

## Supplementary Data

SUPPLEMENTARY TABLE S1. ABBREVIATIONS OF GENERAL TERMS, DEFINITIONS, AND EQUATIONS

AFFFs	Aqueous film-forming foams
AOPs	Advanced oxidation processes
ARPs	Advanced reduction processes
CEC	Cation exchange capacity (cmol/kg)
ISCO	<i>In situ</i> chemical oxidation
pH <sub>pzc</sub>	pH point of zero charge
SA <sub>BET</sub>	Brunauer–Emmett–Teller surface area (m <sup>2</sup> /g)
TNTs	Trinitrotoluenes
TSS	Total suspended solids
WWTP	Wastewater treatment plant
VSS	Volatile suspended solid
Decomposition	C–C or C–F bond cleavage. In general, PFASs can be reduced to shorter-chain PFASs, fluoride ion, CO <sub>2</sub> , sulfate ions, etc., depending on the parent compound. Synonyms include degradation and destruction.
Mineralization	Degradation of the target compound to its elemental form, such as fluoride ion, CO <sub>2</sub> , and sulfate ions.
F <sup>−</sup> yield	$\frac{\text{Fluoride ions produced}(\frac{\text{mg}}{\text{L}})}{[\text{PFASs}(\frac{\text{mg}}{\text{L}})]_0 \times \frac{19 \frac{\text{g}}{\text{mol}} \times Y}{\text{PFASs MW}(\frac{\text{g}}{\text{mol}})}} \times 100 = \text{Fluoride ion Yield (\%)} $ <p>Y = number of fluoride ions in PFASs, where 19 g/mol is the molecular weight of fluoride  MW = molecular weight  Higher fluoride yields are desired and indicate liberation of F<sup>−</sup> ions from the target PFAS and into solution. For example, if the target PFAS concentration is below the detection limit after the reaction, but F<sup>−</sup> yield is about 40%, then 60% F<sup>−</sup> are theoretically still located in target PFAS metabolites and not in solution. F<sup>−</sup> mass balance loss has been observed in some PFAS destruction studies, as discussed in the main text, and can also be sequestered by calcium precipitation or other methods.</p>
Removal efficiency	$\frac{[\text{PFASs}]_0 - [\text{PFASs}]_t}{[\text{PFASs}]_0} \times 100 = \% \text{ Removal}$ <p>The amount of PFASs removed or degraded over time (t). While several studies described in the main text indicate complete removal of the target PFAS(s), metabolites can include shorter-chain PFASs that still exist in the system. Furthermore, the limit of detection for each study should be taken into consideration.</p>

