Table S6], which can then dissociate to e_{aq}^- and iodine atom [Eq. (S44) in Supplementary Table S6] (Qu *et al.*, 2010). Initial studies demonstrating the capabilities of KI were limited and resulted in high concentrations of PFCAs and PFSAs remaining in solution (Park *et al.*, 2009, 2011). This was likely due to quenching and sequestration of e_{aq}^- by the production of triiodide (I_3^-) from high concentrations of KI [Eqs. (S45)–(S50) in Supplementary Table S6] (Qu *et al.*, 2010; Park *et al.*, 2011). Strong greenhouse gases were also produced, including iodinated hydrocarbons, CHF_3, and C_2F_6 (IPCC, 2007; Qu *et al.*, 2010).

KI treatment was further improved by utilizing alkaline conditions and a closed reactor (Qu *et al.*, 2014; Zhang *et al.*, 2014). Alkaline conditions reduced the amount of greenhouse gases produced, and the closed reactor (N_2 gas) prevented the reaction of e_{aq}^- with O_2. Regeneration of iodide was also possible when pH > 8.5 [Eqs. (S51) and (S52) in Supplementary Table S6]. Increasing the temperature and varying ionic strength also improved PFOA decomposition and defluorination ratios.

ARPs: dithionite and sulfite

Dithionite ($S_2O_4^{2-}$) and sulfite (SO_3^{2-}) have been used to degrade PFOA with relatively limited success. Both dithionite and sulfite will produce e_{aq}^- and other reductants. When irradiated with UV (315 nm), dithionite will form two sulfur dioxide radical anions ($2SO_2^{\bullet-}$, $E° = -0.66$ V) (Mayhew, 1978; Makarov, 2001; Vellanki *et al.*, 2013) [Eq. (S53) in Supplementary Table S6]. Other products can be generated during this process, including H_2SO_3, HSO_3^-, and SO_3^{2-}. Aquated electrons and other reductants, such as $H^\bullet$ and sulfite radical ($SO_3^{\bullet-}$), will form as a result of UV irradiation of these products and can then be used to breakdown PFOA [Eqs. (S54) and (S55) in Supplementary Table S6] (Fischer and Warneck, 1996; Lian *et al.*, 2006; Li *et al.*, 2012a). Generated sulfite radicals can also be used, either as a reductant or oxidant [Eqs. (S56)–(S60) in Supplementary Table S6] (Liu *et al.*, 2013b; Vellanki *et al.*, 2013). While PFOA degradation seems promising using dithionite and sulfite, <10% PFOA was removed with UV light and no degradation was observed when using dithionite and sulfite with ultrasound, microwave, or E-beam (Vellanki *et al.*, 2013). Comparatively, Song *et al.* (2013) observed 68.6% defluorination of PFOA within 6 h (N_2 gas), but did not quantitate the PFOA decomposition ratio or metabolite yield.

ARPs: summary

Degradation of PFASs using ARPs needs more research to determine better degradation parameters for dithionite and sulfite and to determine other ARPs that can be used. For example, the ARP, $K_4Fe(CN)_6$, in combination with laser flash photolysis has been studied for trifluoroacetic acid (TFA), PFBA, and PFOA, but this process has limited applications and has not been further optimized for PFAS degradation (Huang *et al.*, 2007). In contrast, KI may be applied to PFCA-contaminated wastewater. Qu *et al.* (2010) observed about 96% PFOA degradation when using KI to destroy PFCAs in wastewater from a fluorochemical plant in China.

Thermal and nonthermal destruction

Thermal degradation of PFASs involves breaking the C–C and C–F bonds with high temperatures to produce perfluoroalkyl radicals that will subsequently decompose and form similar degradation products as photolytic treatment of PFASs. Thermal treatment methods include thermal chemical reactions, incineration, sonochemistry, sub- or supercritical, microwave-hydrothermal, and high-voltage electric discharge (Tables 7 and 8).

