

1 **Remediation of poly- and perfluoroalkyl substances (PFAS) contaminated soils – to**
2 **mobilize or to immobilize or to destroy?**

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102 **List of abbreviations**

103 AC: Activated carbon

104 AECOM: Architecture, engineering, consulting, operations, and maintenance

105 AFFF: Aqueous film-forming foam

106 BAF: Bioaccumulation factors

107 BCF: Bioconcentration factor

108 CFR: Chlorinated flame retardants

109 CNT: Carbon nanotubes

110 CTAB: Cetyltrimethylammonium bromide

111 dAQ: Aquifer into a deeper section

112 DDT: Dichlorodiphenyltrichloroethane

113 diPFCA: Polyfluoroalkyl phosphate diesters

114 DOM: Dissolved organic matter

115 dw: dry weight

116 EC50: Effective Concentration₅₀

117 ECCC: Environment and climate change Canada

118 EDL: Environmental decontamination limited

119 FCSAP: Federal contaminated sites action plan

120 ESTS: Emergencies science and technology section

121 FOSA: Perfluorooctane sulfonamide

- 122 FTCA: Fluorotelomer carboxylic acids
- 123 FTS: Fluorotelomer sulfonates
- 124 FTuCA: Unsaturated fluorotelomer carboxylic acids
- 125 GAC: Granulated activated carbon
- 126 HCB: Hexachlorobenzene
- 127 ISCO: *in-situ* chemical oxidation
- 128 K_d : Soil specific distribution coefficient
- 129 K_{oc} or K_{om} : Partition coefficients with the specific organic particle or organic component of the
- 130 soil
- 131 K_{ow} : Octanol-water partition coefficient
- 132 LC50: Lethal Concentration₅₀
- 133 LD50: Lethal Dose₅₀
- 134 LOQ: Instrumental limit of quantitation
- 135 LOR: Level of reporting
- 136 L-PFBS: Perfluorobutane sulfonate
- 137 L-PFOA: Non branched perfluorooctanoic acid
- 138 MCD: Mechanochemical destruction
- 139 MEP: Multiple extraction procedure
- 140 MTBE: Methyl-tert-butyl ether
- 141 NAPL: Nonaqueous phase liquid
- 142 NOEC: No observable effect concentration
- 143 OC: Organic carbon
- 144 PAC: Powdered activated carbon
- 145 PAH: Polycyclic aromatic hydrocarbon
- 146 PBDE: Polybrominated diphenyl ethers

- 147 PCB: Polychlorinated biphenyls
- 148 PFAA: Perfluoroalkyl acid
- 149 PFAS: Perfluorinated alkyl substances
- 150 PFBA: Perfluorobutanoic acid
- 151 PFBS: Perfluorobutane sulfonic acid
- 152 PFC: Perfluorinated compounds
- 153 PFCA: Perfluorocarboxylic acids
- 154 PFDA: Perfluorodecanoic acid
- 155 PFDDA: Perfluorododecanoic acid
- 156 PFDoDA: Perfluorododecanoic acid
- 157 PFHpA: Perfluoroheptanoic acid
- 158 PFHxA: Perfluorohexanoic acid
- 159 PFHxS: Perfluorohexane sulfonate
- 160 PFNA: Perfluorononanoic acid
- 161 PFOA: Perfluorooctanoic acid
- 162 PFOS: Perfluorooctane sulfonate
- 163 PFOSA: Perfluorooctane sulphonamide
- 164 PFPeA: Perfluoropentanoic acid
- 165 PFSA: Perfluorosulfonic acids
- 166 PFTeDA: Perfluorotetradecanoic acid
- 167 PFUnDA: Perfluoroundecanoic acid
- 168 POP: Persistent organic pollutants
- 169 RO: Reverse osmosis
- 170 sAQ: Aquifer into a shallow section

171 SCF: Supercritical fluid
172 SCISOR: Smart combination *In-situ* oxidation/reduction
173 SDBS: Sodium dodecylbenzene sulfonate
174 SOM: Soil organic matter
175 SPLP: Synthetic precipitation leaching procedure
176 SSA: Specific surface area
177 S/S: Stabilization and solidification
178 THF: Tetrahydrofuran
179 TII: Thermodynamic index of irreversibility
180 TSCF: Transpiration stream concentration factor
181 WWTP: Wastewater treatment plant
182 VEG: Vapor energy generator
183 UCS: Uniaxial compressive strength

184

185 **Highlights**

- 186 • Collated (im)mobilization and destruction techniques for PFAS remediation in soil.
- 187 • Described interrelationships between soil PFAS dynamics and their remediation.
- 188 • Ascribed soil amendments to PFAS bioavailability and mobility in relation to
189 remediation.
- 190 • Covered case studies of successful remediation techniques for PFAS contaminated soil.
- 191 • Highlighted environmental implications of various soil PFAS remediation techniques

192

193 **Abstract**

194 Poly- and perfluoroalkyl substances (PFASs) are synthetic chemicals, which are introduced to
195 the environment through anthropogenic activities. Aqueous film forming foam used in

196 firefighting, wastewater effluent, landfill leachate, and biosolids are major sources of PFAS
197 input to soil and groundwater. Remediation of PFAS contaminated solid and aqueous media
198 is challenging, which is attributed to the chemical and thermal stability of PFAS and the
199 complexity of PFAS mixtures. In this review, remediation of PFAS contaminated soils through
200 manipulation of their bioavailability and destruction is presented. While the mobilizing
201 amendments (e.g., surfactants) enhance the mobility and bioavailability of PFAS, the
202 immobilizing amendments (e.g., activated carbon) decrease their bioavailability and mobility.
203 Mobilizing amendments can be applied to facilitate the removal of PFAS through soil washing,
204 phytoremediation, and complete destruction through thermal and chemical redox reactions.
205 Immobilizing amendments are likely to reduce the transfer of PFAS to food chain through plant
206 and biota (e.g., earthworm) uptake, and leaching to potable water sources. Future studies should
207 focus on quantifying the potential leaching of the mobilized PFAS in the absence of removal
208 by plant and biota uptake or soil washing, and regular monitoring of the long-term stability of
209 the immobilized PFAS.