SUPPLEMENTARY TABLE S2. ABBREVIATIONS AND FORMULAS OF PFASs USED IN THE REVIEW

APFO	Ammonium perfluorooctanoate	$\text{NH}_4^+\text{C}_7\text{F}_{15}\text{COO}^-$
DTFA	1H, 1H, 2H, 2H, 8H, 8H-perfluorododecanol	$\text{CF}_3(\text{CF}_2)_3\text{CH}_2(\text{CF}_2)_5\text{CH}_2\text{CH}_2\text{OH}$
Forafac 110		$\text{C}_6\text{F}_{13}-\text{C}_2\text{H}_4(\text{OC}_2\text{H}_4)_{11.5}\text{OH}$
FTEO	Fluorotelomer ethoxylate	$\text{F}(\text{CF}_2\text{CF}_2)_x(\text{CH}_2\text{CH}_2\text{O})_y\text{H}$
FTOH	Fluorotelomer alcohol (n:2 FTOH)	$\text{CF}_3(\text{CF}_2)_{n-1}\text{CH}_2\text{CH}_2\text{OH}$
FTSA	Fluorotelomer sulfonate (n:2 FTSA)	$\text{CF}_3(\text{CF}_2)_{n-1}\text{CH}_2\text{CH}_2\text{SO}_3^-$
FTUCA	Fluorotelomer unsaturated carboxylic acid	$\text{CF}_3(\text{CF}_2)_{n-2}\text{CF}=\text{CHCOO}^-$ (n:2 FTUCA) $\text{CF}_3(\text{CF}_2)_{n-1}\text{CH}=\text{CHCOO}^-$ (n:3 FTUCA)
NFDOHA	Nonafluoro-3,6-dioxaheptanoic acid	$\text{CF}_3\text{OC}_2\text{F}_4\text{OCF}_2\text{COOH}$
PFBA	Perfluorobutanoic acid	$\text{C}_3\text{F}_7\text{COOH}$
PFBS	Perfluorobutane sulfonic acid	$\text{C}_4\text{F}_9\text{SO}_3\text{H}$
PFCAs	perfluorocarboxylic acids	$\text{C}_n\text{F}_{2n+1}\text{COOH}$
PFDA	Perfluorodecanoic acid	$\text{C}_9\text{F}_{19}\text{COOH}$
PFHpA	Perfluoroheptanoic acid	$\text{C}_6\text{F}_{13}\text{COOH}$
PFHxA	Perfluorohexanoic acid	$\text{C}_5\text{F}_{11}\text{COOH}$
PFHxS	Perfluorohexanesulfonic acid	$\text{C}_6\text{F}_{13}\text{SO}_3\text{H}$
PFOA	Perfluorooctanoic acid	$\text{C}_7\text{F}_{15}\text{COOH}$
PFOS	Perfluorooctane sulfonic acid	$\text{C}_8\text{F}_{17}\text{SO}_3\text{H}$
PFOSF	Perfluorooctanesulfonyl fluoride	$\text{C}_8\text{F}_{18}\text{O}_2\text{S}$
PFNA	Perfluorononanoic acid	$\text{C}_8\text{F}_{17}\text{COOH}$
PFPeA	Perfluoropentanoic acid	$\text{C}_4\text{F}_9\text{COOH}$
PFPrA	Perfluoropropanoic acid	$\text{C}_2\text{F}_5\text{COOH}$
PFSAs	perfluorosulfonic acids	$\text{C}_n\text{F}_{2n+1}\text{SO}_3\text{H}$
TFA	Trifluoroacetic acid	$\text{CF}_3\text{COO}^-$
PFAAs	Perfluoroalkyl acids	

PFASs, perfluoroalkyl and polyfluoroalkyl substances.

SUPPLEMENTARY TABLE S3. ABBREVIATIONS  
OF SORPTION TERMS

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AC	Activated carbon
ACF	Activated carbon fiber
BAC	Bamboo-derived AC
CNTs	Carbon nanotubes
ECH	Epichlorohydrin
E(N)FMs	Electrospun (nanofibrous) membranes
$f_{oc}$	Organic carbon fraction
GAC	Granular activated carbon
HDTMAB	Hexadecyltrimethylammonium bromide
(M)MCN	(Magnetic) mesoporous carbon nitride
MOFs	Metal-organic frameworks
Mt	Montmorillonite
MWCNTs	Multiwalled carbon nanotubes
PAC	Powdered activated carbon
PAC-MBR	Powdered activated carbon membrane bioreactor
PCMA <sub>s</sub>	Permanently confined micelle arrays
PLGA	Poly(D, L-lactide-co-glycolid)
PVA	Polyvinyl alcohol
TRIA	Trimethylolpropane triacrylate

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SUPPLEMENTARY TABLE S4. ABBREVIATIONS  
OF AOP TERMS

BDD	Boron-doped diamond
CB	Conduction band
CHP	Catalyzed H <sub>2</sub> O <sub>2</sub> propagation
MWCNTs	Multiwalled carbon nanotubes
RDE	Rotating disk electrode
SCE	Saturated calomel electrode
Titania	TiO <sub>2</sub>
UNCD	Ultrananocrystalline boron-doped conductive diamond
VB	Valence band
(V)UV	(Vacuum) ultraviolet light

AOPs, advanced oxidation processes.

SUPPLEMENTARY TABLE S5. ABBREVIATIONS OF ARPs

E-beam	Electron beam
$e_{aq}^-$	Aqueous or hydrated electrons
I <sup>-</sup>	Aqueous iodide (from KI)
VOCs	Volatile organic compounds
ZVI	Zero-valent iron powder

ARPs, advanced reduction processes.