Thermal and nonthermal destruction: incineration and thermal chemical reactions

Incineration is one of the most common ways to destroy hazardous compounds and to reduce waste, but can result in harmful emissions. Incineration of PFASs, including fluorotelomer alcohol (FTOH)-based acrylic polymers, PFOS, ammonium perfluorooctanoate (APFO), and PFOA, has been successful at temperatures ranging from 600°C to1,000°C (USEPA, 2003; Krusic and Roe, 2004; Krusic *et al.*, 2005; Yamada *et al.*, 2005; Taylor *et al.*, 2014). This may lead to the formation of 1H-substituted perfluoroalkyl substances, such as 1-H-perfluoroheptane, which are volatile and mobile products (Krusic and Roe, 2004; Krusic *et al.*, 2005). Other harmful emissions, such as dioxins and furans, can be produced if PFASs are incinerated with other wastes (Tuppurainen *et al.*, 1998; McKay, 2002). Strong greenhouse gases have been observed from the combustion of PFOS, including tetrafluoromethane (CF_4) and hexafluoroethane (C_2F_6) (Yamada *et al.*, 2005). The global-warming potentials are 5,700 and 11,900, respectively (IPCC, 2007), with long atmospheric lifetimes of 50,000 and 10,000 years, respectively (IPCC, 2013). These harmful by-products may be reduced with certain additives, such as calcium hydroxide (Wang *et al.*, 2011a, 2013). More research is needed to fully understand the effects of incineration on PFASs and by-products formed.

Other studies have observed PFCA and perfluoroether carboxylic acid decomposition to shorter-chain carboxylic acids, F^-, and CO_2 under much more benign, thermal chemical methods by combining heat (30–85°C) with persulfate. Similar reactions took place as with persulfate and UV light [Eqs. (S28)–(S33) in Supplementary Table S6] and resulted in PFOA decomposition to nondetectable levels, with faster degradation occurring with increasing temperatures (Lee et al., 2012b; Liu *et al.*, 2012a). Other PFCAs, perfluoroether carboxylic acids, and PFNA from floor wax were also observed to degrade to nondetectable levels within 6 h (Hori *et al.*, 2008a).

Thermal and nonthermal destruction: sonochemical degradation

Sonochemical degradation of PFASs occurs through the application of ultrasound to an aqueous medium. When ultrasound is applied, cavitation bubbles form during the rarefaction (negative pressure) portion of sound waves (Thompson and Doraiswamy, 1999; Joseph *et al.*, 2009). The cavitation bubbles will implode adiabatically, creating extreme temperatures (>9,700°C in the vapor core) and pressures (~ 14,000 psi) within its cavity (Didenko *et al.*, 1999; Ashokkumar and Grieser, 2005; Ciawi *et al.*, 2006; Eddingsaas and Suslick, 2007; Park *et al.*, 2009). Highly reactive intermediates and radicals, including hydroxyl radicals, hydrogen atom, and oxygen atom, form during cavitation bubble collapse (Leighton, 1994). This combination of highly reactive species and high temperatures and pressures has made sonolytic decomposition of PFASs successful.

PFOS and PFOA were completely mineralized through sonolysis to CO, CO_2, F^-, and SO_4^{2-}, as detected by HPLC-MS, ion chromatography, FT-IR, and GC-MS. Two studies report no detection of reaction intermediates and complete defluorination of PFOS within 3 h and PFOA within 2 h (Vecitis *et al.*, 2008b, 2010). There was immediate production of inorganic sulfur and fluorine atoms, with a slight delay in production of CO and CO_2 (Vecitis *et al.*, 2008b).

Complete mineralization was possible due to the presence of three different reactivity sites: inside the cavitation bubble, at the interfacial region between the cavitation bubble and the bulk aqueous solution, and in the bulk aqueous solution and vapor phase (Moriwaki *et al.*, 2005; Vecitis *et al.*, 2008b).

The sonolytic decomposition of PFASs depended on the type of gas used and the initial PFAS concentration. All sonolytic decomposition processes of PFASs have been conducted using argon gas since it will produce higher temperatures and increased reaction yields compared with air (Moriwaki *et al.*, 2005). However, Phan Thi *et al.* (2014) observed 100% PFOA decomposition when using nitrogen gas with $NaHCO_3$. The initial concentration of PFASs is important as saturation kinetics could influence the reaction. At higher concentrations of PFOS or PFOA (Table 8), zero-order kinetics were observed, indicating saturation of adsorption sites on the interfacial region, while at lower concentrations, pseudo-first-order kinetics took place (Vecitis *et al.*, 2008a).