210

211 **Key words:** PFAS; Aqueous firefighting foam; Soil remediation; Biosolids; Mobilization and
212 immobilization

213

214 **1. Introduction**

215 The substances of both organic and inorganic origin containing at least one fluorine (F) atom
216 are generally termed as fluorinated substances or fluorochemicals or fluorinated chemicals.
217 Among them, a specific sub-group of chemicals are known as perfluoroalkyl and
218 polyfluoroalkyl substances (PFASs) that contain a perfluoroalkyl moiety in their structures
219 (Banks et al., 1994; Buck et al., 2011). The perfluoroalkyl moiety, generally represented by
220 C_nF_{2n+1} , is a one or more carbon (C) atom-containing fluorinated aliphatic chain in which most

221 of its hydrogen (H) atoms are substituted by F atoms (Banks et al., 1994). To be classified as a
222 PFAS, the substance should have at least one perfluoroalkyl moiety in its chemical structure.
223 The key groups of PFASs include perfluorinated carboxylic acids (PFCAs), perfluorinated
224 sulfonic acids (PFSAs), and perfluorinated phosphonic acids (PFPA), whereas polyfluorinated
225 compounds include fluorotelomer alcohols (FTOHs), fluorotelomer sulfonic acids (FTSs),
226 polyfluorinated alkyl phosphates (PAPs), perfluorooctane sulfonamide (PFOSA), and their
227 derivatives (Buck et al., 2011). It is estimated that >4,000 synthetic fluorinated compounds
228 belong to the complex PFAS family.

229 The perfluorinated or polyfluorinated compounds are anthropogenic in nature and released into
230 the environment due to human and industrial activities (Buck et al., 2011). Owing to their
231 strong C-F bond strengths, these compounds are remarkably resistant to external environmental
232 conditions such as temperature, water or oil status of the medium, and microbial attack. As a
233 result, PFASs have found widespread applications in various industries including plastic
234 manufacturing, textiles and leather industries, surfactants preparation, and even in medical
235 applications (Prevedouros et al., 2006). Daily-life applications of PFASs extend to food
236 wrapping materials, drink can-lining materials, non-sticky cookware, water-resistant fabrics
237 and clothing, grease/oil resistant papers and surfaces, and firefighting foam (Darrow et al.,
238 2013; Mahinroosta and Senevirathna, 2020; Yeung et al., 2016).

239 Due to their widespread use in industries and daily-life products, PFAS have entered the soil
240 and water environments, and now they are found in microorganisms, plants, higher animals,
241 and humans globally, including in the Arctic and Antarctic ecosystems (Ahrens et al., 2016;
242 Dreyer et al., 2009; Sunderland et al., 2019). Due to their unique chemical structures and
243 stability, several PFASs have been proven to be bioaccumulative and toxic in higher animals
244 including humans (Ahrens et al., 2019; Lau et al., 2007; Xiao et al., 2017). Among various
245 usages, aqueous film forming foam (AFFF) is recognised as one of the key sources of PFAS

246 entry into the soil and water environments. Perfluorooctane sulfonate (PFOS),
247 perfluorooctanoic acid (PFOA), and perfluorohexane sulfonate (PFHxS) are the most
248 commonly found PFASs in AFFF. The Class B fluorine-based AFFF is used to extinguish
249 flammable liquid-caused fires, and firefighting training sites at and around fire brigades,
250 airports, and defence establishments have been reported to contaminate the surrounding soil,
251 water bodies, and groundwater with PFAS (Cousins et al., 2019). In addition to such point
252 sources of PFAS, diffused pollution of soil and groundwater has taken place through
253 transportation of these contaminants from household products and activities via biosolids,
254 wastewater treatment plants, and landfill leachates (Sinclair and Kannan, 2006; Bolan, 2019).
255 Because of the high chemical, thermal, and biological stability of PFAS compounds, and also
256 due to their existence as mixtures in environmental matrices, their remediation both in water
257 and soil/sediment is extremely challenging. Approaches taken to remediate PFAS in solid and
258 liquid media can be different. For example, they can be removed from a solid medium (e.g.,
259 soil or biosolid) following mobilization using selective chemical species, or their concentration
260 can be reduced by plant uptake and by chemical or microbial destruction (Jeon et al., 2011;
261 Simon et al., 2019). In the case of liquid media, PFAS compounds are removed mostly by using
262 adsorbents or chemical destruction (Merino et al., 2016). PFAS immobilization in solid media
263 has also been reported by adsorbents or fixing agents (Darlington et al., 2018). In terms of
264 biotic degradation of PFAS compounds in soil and water, available information is limited (Wei
265 et al., 2019).

266 A few review articles have concentrated on strategies for removing PFAS compounds from
267 water, including sorption on various adsorbents (Du et al., 2014; Zhang et al., 2019; Vo et al.,
268 2020), focussing on field testing of selected adsorbents (España et al., 2015), and advanced
269 defluorination and degradation (thermal and non-thermal) methods (Ahmed et al., 2020; Vo et
270 al., 2020), but review articles critically analysing soil PFAS remediation are scarce in the

271 literature. Only two reviews have been written on this topic in the recent past (Ross et al., 2018;
272 Mahinroosta and Senevirathna, 2020). However, more information is needed concerning the
273 scientific and technological soundness of PFAS remediation approaches, especially in soils.
274 So, the current review paper aims to present information on the remediation of PFAS
275 contaminated soils by critically reflecting on the pros and cons of contaminant mobilization,
276 immobilization, and destruction strategies using a wide range of soil amendments. The
277 mobilizing amendments help to desorb PFAS compounds that are bound to soil minerals or
278 organic matter, and hence increase their bioavailability and mobility (Milinovic et al., 2015,
279 2016; Pan et al., 2009; Tang et al., 2017). In contrast, the immobilizing amendments adsorb or
280 fix the PFAS compounds in soils reducing their bioavailability and mobility (Aly et al., 2019;
281 Das et al., 2013; Hale et al., 2017). This review discusses all the above strategies by first
282 presenting an overview of various soil PFAS sources, PFAS interactions with soil components
283 and bioavailability, soil PFAS remediation through manipulating the bioavailability using
284 conventional and advanced soil amendments along with some exemplary case studies, and
285 highlighting their respective techno-economic advantages and disadvantages.

286

287 **2. Sources of PFAS in soil**

288 Soil and water environments receive PFAS contaminants mainly through discharge of AFFF,
289 effluent discharge from wastewater treatment plants and landfills, and contaminated wastes
290 such as biosolids (Table 1; Figure 1).