SUPPLEMENTARY TABLE S6. PROPOSED EQUATIONS FOR THE TRANSFORMATIONS OF PFASs

Electrochemical oxidation, the PFCA example		
1. Radical formation on BDD surface	$C_7F_{15}COO^- \rightarrow C_7F_{15}COO^\bullet + e^-$	S1
	$H_2O \rightarrow \bullet OH + H^+ + e^-$	S2
2. Mineralization	$C_7F_{15}COO^\bullet \rightarrow C_7F_{15}^\bullet + CO_2$	S3
	$C_7F_{15}^\bullet + \bullet OH \rightarrow C_7F_{15}OH$	S4
	$C_7F_{15}OH \rightarrow C_6F_{13}COF + H^+ + F^-$	S5
	$C_6F_{13}COF + H_2O \rightarrow C_6F_{13}COO^- + 2H^+ + F^-$	S6
	Repeat Equations (S1)–(S6)	
Electrochemical oxidation, PFSA example		
1. Radical formation on BDD surface	$C_8F_{17}SO_3^- \rightarrow C_8F_{17}SO_3^\bullet + e^-$	S7
2. Mineralization	$C_8F_{17}SO_3^\bullet + H_2O \rightarrow C_8F_{17}^\bullet + SO_4^{2-} + 2H^+$	S8
	Repeat Equations S4–S6	
Proposed photocatalysis mechanisms		
1. Radical formation	$C_nF_{2n+1}COO^- + (1) \rightarrow C_nF_{2n+1}^\bullet + COO^{\bullet -}$	S9a
	$C_nF_{2n+1}COO^- + (2) \text{ or } (3) \rightarrow C_nF_{2n+1}COO^\bullet$	S9b
2. Decarboxylation	$C_nF_{2n+1}COO^\bullet \rightarrow CO_2 + C_nF_{2n+1}^\bullet$	S10
	$C_nF_{2n+1}^\bullet + OH^\bullet \rightarrow C_nF_{2n+1}OH$	S11
3. HF elimination	$C_nF_{2n+1}OH \rightarrow C_{n-1}F_{2(n-1)+1}COF + HF$	S12
4. Formation of shorter-chain PFAS	$C_{n-1}F_{2(n-1)+1}COF + H_2O \rightarrow C_{n-1}F_{2(n-1)+1}COOH + HF$	S13
Proposed iron photocatalysis decomposition mechanism		
	$C_7F_{15}COO^- + Fe^{3+} \rightarrow [C_7F_{15}COO-Fe]^{2+}$	S14
	$[C_7F_{15}COO-Fe]^{2+} + h\nu \rightarrow Fe^{2+} + C_7F_{15}COO^\bullet$	S15
	$Fe^{2+} + O_2 \rightarrow Fe^{3+} + O_2^{\bullet -}$	S16
	$Fe^{2+} + \bullet OH \rightarrow Fe^{3+} + OH^-$	S17
Proposed iron–hydrogen peroxide photocatalysis decomposition mechanism		
	$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + \bullet OH + OH^-$	S18
	$Fe^{2+} + \bullet OH \rightarrow Fe^{3+} + OH^-$	S19
	$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^\bullet + H^+$	S20
	$Fe^{3+} + HO_2^\bullet \rightarrow Fe^{2+} + O_2 + H^+$	S21
	$H_2O_2 + \bullet OH \rightarrow HO_2^\bullet + H_2O$	S22
CHP reaction mechanism		
	$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^\bullet + H^+$	S23
	$\bullet OH + H_2O_2 \rightarrow HO_2^\bullet + H_2O$	S24
	$HO_2^\bullet \leftrightarrow O_2^{\bullet -} + H^+ (pK_a = 4.8)$	S25