Sonolysis of PFOA and PFOS was affected by cocontaminants and electrolyte concentrations. Volatile organic compounds (VOCs) (e.g., methanol, acetone, and methyl isobutyl ketone) decreased the decomposition rate, likely caused by competitive adsorption onto the interfacial region or evaporation of VOCs into the bubble vapor phase, decreasing bubble vapor and interfacial temperatures (Cheng *et al.*, 2008). In contrast, dissolved organic matter did not significantly impact PFOA and PFOS sonochemical degradation kinetics (Cheng *et al.*, 2008). Electrolyte concentration could either increase or decrease the sonolytic degradation rate constant of PFOS and PFOA (Cheng *et al.*, 2010). For example, the effect of electrolytes on sonochemical rates could be ordered as $ClO_4^- > NO_3^- \sim Cl^- \geq 0 > HCO_3^- > SO_4^{2-}$ (Cheng *et al.*, 2010), where ClO_4^-, NO_3^-, and Cl^- increased decomposition, while HCO_3^- and SO_4^{2-} decreased decomposition. The electrolyte probably influenced interfacial conditions by either increasing the number of surface sites available or by altering heat transfer from the bubble vapor to the bulk liquid. In addition to PFOA and PFOS, shorter-chain PFASs, including PFHxA, PFHxS, PFBA, and PFBS, can be degraded by sonolysis with pseudo-first-order kinetics (Campbell *et al.*, 2009).

Sonolysis of PFASs may be improved in conjunction with other treatment methods, such as ozone, microwave irradiation, persulfate, and VUV (Yang *et al.*, 2013). When sonolysis and ozone were applied to groundwater containing PFOS and PFOA, the degradation rates increased by 79% for PFOS and 70% for PFOA when compared with Milli-Q water (Cheng *et al.*, 2008). Similarly, microwave irradiation combined with an ultrasonic homogenizer decomposed PFOA within only 90 s with 59% defluorination yields (Horikoshi *et al.*, 2011). Temperatures reached 1,000°C at the tungsten tip and 51°C in the bulk liquid. Active species were also generated, including hydroxyl radical, hydrogen atom, and oxygen atom, causing decarboxylation and oxidation of PFOA and its intermediates. Comparatively, a more benign treatment approach can be used with persulfate and sonolysis under either air or argon gas and has been used to degrade five perfluoroether carboxylic acids and two perfluoroether sulfonates (Hori *et al.*, 2012). Argon gas increased removal yields compared with air due to the occurrence of higher temperatures when the cavitation bubbles collapsed.

Thermal and nonthermal destruction: sub- or supercritical treatment

Treatment methods using sub- or supercritical water can be environmentally benign. Subcritical water temperatures range from 100°C to 350°C and are maintained at a certain pressure to hold a liquid state. In comparison, supercritical water temperatures reach >350°C and pressures >22.1 MPa (Jessop and Leitner, 1999). At these temperatures and pressures, sub- and supercritical water has useful properties for degrading hazardous compounds, including high diffusivity and low viscosity.

Iron has been used in combination with sub- or supercritical water to increase decomposition of PFASs. Compared with aluminum, copper, and zinc (Hori *et al.*, 2006), iron increased decomposition of PFOS (Hori *et al.*, 2006) and Nafion NRE-212, a perfluorinated ion-exchange membrane (Hori *et al.*, 2010), in an argon-saturated aqueous subcritical solution. The PFOS degradation efficiency of each metal could be ordered as $Al < Cu < Zn << Fe$, while the redox potential of each metal could be ordered as $Cu < Fe < Zn < Al$, suggesting that the metal surface plays a more important role than redox potential (Hori *et al.*, 2006). Increasing the surface area and using zero-valent iron also improves decomposition of PFASs (Hori *et al.*, 2008b, 2013b, 2015).

Compared with subcritical water, PFAS degradation was enhanced under supercritical conditions. Hori *et al.* (2008b) observed increased consumption of PFHxS under supercritical conditions (94.8% PFHxA decomposed) compared with subcritical conditions (83.6% PFHxS decomposed) when Fe powder was added. However, under supercritical conditions, more CF_3H was produced, a greenhouse gas with a global warming potential of 14,800 for a 100-year horizon and an atmospheric lifetime of 270 years (IPCC, 2007).

Thermal and nonthermal destruction: microwave-hydrothermal treatment

Microwave-hydrothermal treatment is more cost-efficient when compared with other thermal treatment processes and can save up to 50% in energy consumption. Higher decomposition rates, enhanced kinetics, and rapid and homogeneous heating have also been observed (Park *et al.*, 2000; Jones *et al.*, 2002).