291

292 **2.1 Firefighting foams**

293 As an important point source, PFAS from AFFF can be introduced into the terrestrial and
294 aquatic environments during storage, handling, use, and post-use cleaning stages of these
295 chemicals (Cousins et al., 2019). For example, a small PFAS volume can be released in the

296 form of a concentrated foam at the time of storage, careless handling during transfer of
297 containers, and usage and calibration of equipment. However, a more occasional release than
298 the above occurs when a large volume of PFAS enters into the environment during real
299 firefighting operations (Houtz et al., 2012, 2013). Additionally, downward leakage and/or
300 lateral overflow from temporary ponds storing AFFF-contaminated water following fire
301 training operations can also become an important diffused source of contamination in the
302 surrounding areas (Eschauzier et al., 2013; Houtz et al., 2012; 2013).

303 Soil and groundwater contamination resulting from the use of firefighting foams at defence
304 sites, airports, and fire brigade training sites has been noticed in many countries including
305 Australia and the USA. For example, currently around 90 sites in Australia are being
306 investigated for PFAS contamination from the regular use of firefighting foam (Australian
307 Defense, 2019). Similarly, around 26,000 PFAS contaminated sites exist across the USA,
308 impacting more than six million people through drinking water contamination (Darlington et
309 al., 2018). Both in Australia and USA, the impacted areas are located around and near defence
310 facilities where AFFF is used either for real fire extinguishing or training purposes. There are
311 sources of PFAS other than AFFF that contaminate the environment, but given the great extent
312 of PFAS contamination (i.e., high PFAS concentration and toxic chemical constituents) in and
313 around defence sites globally, these sites warrant immediate risk assessment and remediation
314 actions.

315

316 2.2 Wastewater effluents and sludges

317 Municipal waste disposal sites, landfills, wastewater treatment facilities, and biosolids provide
318 major diffused sources of PFAS contamination of soil and water. For example, household
319 wastewater containing PFAS chemicals in low concentration can reach municipal wastewater
320 treatment plants and finally accumulate in biosolids (Bossi et al., 2008; Campo et al., 2014;

321 Chen et al., 2012a; Gallen et al., 2018; Guo et al., 2010; SI Figure 1). A number of PFAS
322 compounds including PFOA and PFOS were found in Australian biosolids in recent years
323 (Gallen et al., 2016; 2017; 2018). Higgins et al. (2005) reported total PFAS concentrations
324 ranging from 55 to 3370 ng/g in domestic sludge in the USA. Venkatesan and Halden (2013)
325 measured PFAS concentration in 113 biosolid samples collected from 94 waste water treatment
326 plants in USA, and obtained a mean concentration of PFOS = 403 ± 127 ng/g, PFOA = $34 \pm$
327 22 ng/g, and PFDA = 26 ± 20 ng/g. Similarly, Sun et al. (2011) reported total PFAS
328 concentrations in digested sewage sludge in Switzerland ranging from 28 to 637 ng/g, while
329 total PFOS concentrations ranged from 15 to 600 ng/g. Kallenborn et al. (2004) demonstrated
330 that Nordic countries had relatively low PFAS concentrations (0.6 to 15.2 ng/g) in sludges.
331 Levels of PFAS contamination in Swedish sludge ranged from 0.6 to 23.9 ng/g and 1.6 to 54.8
332 ng/g for PFOA and PFOS, respectively (Haglund and Olofsson, 2009). Other estimates
333 suggested that annually around 2749-3450 kg of total PFAS was present in biosolids across the
334 USA, and around 1375-2070 kg PFAS ended up in agricultural land through soil applications
335 (Venkatesan and Halden, 2013). Sepulvado et al. (2011) found that PFASs were found to
336 concentrate to the 120 cm soil depth and reached a concentration up to 483 ng/g in field soils
337 that had received long term biosolid applications. Washington et al. (2010) investigated sludge-
338 applied soils in proximity to a wastewater treatment plant handling sewage waste from PFAS
339 industries. The PFOS and PFOA concentrations in the sludge applied soil reached a maximum
340 of 408 $\mu\text{g}/\text{kg}$ and 312 $\mu\text{g}/\text{kg}$, respectively. Sludge from this wastewater treatment plant was
341 found to contain PFOA concentrations up to 1875 ng/g.

342 PFAS can enter the sewage system through a variety of industrial sources that include PFAS
343 manufacturing, fluoropolymer manufacturing, and AFFF manufacturing (Prevedouros et al.,
344 2006). Many studies reported the degree of PFAS pollution in wastewater sludge (Table 1).
345 The PFAS issue arises in sewage sludge because conventional wastewater and sewage

346 treatment methods cannot efficiently eliminate these recalcitrant compounds from the system.
347 The increase in concentration of some PFAS compounds such as perfluoroalkyl acid (PFAA)
348 in sewage effluents over that in the influent is attributed to the degradation of more complex
349 PFAA precursors during activated sludge treatment (Houtz et al., 2012; 2013). For example,
350 wastewater treatment plants could show 9–352% increase in PFOA concentration in effluents
351 over influents (Schultz et al., 2006). However, PFOS often could exhibit a decrease in
352 concentration in the effluent, attributed to high K_d values causing retention of PFOS in the
353 sludge and lowering final PFOS concentrations in effluents (Yu et al., 2009). Becker et al.
354 (2008) observed a 20-fold increase in PFOA concentrations from influents to effluents, and an
355 additional 10 and 50% PFOA and PFOS, respectively, adsorbed in the sludges.

356

357 2.3 Landfill leachate

358 Household wastes composed of goods containing hydrophobic and stain-resistant coatings
359 (e.g., carpets) can release PFAS chemicals when dumped into the landfill (Gallen et al., 2016;
360 Wei et al., 2019; Yan et al., 2015; Lang et al., 2017). As a result, PFAS can be released into
361 the groundwater through contaminated leachates from landfills, or laterally move to
362 surrounding land areas if appropriate lining is not in place (SI Figure 2). In addition to sewage
363 and wastewater, industrial wastes such as fabrics, building and coating materials can be a
364 diffuse source of PFAS and their related chemicals (Janousek et al., 2019) when they are
365 disposed in landfill sites. For example, long-chain PFAAs (e.g., PFOA and its precursors) were
366 detected in landfill sites where the above types of wastes were dumped off for disposal. Knutsen
367 et al. (2019) warned that short-chain PFAS compounds released from a range of household and
368 industrial wastes could dominate over long-chain compounds in the leachates of historic
369 landfill sites. The removal of such short-chain PFAS contaminants from water can be extremely
370 challenging, which, in addition to the general challenges of landfill leachate treatment methods,

371 underscores the PFAS contamination issue worldwide in ground and surface water resources
372 from landfill sites.