	$HO_2^\bullet + Fe^{2+} \rightarrow Fe^{3+} + HO_2^-$	S26
	$H_2O_2 \leftrightarrow HO_2^- + H^+ (pK_a = 11.8)$	S27
Proposed persulfate oxidation mechanism		
1. Persulfate activation	$S_2O_8^{2-} + h\nu \rightarrow 2SO_4^{\bullet -}$	S28
	$SO_4^{\bullet -} + OH^- \rightarrow SO_4^{2-} + \bullet OH$	S29
	$SO_4^{\bullet -} + H_2O \rightarrow \bullet OH + SO_4^{2-} + H^+$	S30
2a. Sulfate radical scavenging	$SO_4^{\bullet -} + SO_4^{\bullet -} \rightarrow S_2O_8^{2-}$	S31
	$SO_4^{\bullet -} + S_2O_8^{2-} \rightarrow SO_4^{2-} + S_2O_8^{\bullet -}$	S32
2b. Sulfate radical reaction with PFASs	$SO_4^{\bullet -} + PFASs \rightarrow SO_4^{2-} + PFASs^{\bullet +}$	S33
Proposed advanced reduction of PFAS mechanism		
1. Cleavage of $\alpha$ -position C–F bond by $e_{aq}^-$	$C_nF_{2n+1}COOH + e_{aq}^- \rightarrow \bullet C_nF_{2n}COOH + F^-$	S34
	$\bullet C_nF_{2n}COOH + H_2O \rightarrow C_nF_{2n}HCOOH + \bullet OH$	S35
	$C_nF_{2n}HCOOH + e_{aq}^- \rightarrow \bullet C_nF_{2n-1}HCOOH + F^-$	S36
	$\bullet C_nF_{2n-1}HCOOH + H_2O \rightarrow C_nF_{2n-1}H_2COOH + \bullet OH$	S37
2. Free radical formation (e.g., via UV irradiation)	$C_nF_{2n-1}H_2COOH \rightarrow \bullet C_{n-1}F_{2n-1} + \bullet CH_2 + \bullet COOH$	S38
	$\bullet C_{n-1}F_{2n-1} + COOH \rightarrow C_{n-1}F_{2n-1}COOH$	S39
3. Carbene reaction	$\bullet CH_2 + \bullet H \rightarrow \bullet CH_3$	S40
	$\bullet CH_3 + \bullet COOH \rightarrow CH_3COOH$	S41
Proposed hydrated electron generation and recycle mechanism from iodide ion		
1. Formation of caged complex	$I^- + H_2O + h\nu (254 \text{ nm}) \rightarrow I^\bullet H_2O^*$	S42
	$I^\bullet H_2O^* \rightarrow (I^\bullet, e^-) + H_2O$	S43
2. Radical formation	$(I^\bullet, e^-) \rightarrow I^\bullet + e_{aq}^-$	S44
3. Iodide radical reactions	$I^\bullet + I^- \rightarrow I_2^{\bullet -}$	S45
	$I^\bullet + I^\bullet \rightarrow I_2$	S46
	$2I_2^{\bullet -} \rightarrow I^- + I_3^-$	S47
	$I^- + I_2 \rightarrow I_3^-$	S48
4a. $e_{aq}^-$ consumption: $I_2$ or $I_3$ present	$e_{aq}^- + I_2 \rightarrow I_2^{\bullet -}$	S49
	$e_{aq}^- + I_3^- \rightarrow I_2^{\bullet -} + I^-$	S50
4b. $e_{aq}^-$ consumption: $H^+$ present or low pH	$e_{aq}^- + H_3O^+ \rightarrow H^\bullet$	S51
5. $I^-$ regeneration (pH >8.5)	$3I_2 + 6OH^- \rightarrow 5I^- + IO_3^- + 3H_2O$	S52