Persulfate has been used in combination with microwave-hydrothermal treatment to decompose PFOA and will form sulfate radicals with heat, similar to persulfate and UV light, as shown in Equations (S28)–(S33) in Supplementary Table S6 (Lee *et al.*, 2009). At 90°C, PFOA was decomposed to nondetectable levels after 6 h, and at 60°C, the reaction took twice as long to achieve the same PFOA removal (Lee *et al.*, 2009). Although microwave-hydrothermal treatment with persulfate is quick, it requires low pH, as found in certain industrial wastewaters (e.g., chromium plating), to form more sulfate radicals, as seen in Equations (S61) and (S62) in Supplementary Table S6 (Lee *et al.*, 2012a). In addition, the pH will drop quickly as the reaction proceeds from pH 3.6 to 2.3 in 1 h and less than 0.1 U every hour afterward due to the formation of more protons.

Persulfate activated by microwave-hydrothermal treatment was improved with the addition of zero-valent iron (ZVI) powder and inhibited by the addition of chloride ions. ZVI acted as a source of ferrous ions and led to faster activation of persulfate [Eq. (S63) in Supplementary Table S6]. Within 1 h at 90°C, about 60% PFOA was degraded ($\sim$15% fluoride yield) (Lee *et al.*, 2010). While PFOA degradation efficiency increased with ZVI, high concentrations of ZVI (14.4–18 mM) resulted in less PFOA degradation due to the release of ferrous ions that competed with PFOA for sulfate radicals. Chloride ions were also observed to inhibit PFOA degradation rate (Lee *et al.*, 2012b).

Thermal and nonthermal destruction: high-voltage electric discharge

High-voltage electric discharge reactors apply electric discharges directly in water (electrohydraulic discharge) or above water (nonthermal plasma, NTP) and have been utilized for inactivation of microorganisms and removal of organic substances, such as phenols, chlorinated solvents, and organic dyes (Locke *et al.*, 2006). These electric discharges will generate strong electric fields and highly active species, such as hydroxyl radicals, ozone, oxygen radicals, and hydrogen radicals. Shock waves and UV light may also occur as a result of electric discharge application. During electrohydraulic discharge, liquid–gas phase reactions with reactive species and other organic compounds can occur as bubbles form in the electric field. NTP has also been shown to generate electrons that have temperatures >9,700°C and can activate highly reactive species (Holzer *et al.*, 2002; Roland *et al.*, 2002; Oda, 2003; Locke *et al.*, 2006). High-voltage electric discharge can be cost-efficient, depending on the reaction time and energy utilization [e.g., decomposition energy yield of 16 g/kWh or 1 W power (Magureanu *et al.*, 2010)].

A DC electrohydraulic plasma discharge reactor was utilized for PFOA and PFOS decomposition (Yasuoka *et al.*, 2010, 2011; Matsuya *et al.*, 2014; Takeuchi *et al.*, 2014; Hayashi *et al.*, 2015). DC plasmas were generated within oxygen gas bubbles and reached temperatures $\sim$2,000 K. PFOA and PFOS molecules adsorb onto the gas–liquid interface in high concentration (Matsuya *et al.*, 2014), where generated positive species (M^+) collided and reacted with anionic forms of PFOA or PFOS. The reaction can cause decarboxylation degradation pathways [Eqs. (S64)–(S66) in Supplementary Table S6] or C–C bond cleavage [Eqs. (S67)–(S69) in Supplementary Table S6]. C–C bond cleavage is likely since the gaseous fluorocarbons, CF_m ($m = 1$–3), were detected (Takeuchi *et al.*, 2014). Other gaseous fluorocarbons detected during only the first 10 min of the reaction include CHF_3, C_2F_6, and C_2HF_5. Shorter-chain perfluorocarboxylates, especially TFA [Eq. (S68) in Supplementary Table S6] and PFHxA [Eq. (S69) in Supplementary Table S6], fluoride ions, or sulfate ions, were also observed. Recently, Hayashi *et al.* (2015) demonstrated the successful use of a two-parallel operation of a DC electrohydraulic plasma discharge reactor for large-scale treatment of PFOS [Eqs. (S70)–(S73) in Supplementary Table S6].