373

374 **3. Dynamics of PFAS in soils**

375 PFAS compounds that reach soil through various sources (Table 2) can undergo sorption,
376 partition, and complexation reactions that enable them to be retained in the soil (Zhang et al.,
377 2019). Sorption refers to electrostatic interaction of PFAS with charged clay and organic matter
378 surfaces, whereas partition refers to hydrophobic interaction of PFAS with organic substrates
379 such as soil organic matter. Complexation involves partition of PFAS with dissolved organic
380 matter forming soluble PFAS-organic matter complexes (Li et al., 2018; Zhu and Kannan,
381 2019). In contrast, plant uptake, leaching, degradation/transformation, and volatilization
382 potentially can remove these chemicals from contaminated soils (Figure 2). The dynamics of
383 PFAS compounds and their fate in the soil depend on the characteristics of PFAS compounds
384 (e.g., solubility and chain length), soil properties (e.g., organic matter and pH), and
385 environmental factors (e.g., precipitation) (Milinovic et al., 2015). PFAS compounds can be
386 adsorbed on soil particles through hydrophobic interaction and/or electrostatic attraction
387 (Johnson et al., 2007; Wei et al., 2019). The major PFAS compounds such as PFOA and PFOS
388 tend to exist as dissociated anions of acids under natural soil environmental conditions. PFAS
389 compounds in the soil system can show both hydrophilic and hydrophobic characteristics.
390 While the long-fluorinated alkyl chain of PFAS compounds confers on them hydrophobic
391 properties, the sulfonate and carboxylate functional groups provide them hydrophilic
392 characteristics (Darlington et al., 2018; Ross et al., 2018). Vapor state mobility of PFAS
393 compounds may rarely occur under soil environmental conditions, because of the low to very
394 low vapor pressures of most of the PFAS compounds (Kucharzyk et al., 2017), or in other
395 words these compounds have high molecular weight and are highly stable in nature.

396

397 3.1 Sorption/desorption process

398 This section covers both sorption as indicated by electrostatic interaction of PFAS with charged
399 clay and organic matter surfaces, and partition as indicated by hydrophobic interaction of PFAS
400 with organic substrates such as soil organic matter. Because of simultaneously having
401 hydrophobic fluoroalkyl long chains and hydrophilic ionizable functional groups, PFAS
402 compounds show complex behaviours in the environment in terms of their sorption and
403 desorption processes (Ahrens, 2011; Kannan, 2011). The sorption of PFAS in soils has been
404 shown to increase with an increase in the chain length of PFAS compounds and also with an
405 increase in the fraction of organic components (f_{oc}) in the soil (Brusseau et al., 2018; Milinovic
406 et al., 2015). Simultaneously, PFAS sorption is also influenced by the soil pH and soil solution
407 ionic strength. It has been shown that PFAS sorption in soil increases with an increasing
408 electrolyte concentration (i.e., ionic strength) and/or due to the presence of higher valent
409 cations in the soil solution (Wang and Shih, 2011). A decreasing pH of the soil (i.e., dominance
410 of protons (H^+) on the soil surface) also increases the sorption of PFAS compounds, which is
411 attributed mainly to an increase in positive charge with decreasing pH (Du et al., 2014; Jeon et
412 al., 2011; Bolan et al., 1999). Therefore, two key mechanisms can be identified for PFAS
413 sorption in the soil environment: (1) hydrophobic interaction with soil particles rich in aromatic
414 hydrophobic components, and (2) surface electrostatic interaction with charged soil minerals
415 (Figure 3; Hellsing et al., 2016).

416 The sorption of PFAS compounds in soils and sediments has been studied extensively. In
417 general, long chain PFAS compounds are adsorbed in soils and sediments dominantly via
418 hydrophobic attraction, and short chain compounds via polar-polar interaction (e.g.,
419 electrostatic attraction) (Zhao et al., 2012). Soils and sediments containing high contents of
420 organic carbon (OC) or black carbon particles thus tend to show high sorption of PFAS

421 compounds. The physicochemical behaviours of PFAS compounds, especially the net
422 hydrophobicity evolving from their chemical structures are critical to predict the sorption
423 strengths of these chemicals to soils and sediments. For example, Milinovic et al. (2015) reported
424 that among three studied PFAS compounds, namely PFOS, PFOA and perfluorobutane
425 sulfonic acid (PFBS), PFOS was the most strongly adsorbed by six different soils. The authors
426 attributed the strong interaction of PFOSs with soil particles to hydrophobic interaction, as
427 indicated by a strong correlation between the log K_{ow} values of the three PFAS compounds and
428 the log K_{oc} values of the soils (Figure 4). Chen et al. (2013) demonstrated that PFOS and PFOA
429 sorption (K_d value) increased with an increase of the organic carbon fraction and ionic strength
430 of five different soils, whereas the sorption decreased with an increasing humic acid (HA)
431 concentration in the solution. The authors suggested that HA or other dissolved organic matter
432 might form complexes with PFAS compounds in the soil solution and inhibit sorption of those
433 chemicals on to soil components such as clay minerals and particulate organic matter.

434 Helling et al. (2016) found that a negatively charged silica surface was not able to adsorb
435 anionic PFAS compounds such as perfluorohexanoic acid (PFHxA), PFOA, PFOS, and
436 perfluorononanoic acid (PFNA). On the contrary, positively charged alumina surface adsorbed
437 significant amounts of these compounds, indicating that an electrostatic mechanism might
438 come into partial effect for adsorbing PFAS compounds on electrically charged soil
439 components (Higgins and Luthy, 2006). Johnson et al. (2007) suggested that PFOS sorption
440 mechanisms to the surfaces of minerals such as kaolinite, goethite, high iron sand, and Ottawa
441 sand could be dominantly controlled by electrostatic attraction when surfaces of these minerals
442 were OC free. The presence of organic carbon of the mineral surfaces drives the sorption
443 mechanism toward hydrophobic interaction. According to their charge characteristics, the
444 above minerals adsorbed PFOS in the order: goethite > kaolinite > high iron sand > Ottawa
445 sand. Tang et al. (2010) observed that pH, ionic strength, and Ca^{2+} concentration of solutions

446 significantly influenced the sorption of PFOS by goethite, but their effects were only marginal
447 when sorption occurred on silica. In the case of goethite, low pH values and high Ca^{2+}
448 concentrations enhanced PFOS sorption via possible electrostatic attraction. Likewise, Ferrey
449 et al. (2012) suggested that at around solution pH = 7, iron oxides adsorbed PFOS and PFOA
450 predominantly through electrostatic attraction rather than hydrophobic interaction.