(continued)

SUPPLEMENTARY TABLE S6. (CONTINUED)

Proposed dithionite and sulfite reaction mechanism			
1a. Radical formation: dithionite	$S_2O_4^{2-} \rightarrow 2SO_2^{\bullet-}$ (or $H_2SO_3$ , $HSO_3^-$ , $SO_3^{2-}$ )		S53
1b. Radical formation: hydrogen sulfide	$HSO_3^- + h\nu \rightarrow SO_3^{\bullet-} + H^+$		S54
1c. Radical formation: sulfite	$SO_3^{2-} + h\nu \rightarrow SO_3^{\bullet-} + e_{aq}^-$		S55
2a. Sulfite radical: oxidant	$SO_3^{\bullet-} + e_{aq}^- \rightarrow SO_3^{2-}$		S56
2b. Sulfite radical: reductant	$SO_3^{\bullet-} + O_2 \rightarrow SO_5^{\bullet-}$		S57
	$2SO_3^{\bullet-} + 2SO_3^{\bullet-} + H_2O \rightarrow S_2O_6^{2-} + SO_4^{2-} + H^+ + HSO_3^-$		S58
	$SO_3^{\bullet-} + SO_3^{\bullet-} \rightarrow S_2O_6^{2-}$		S59
	$SO_3^{\bullet-} + SO_3^{\bullet-} + H_2O \rightarrow SO_4^{2-} + H^+ + HSO_3^-$		S60
Microwave-hydrothermal treatment—effects of pH on sulfate radical			
All pHs	$SO_4^{\bullet-} + H_2O \rightarrow SO_4^{2-} + OH^{\bullet} + H^+$	$k < 60 M^{-1}s^{-1}$	S61
Alkaline pH	$SO_4^{\bullet-} + OH^- \rightarrow SO_4^{2-} + OH^{\bullet}$	$k = 7 \times 10^7 M^{-1}s^{-1}$	S62
Microwave-hydrothermal treatment—effects of ZVI on sulfate radical			
	$S_2O_8^{2-} + Fe^{2+} \rightarrow Fe^{3+} + SO_4^{\bullet-} + SO_4^{2-}$		S63
Electrohydraulic plasma discharge reactor			
1. Decarboxylation	$C_nF_{2n+1}COO^- + M^+ \rightarrow C_nF_{2n+1}COO^{\bullet} + M^+ + e^-$		S64
	$C_nF_{2n+1}COO^{\bullet} \rightarrow C_nF_{2n+1}^{\bullet} + CO_2$		S65
	$C_nF_{2n+1}^{\bullet} + 2H_2O \rightarrow C_{n-1}F_{2n-1}COO^- + 3H^+ + 2F^- + H$		S66
2. C–C cleavage (PFOA example)	$C_7F_{15}COO^- + 2M^+ \rightarrow CF_3^{\bullet} + \bullet C_6F_{12} + \bullet COO^- + 2M^+$		S67
	$CF_3^{\bullet} + \bullet COO^- \rightarrow CF_3COO^{\bullet}$		S68
	$\bullet C_6F_{12} + 2H_2O \rightarrow C_5F_{11}COO^- + 2H^+ + F^- + 2H$		S69
3. PFOS example	$C_8F_{17}SO_3^- + M^+ \rightarrow C_8F_{17}SO_3^{\bullet} + M^+ + e^-$		S70
	$C_8F_{17}SO_3^{\bullet} \rightarrow C_8F_{17}^{\bullet} + SO_3$		S71
	$C_8F_{17}^{\bullet} + 2H_2O \rightarrow C_7F_{15}COO^- + 3H^+ + 2F^- + H$		S72
	$SO_3 + H_2O \rightarrow 2H^+ + SO_4^{2-}$		S73
Nonthermal plasma–Glidarc			
	$H_2O + e^- \rightarrow \bullet OH + H^{\bullet} + e^-$		S74
	$N_2 + e^- \rightarrow N(^4S) + N(^2D) + e^-$		S75
	$N(^2D) + O_2 \rightarrow NO^{\bullet} + O$		S76
	$NO^{\bullet} + \bullet OH \rightarrow NO_2 + H^{\bullet}$		S77
	$NO_2 + HO_2^{\bullet} \rightarrow HNO_2 + O_2$		S78
	$HNO_2 + 2HO^{\bullet} \rightarrow HNO_3 + H_2O$		S79

 CHP, catalyzed  $H_2O_2$  propagation.