The NTP method, Glidarc, was shown to decompose a perfluorinated nonionic surfactant known as Forafac 110 (C_6F_{13}–$C_2H_4(OC_2H_4)_{11.5}OH$) (Marouf-Khelifa *et al.*, 2008). Within 6 h, 96.7% Forafac was removed, and more efficient reaction time (within 1 h) was observed when adding anatase or rutile TiO_2. Glidarc involves the production of an electric arc between two electrodes in a gaseous medium (Marouf-Khelifa *et al.*, 2006). When the glidarc is exposed to humid air plasma, $NO^\bullet$ and $OH^\bullet$ are formed. The $NO^\bullet$ leads to the formation of NO_2, NO_2^-, and NO_3^- [Eqs. (S74)–(S79) in Supplementary Table S6] (Marouf-Khelifa *et al.*, 2006). The $OH^\bullet$ makes the glidarc a strong oxidizer, while NO species acidifies the reaction. Other species, such as H_2O_2 and O_3, are also produced to improve decomposition (Marouf-Khelifa *et al.*, 2008).

Thermal and nonthermal destruction: summary

Studies using thermal and nonthermal processes for PFAS destruction have been successful with some limitations, including production of toxic by-products and greenhouse gases. These methods are also relatively more expensive than AOPs and sorption removal processes. For example, to treat phenol-contaminated wastewater at a 1,000 L/min capacity treatment plant, sonolysis would cost $15,537 per 1,000 gallons of treated water. When combined with other treatment processes, such as ozone or UV, this cost can be lowered to $25.80. In comparison, treating the same wastewater would cost $7–11 per 1,000 gallons of treated water with a commercial process known as Perox-pure™ (Calgon Carbon Oxidation Technologies) (Mahamuni and Adewuyi, 2010). The cost efficiency of other methods for PFAS removal is still unknown, especially when dealing with PFAS mixture and cocontaminants.

Microbial treatment processes

Microbial degradation of PFASs has only been observed to occur with polyfluoroalkyl substances (Butt *et al.*, 2014), which contain some C–H bonds instead of C–F bonds (Buck *et al.*, 2011). Although reductive defluorination of perfluoroalkyl substances may be possible, as observed when using vitamin B12 and Ti(III)-citrate (Ochoa-Herrera *et al.*, 2008), there are no known reports of biotransformation occurring. Vitamin B12 is needed in microbial reductive dehalogenation processes and it can be used *in vitro* when Ti(III) is supplied as the reducing agent.

Butt *et al.* (2014) published a comprehensive review on the biotransformation pathways (microbial, mammalian, and fish) of fluorotelomer-based compounds. Many polyfluoroalkyl substances, such as 6:2 FTOH, are transformed to perfluoroalkyl substances, such as PFHxA, PFPeA, and PFBA, by pure bacterial cultures and environmental samples. Perfluoroalkyl metabolite production has been observed in other studies published after Butt *et al.* (2014), including WWTP effluent (Guerra *et al.*, 2014), AFFF-amended microcosms (Harding-Marjanovic *et al.*, 2015), and soil–plant microcosms with WWTP biosolids (Rankin *et al.*, 2014). Comparatively, the wood-rotting fungi, *Phanerochaete chrysosporium*, transformed 6:2 FTOH toward polyfluoroalkyl substances, including 5:3 polyfluorinated acid (5:3 acid) and 5:3 acid conjugates within 28 days (Tseng *et al.*, 2014). While many studies demonstrated perfluoroalkyl metabolite production, this may not occur under all environmental conditions, such as anaerobic bioreactors (Alder and van der Voet, 2015). More research on the microbial degradation of PFASs must be conducted to fully understand the biotransformation of PFASs in the environment.

Other treatment processes

Several other studies have used different treatment methods to degrade PFASs, including ozonation under alkaline conditions, permanganate, and ball milling. Ozonation is a commonly used AOP in at least one-third of water treatment plants in the United States (Crittenden *et al.*, 2012). Ozonation of PFOA and PFOS was viable within 4 h when pretreating with O_3 at pH 4–5, followed by pH adjustment to 11, but environmental matrices containing humic acid may inhibit ozonation (Lin *et al.*, 2012a). Permanganate is also widely used as an oxidizing agent for iron and manganese, taste and odor control, microorganism control, and degradation of other hazardous pollutants (Crittenden *et al.*, 2012; Liu *et al.*, 2012b). Permanganate removed about 50% PFOS, but with only 5% fluoride yield at 65°C and pH 4.2 (Liu *et al.*, 2012b). Although complete PFOS decomposition could not be achieved, degradation efficiency of permanganate improved with increasing temperatures and was not inhibited by the addition of organic acids, including oxalic, tartaric, succinic, citric, and humic acid.