451 Literature suggests that PFAS sorption and desorption studies are mostly concentrated on soils
452 and sediments, where point source pollution occurred mainly surrounding AFFF handling and
453 storage facilities. However, due to the high mobility of these compounds, reports now exist
454 that agricultural soils globally are also contaminated or vulnerable for contamination by these
455 toxic compounds (Yao et al., 2015). The sorption and desorption behaviour of PFAS in
456 agricultural soils can be different than that of other soils, because agricultural soils receive a
457 continuous supply of amendments, fertilizers, and irrigation water. Therefore, understanding
458 the retention of PFAS in agricultural soils requires future research attention, and the fate and
459 behaviour of these contaminants should be studied using advanced biogeochemical prediction
460 models.

461

462 3.2 Leaching

463 Most PFAS compounds are relatively more soluble in water than other persistent organic
464 pollutants such as PAHs (Post et al., 2017). Hence, PFASs are liable for leaching, especially in
465 soils with low sorption capacity (e.g., sandy soils). The sorption of the PFAS compounds in
466 soils influences their leaching behaviour through the soil profile (Gellrich et al., 2012). Gellrich
467 et al (2012) observed that in groundwater, PFASs with short chain lengths (<7 fluorinated
468 carbon atoms) predominate in concentrations. The short chain PFASs can be less toxic than
469 long chain ones (e.g., PFOS and PFOA); short chain PFASs display a higher mobility but lower
470 bioaccumulation potential in the environment (Das et al., 2008; Newsted et al., 2008; Gellrich

471 et al., 2012). However, despite the restricted use of PFAS, the concentrations of PFOA and
472 PFOS in water and other environmental matrices are likely to increase because of (1) the
473 continuous desorption of PFOS and PFOA that are still bound to soil particles, and (2) the slow
474 transformation of precursors of these compounds in environmental compartments (Frömel and
475 Knepper, 2010). Gellrich et al. (2012) detected PFOA in the leachate percolating through a soil
476 column until about four months after the experiment began, but PFOS was found below the
477 detection limit (1 ng/L) even after 2.5 years. Similarly, Stahl et al. (2013) reported that PFOA
478 moved rapidly with water through a soil lysimeter, while PFOS travelled slowly (SI Figure 3).
479 Thus, the leaching behaviour of PFAS is in part influenced by the extent to which the PFAS is
480 adsorbed onto soil and sediments during transport. The sorption of PFAS onto soil and
481 sediments during transport facilitates partial removal of PFAS from aqueous media, which
482 potentially retards PFAS flow velocity relative to the velocity of water, thereby attenuating the
483 concentration of PFASs over distance and time across the stream (Higgins and Luthy, 2006; Li
484 et al., 2019; Ross et al., 2018).

485

486 3.3 Plant uptake

487 Unlike nonpolar contaminants (e.g., petroleum hydrocarbons) that tend to accumulate on plant
488 root surfaces via lipid partitioning, polar contaminants including PFAS may enter into the
489 transpiration stream of plants and move across the whole system (Ahrens et al., 2009; Blaine
490 et al., 2013; Dalahmeh et al., 2018; Garcia-Valcarcel et al., 2014). For those contaminants that
491 enter the transpiration stream, the ratio of concentrations of the solute in the transpiration
492 stream to the soil solution is defined as the transpiration stream concentration factor (TSCF)
493 (Felizeter et al., 2012).

494 $TSCF = [\text{Concentration of solute in the transpiration stream} / \text{Concentration of solute in the soil}$
495 $\text{solution}]$

496 Plant uptake of PFAS compounds is dependent on PFAS chain length and the sorption
497 behaviour in soils. Being relatively hydrophilic, the short chain PFAS are expected to have
498 high TSCF values. Since ionisable contaminants such as PFAS are soluble and non-volatile, a
499 high concentration of PFAS can potentially accumulate in plants (Ghisi et al., 2019). PFAS
500 accumulation occurs predominately in the leaves, because water along with PFAS enters into
501 the roots and then translocates to the leaves before getting evaporated, resulting in the
502 accumulation of PFAS in the leaves (Ghisi et al., 2019). The bioaccumulation factor (BAF) of
503 PFAS can be calculated as follows (Liu et al., 2017):

$$504 \text{ BAF} = [\text{PFAS concentration in the plant } (\mu\text{g/kg plant}) / \text{PFAS concentration in the soil } (\mu\text{g/kg} \\ 505 \text{ soil})]$$

506 Only a few studies have documented the bioaccumulation potential of PFAS, particularly
507 PFOA and PFOS, into food crops (SI Figure 4). In an artificially contaminated soil, Stahl et al.
508 (2009) observed an uptake of PFOS and PFOA in maize, wheat, potato, and oats, and
509 particularly high concentrations accumulated in the vegetative portions of the plants. Lechner
510 et al. (2011) also reported similar PFOA and PFOS uptake patterns in the vegetative portions
511 of carrot, cucumber, and potato. Blaine et al. (2013) found that the dry weight concentrations
512 of PFBA and PFPeA in plants grown in a PFAS-contaminated biosolid-amended soil under
513 glasshouse conditions reached 266 and 236 $\mu\text{g/kg}$ in lettuce, and 56 and 211 $\mu\text{g/kg}$ in tomato,
514 respectively. PFBA showed the highest BAF (56.8) in the case of lettuce, while the highest
515 BAF for PFPeA was 17.1 in the case of tomato. Under field conditions, while no PFBA and
516 PFPeA were detected in corn grains, concentrations of the chemicals in the corn stover were
517 ultralow (Blaine et al., 2013). In another study, Blaine et al. (2014a) found that among various
518 PFAS compounds present in a contaminated biosolid-amended soil, crops such as radish,
519 celery, and pea accumulated the highest concentrations of PFOA (67 $\mu\text{g/kg}$), PFBA (232
520 $\mu\text{g/kg}$), and PFBA (150 $\mu\text{g/kg}$), respectively. However, an increasing chain length of PFAS

521 compounds significantly decreased the shoot-soil concentration factor (SCF) for all the crops.
522 Under greenhouse conditions, Blaine et al. (2013) also monitored the entry of PFAS into the
523 human food chain via irrigating food crops (lettuce and strawberry) with reclaimed water
524 (PFAS concentration = 0.2-40 µg/L). PFBA and PFPeA (short-chain PFAS) showed the overall
525 highest accumulation of any PFAS in the edible parts of both the crops. The authors also
526 reported that the OC content of soils had an inverse relationship with the bioaccumulation of
527 PFAS. Zhu and Kannan (2019) conducted a field study within a one-mile radius of a five-
528 decade old fluoropolymer (PFCA) industry and observed that, while the soil samples and plant
529 tissues contained mainly PFOA, the earthworms accounted for higher proportions of long-
530 chain PFCAs (e.g., PFUnDA and PFDoDA). The biota-soil accumulation factor in earthworms
531 and root-soil accumulation factor in plants/grasses increased with an increasing chain length of
532 the PFCAs. Thus, the bioaccumulation of PFAS in crops from soils would depend on PFAS
533 concentration, soil properties, crop species, and the specific PFAS analyte, and it is not clearly
534 understood. Also, reports concerning PFAS bioaccumulation in beneficial soil animals such as
535 earthworms are scant (Karnjanapiboonwong et al., 2018; Zhao et al., 2013; Zhu and Kannan,
536 2019), and, given the importance of these animals in ecosystem functions, it warrants future
537 studies.

538

539 3.4 PFAS transformation

540 The major transformation processes of PFAS include abiotic and biotic degradation with a
541 limited extent of volatilization. Transformation of various precursor substances can provide an
542 indirect source of PFAS input to soils (Ruan et al., 2015). For example, fluorotelomer alcohols
543 (FTOHs; $F(CF_2)_nCH_2CH_2OH$) are some of the major indirect sources of PFAS input in soil
544 (Dinglasan et al., 2004; Yuan et al., 2016). Studies showed that FTOHs might undergo
545 degradation and produce secondary chemicals such as polyfluoroalkyl carboxylic acids

546 (PFCAs), fluorotelomer aldehydes, and secondary polyfluorinated alcohols (Liu et al., 2007;
547 Liu et al., 2010; Zhao et al., 2017). For example, an aerobic biotransformation of 6:2 FTOH
548 (F(CF₂)₆CH₂CH₂OH) was reported to produce PFCAs including PFBA, PFPeA, and PFHxA,
549 *x*:3 acids, such as 5:3 acid (F-(CF₂)₅CH₂CH₂COOH), and 4:3 acids, such as (F-
550 (CF₂)₄CH₂CH₂COOH) (Zhao et al., 2013a). The profiles of FTOH degradation products varied
551 depending on the bacterial strains involved. For example, mixed bacterial strains yielded equal
552 amounts of PFCAs and *x*:3 acids, and a small amount of other transient intermediates (Liu et
553 al., 2010; Zhao et al., 2013a; Zhao et al., 2013b). In contrast, a single strain of *Pseudomonas*
554 sp. transformed 6:2 FTOH yielding a high quantity of transient intermediates and low quantities
555 of PFCAs and *x*:3 acids (Kim et al., 2012). Similarly, Tseng et al. (2014) indicated that a white-
556 rot fungus (*Phanerochaete chrysosporium*) degraded 6:2 FTOH yielding mainly 5:3 acids.
557 FTOHs biodegradation rates and pathways differed among aerobic/anoxic/anaerobic
558 conditions, with the first-order rate constants in the following decreasing order: aerobic >
559 anoxic > anaerobic conditions (Yu et al., 2016). The anaerobic biodegradation of FTOH was
560 found inefficient to produce PFCAs, but might form polyfluorinated acids (Zhang et al.,
561 2013b). FTOHs could also be transformed by terrestrial plants and animals. For example, Zhao
562 and Zhu (2017) observed that 10:2 FTOH was biotransformed to PFDA, PFNA, and PFOA by
563 soil microorganisms, PFDA, PFHxA and PFPeA by wheat roots, and PFDA and PFNA by
564 earthworms (*Eisenia foetida*). In the atmosphere, peroxy radical reactions could degrade
565 FTOHs producing a series of homologous PFCAs (Ellis et al., 2004), which also contributes to
566 widespread contamination of PFCAs in soil. Other fluorotelomer derivatives, such as
567 fluorotelomer sulfonate, polyfluoroalkyl phosphate, fluorotelomer acrylate, and fluorotelomer
568 stearate monoester, could act as precursors of FTOHs, which could subsequently be subjected
569 to the same degradation pathways as mentioned above to form PFCAs (Lee et al., 2010; Russell

570 et al., 2008; Butt et al., 2014; Wang et al., 2011; Lewis et al., 2016; Zhang et al., 2016b; Liu
571 and Liu, 2016; Dasu et al., 2012).

572 The potential PFAS precursors from electrochemical fluorination (ECF) include mixtures of
573 linear and branched isomers of perfluorooctane sulfonamide (FOSA), sulfonamido ethanol
574 (FOSE), FOSE-based phosphate diester (SAmPAP diester), and perfluoroalkyl sulfonamide
575 derivatives (e.g., *N*-methylperfluorooctane sulfonamidoethanol (MeFOSE), *N*-
576 ethylperfluorooctane sulfonamidoethanol (EtFOSE), *N*-ethylperfluorooctane sulfonamide
577 (EtFOSA), and *N*-methylperfluorooctane sulfonamidethylacrylate (MeFOSEA)) (Ruan et al.,
578 2015). A few studies confirmed that biotransformation of sulfonamide derivatives was an
579 indirect source of PFOS in soils. For example, Benskin et al. (2013) reported that SAmPAP
580 diester was persistent in marine sediments with an approximate half-life of >380 days at 25 °C,
581 whereas EtFOSE was transformed by bacteria to a number of products, including *N*-ethyl
582 perfluorooctanesulfonamido acetic acid (EtFOSAA), perfluorooctane sulfonamide acetate
583 (FOSAA), EtFOSA, FOSA, and PFOS (Benskin et al., 2013). Rhoads et al. (2008) proposed a
584 transformation route of EtFOSE as follows: EtFOSE → EtFOSAA → EtFOSA → FOSA →
585 perfluorooctane sulfinate (FOSI) → PFOS. The proposed biodegradation pathways of PFAS in
586 the soil system largely follow those reported in activated sludge and sediments. For instance,
587 Mejia-Avendaño and Liu (2015) investigated aerobic biotransformation of EtFOSE and
588 EtFOSA in soil. PFOS was identified from the biotransformation products of EtFOSA (4.0
589 mol%) after 182 days of aerobic incubation, which demonstrated that EtFOSE and EtFOSA
590 were precursors of PFOS in the soil environment. No further degradation of PFOS was reported
591 in the soil. Similarly, Zhao et al. (2016b) found that EtFOSE was sequentially transformed to
592 EtFOSAA, FOSAA, FOSA, and PFOS in an earthworm-soil system. Zhao et al. (2018) further
593 reported that FOSA could be degraded to PFOS in soil-wheat and soil-plant-earthworm
594 systems. Only a few studies to date have examined biotransformation of perfluoroalkyl