In contrast to ozone and permanganate, ball milling is a type of mechanochemical (MC) destruction method that has been used to destroy PFOS and PFOA (Zhang *et al.*, 2013b). Reactions take place at the surface of the ball mills while mechanic force is applied, such as shaking. This process effectively destroyed PFOS (<0.2% PFOS remained with 92.3% fluoride yield) after 6 h of ball milling. When potassium hydroxide (KOH) was added, PFOS and PFOA were completely destroyed with higher fluoride yields.

Conclusion

A wide variety of technologies to remove or destroy PFASs have been tested by researchers and practitioners. Results show that a variety of PFASs can effectively be removed from water and wastewater using sorption onto AC, ion exchange, MIPs, and other sorbents. Knowledge of PFAS sorption mechanisms has been used to design more efficient sorbents and to predict their performance under a range of environmental conditions. Technologies for destroying PFASs include a variety of AOPs, ARPs, thermal and nonthermal destruction methods, and other innovative approaches (e.g., ball milling). These technologies effectively destroyed select PFASs under idealized laboratory conditions. However, many studies discussed in this review may not have achieved detection limits below provisional guidelines set by EPA (ng/L) (USEPA, 2016). While analytical methods to measure PFASs have become more sensitive within the past decade and can attain detection limits of ng/L (Naile *et al.*, 2010; Cao *et al.*, 2011; Wang *et al.*, 2011b; ASTM, 2015), most PFAS removal methods have not been retested using new analytical techniques. Further research is needed on promising PFAS removal methods that attained nondetectable PFAS levels for the development of applicable remediation strategies.

Despite these advances, more work is required to develop a design basis for confidently employing PFAS remediation strategies. Several technologies (e.g., ARPs) require additional basic research to elucidate reaction mechanisms, determine degradation parameters, decomposition products, and defluorination yields. Destruction technologies can likely be improved for field implementation (e.g., lower reagent doses, temperatures, pressures, energy consumption). Comprehensive research studies are needed to predict and address the effects of complex field conditions on treatment technology performance. Field conditions are typically affected by the presence and distribution of PFAS mixtures, cocontaminants (e.g., chlorinated solvents, metals, and 1,4-dioxane), and environmental matrix parameters (e.g., temperature, pH, organic matter content, inorganic ions, oxygen concentrations, groundwater, sediment geochemistry). Due to the complexity of PFAS mixtures in the environment, comprehensive studies may not be possible until accurate and efficient analytical methods for PFAS precursors and metabolites are developed and standardized.

Application of promising treatment technologies at wastewater and industrial treatment plants and at pilot-scale or full-scale remediation systems also merits further research. Such studies need to take into account the effect of natural environmental conditions on PFAS transformation and distribution. PFAS fate and transport can also be affected by remediation systems designed and previously implemented for cocontaminants. Currently, two field studies have observed the likely transformation of PFAS precursors to terminal PFAS products, such as PFOA and PFOS, at former firefighting training bases, where several remediation methods have been utilized to remove other contaminants (McGuire *et al.*, 2014; Anderson *et al.*, 2016). A bench-scale study observed no transformation of PFAAs using ISCO with activated persulfate, permanganate, or catalyzed hydrogen peroxide (McKenzie *et al.*, 2015) and demonstrated that ISCO remediation efforts for other contaminants can affect PFAS fate and transport. Furthermore, certain ISCO treatments may be used for PFAS containment (persulfate) or for pump-and-treat efforts (permanganate and catalyzed hydrogen peroxide). More field studies and PFAS monitoring must be conducted to fully understand the removal, fate, and transport of these compounds.

Another direction for further research is the effective use of PFAS treatment technologies in treatment trains. Due to the stable nature of these compounds, treatment processes will likely need to be combined to achieve cost-efficient removal. For example, photocatalysis could be combined with membrane separation processes, sonolysis, or biological treatment. Unfortunately, current studies have been limited to understanding the removal efficiencies of one technology under fairly simplistic conditions. Ideally, the assortment of research avenues will eventually provide different treatment methods to cost-effectively remove PFASs under various circumstances and field conditions.

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