595 sulfonamide derivatives in soil. The available studies have not fully identified intermediate
596 products, suggesting that the proposed pathway of transformation of PFAS is still uncertain.
597 Considering the role of soil as a sink for PFAS in the environment, future studies are necessary
598 to examine the biotransformation of legacy and emerging PFASs in the soil environment.

599

600 **4. Remediation of PFAS in soil**

601 Remediating PFAS contaminated solid and aqueous media can be extremely challenging
602 because of the following key reasons: (1) high chemical and thermal stability of PFAS
603 compounds, (2) frequent occurrence of complex mixture of PFAS compounds in the
604 contaminated environment, (3) unique physicochemical properties of PFAS compounds (i.e.,
605 both hydrophobic and oleophobic behaviours), and (4) extremely persistent nature (i.e., limited
606 or no biodegradation). Although various methods have been reported to remove PFAS
607 compounds from aqueous media (Carter et al., 2010; Ding and Peijnenburg, 2013; Du et al.,
608 2014; Wagner et al., 2013; Zhang et al., 2011), these methods may not be readily applicable
609 for remediating PFAS contaminated soils or waste materials (e.g., biosolids) (Darlington et al.,
610 2018). Two broad approaches, namely mobilization and immobilization, using soil
611 amendments, as discussed below, may prove logistically and economically viable for the
612 remediation of PFAS contaminated soil. While an immobilization approach can be used to
613 reduce mobility and bioavailability of PFAS, the mobilization approach can be used to remove
614 PFAS through soil washing and phytoremediation. Both these two approaches can facilitate
615 the destruction of PFAS from soil through abiotic and biotic degradation processes.

616

617 **4.1 Mobilization of PFAS compounds**

618 The key mechanisms involved in the mobilization of contaminants in soils/sediments include
619 solubilisation, desorption, and complexation reactions. Mobilization processes can be used to

620 remove contaminants from soil through washing (i.e., soil flushing) and plant uptake (i.e.,
621 phytoremediation). In the case of organic contaminants such as PFAS, mobilization processes
622 can be facilitated using various soil amendments (Table 3), which lead to the complete
623 destruction of these contaminants through abiotic and biotic degradation reactions.

624

625 *4.1.1 Soil flushing and soil washing*

626 Soil flushing is an *in-situ* process that involves injection of a flushing solution into the ground
627 for extracting contaminants (Hale et al., 2017; Yao et al., 2015). The main advantage of soil
628 flushing is that large quantities of soil can be treated *in-situ* without the need for excavation
629 and transport (Jawitz et al., 2000; Svab et al., 2009; Zheng et al., 2012). Surfactants having
630 both hydrophobic and hydrophilic structural groups are used to facilitate the desorption of
631 persistent organic pollutants (POPs) and subsequent soil flushing. However, many PFAS,
632 including PFOS and PFOA, are themselves surfactants, which can make PFAS behaviour
633 difficult to predict during PFAS mobilization. For example, Pan et al. (2009) found that a
634 cationic surfactant (cetyltrimethylammonium bromide (CTAB)) was able to significantly
635 enhance the sorption of PFOS to sediments due to the initial sorption of CTAB to sediments,
636 thereby exposing CTAB's hydrophobic tails to adsorb PFOS. However, an anionic surfactant
637 (sodium dodecyl-benzene sulfonate (SDBS)) showed a concentration-dependent effect where
638 a SDBS concentration <4.34 mg/L increased PFOS sorption to sediments, but SDBS
639 concentration >21.7 mg/L increased PFOS desorption (Pan et al., 2009). Guelfo and Higgins
640 (2013) found that an anionic surfactant (sodium dodecyl sulphate (SDS)) at low concentration
641 decreased the sorption of PFOS, PFNA, and PFDA, but increased the sorption of long chain
642 PFAS, such as PFBA, PFPeA, PFHxA, PFHpA, and PFBS. In general, anionic surfactants
643 could enhance the solubility of PFAS in water, which would decrease PFAS sorption to
644 soils/sediments and, thereby, facilitate PFAS mobilization.

645 Other common soil flushing additives such as organic/inorganic acids/bases and solvents such
646 as methanol or ethanol might be suitable for removing PFAS from soils. For example, Schröder
647 (2003) applied organic solvents (e.g., ethyl acetate (EtOAc), dimethylformamide (DMF),
648 pyridine, tert-butyl methyl ether (MTBE), 1,4-dioxane, or tetrahydrofuran (THF)) to determine
649 the ability of a solvent or combination of solvents for sludge PFAS extraction. Under
650 pressurised solvent extraction at 150 °C and 143 bar, a sequential flushing with a mixture of
651 EtOAc and DMF followed by methanol modified with phosphoric acid appeared to be the most
652 effective extractants of sludge PFAS. Omitting DMF from the flushing mixture made the
653 procedure greener without significantly reducing the PFAS extraction efficiency.

654 Advanced methods such as reverse osmosis (RO) and supercritical fluid (SCF) assisted
655 extraction using comparatively non-toxic organic solvents were also attempted for removing
656 PFAS from aqueous and solid matrices, respectively. For example, Tang et al. (2006) found
657 that isopropyl alcohol increased the solubility of PFOS during reverse osmosis (RO), but
658 decreased the membrane flux. Chen et al. (2012a) reported that at a critical point of CO₂ (50
659 °C, 20.3 MPa), HNO₃ (16 N) first suppressed the polarity of PFOS and PFOA molecules
660 increasing their solubility in supercritical CO₂, and then methanol led to increased mobilization
661 of PFOS and PFOA from a sand matrix with 59 and 77% extraction efficiencies, respectively,
662 and from paper and fabric with 80 and 100% efficiencies, respectively.

663 The first stage of soil washing would concentrate PFAS of soils/sediments into a solution. Once
664 the contaminant is flushed out and collected, the solution could be decontaminated using water
665 treatment technologies such as sorption, ion-exchange, or filtration including RO technique for
666 reuse or safe disposal. Special research in the future should be given to developing
667 environmentally benign treatment methods, such as using natural and green adsorbents and
668 supercritical CO₂ extraction.

669

670 4.1.2 Phytoremediation

671 Unlike other POPs, PFAS are relatively soluble and remain in the soil solution, leading to their
672 ready uptake by plants and subsequent removal using phytoremediation technology. The plant
673 uptake of PFAS compounds is dependent on their chain length and the sorption behaviour of
674 soils. Promising PFAS compounds suitable for phytoremediation include those with relatively
675 low log K_{ow} and a small C chain. For example, the mass uptake of PFBA was calculated to be
676 high at 11.27 mg/m²/year, assuming log K_{ow} (0.001), concentration (0.1 mg/L), transpiration
677 (500 L/m²/year), and fractional water use (0.3) (Austin et al., 2017). The carbon chain length
678 (6 or less) is possibly a more appropriate screening metric for phytoextraction than log K_{ow} , as
679 the short chain PFASs show the highest TSCF values (Austin et al., 2017).

680 Huff et al. (2019) considered the bioconcentration factor (BCF) as a key metric in assessing
681 the suitability of plant species for phytoremediation of contaminated sites. They demonstrated
682 hyperaccumulation of multiple PFAS compounds; a greater than 10-fold soil to leaf
683 translocation of PFAS in above-ground plant tissues was exhibited. The BCFs of PFOA
684 hyperaccumulating species ranged from 11.5 to 46.5, and that of PFOS ranged from 10.3 to
685 17.9. They also showed higher plant uptake of PFOA and PFOS (44 – 344%) with the
686 application of a proprietary soil amendment than without the amendment. Similarly, Gobelius
687 et al. (2017) reported successful uptake of 26 PFAS compounds in plants from contaminated
688 soils around a firefighting training site in Stockholm.

689 Phytoremediation of a PFAS contaminated site can be a slow process, but it involves low
690 capital cost and almost no maintenance cost. Thus, phytoremediation remains the most
691 economic and sustainable green technology available for the remediation of PFAS
692 contaminated sites.

693

694 4.2 Immobilization of PFAS compounds

695 The immobilization technique redistributes PFAS contaminants from the solution to solid
696 phase, thereby reducing their mobility and bioavailability. A range of amendments have been
697 tested to enhance the immobilization of PFAS contaminants in soils and sediments (Table 4).

698

699 *4.2.1 Sorption*

700 The materials used for sorbing PFAS in soil and water mainly include carbon-based and clay-
701 based materials, ionic surfactants, and anion-exchange resins (Ahrens and Bundschuh, 2014;
702 Yu et al., 2009; Senevirathna et al., 2010). Carbon-based materials include activated carbon
703 (AC), multi-walled carbon nanotubes (CNTs) and biochars (Darlington et al., 2018; Liu et al.,
704 2019). Of these, CNT and AC were reported to show remarkable PFAS sorption capacity
705 (Ochoa-Herrera et al., 2008; Sarkar et al., 2018; Wagner et al., 2013; Xiao et al., 2017). The
706 non-polar functional groups of carbon-based materials enable them to be highly useful for
707 hydrophobic PFAS sorption.

708 Powdered or granulated AC (GAC) was effective in removing PFOA and PFOS in soil and
709 water (Cummings et al., 2015). A small number of commercial adsorbents (e.g., RemBind™,
710 PefluorAd) available on the market could be used as a complement or as an alternative of AC
711 (Birk, 2016). Clay mineral candidates, such as montmorillonite (Zhou et al., 2010; Zhang et
712 al., 2014), kaolinite (Zhang et al., 2014; Zhao et al., 2014), hematite (Zhao et al., 2014), alumina
713 (Wang and Shih, 2011), and boehmite (Wang et al., 2012), were used for batch-scale removal
714 of PFAS compounds, specially focussing on PFOS. For example, Hale et al. (2017) tested AC,
715 compost soil, and montmorillonite to immobilise PFAS in contaminated soils. PFAS leaching
716 was reduced by 94–99.9, 29–34, and 28–40% for AC, compost, and montmorillonite
717 amendments, respectively. Natural clay minerals have a hydrophilic surface, which is
718 intrinsically negatively charged, rendering them ineffective for hydrophobic and anionic PFAS
719 removal. However, when modified with a surfactant, its hydrophobic alkyl chain enhances

720 PFAS sorption through hydrophobic partitioning. Additionally, in contrast to natural clay
721 minerals, quaternary ammonium surfactant-modified products generate positive surface charge
722 (Sarkar et al., 2011; 2012), which could attract anionic PFOS and PFOA via electrostatic
723 interaction. Das et al. (2013) reported the immobilization and reduced leaching of PFOS
724 (>90%) from four AFFF-contaminated soils using a palygorskite-based organoclay prepared
725 with oleylamine. Like organoclays, a swellable organically modified silica was shown to
726 outperform GAC for PFAS sorption (Figure 5), including short-chain PFAA compounds
727 (Stebel et al., 2019). However, care should be taken in choosing the organic agents for
728 modifying mineral materials, because chemicals such as oleylamine or hexadecyltrimethyl
729 ammonium (HDTMA) can be extremely toxic to native soil micro- and macro-organisms
730 (Sarkar et al., 2010; 2013).

731 Some liquid-based amendments were also used to facilitate PFAS immobilisation in
732 contaminated solid media. For example, Aly et al. (2019) observed a six-fold increase of PFAS
733 retention in contaminated soils by applying a commercially available coagulant. Similarly, Pan
734 et al. (2009) calculated the thermodynamic index of irreversibility (TII) for PFOS to sediments
735 in the presence of a cationic surfactant, cetyltrimethylammonium bromide (CTAB), to quantify
736 the degree of sorption irreversibility caused by CTAB, with a value of 0 representing a highly
737 reversible system and 1 representing irreversible sorption. A value of 1 was approached for
738 CTAB concentrations of 18.1 and 36.1 mg/L, indicating its potential use for PFOS
739 immobilization. The cationic surfactant could be delivered to the source of pollution using *in*
740 *situ* percolation or injection.

741 Many factors including media characteristics, PFAS characteristics, and adsorbent
742 characteristics would influence the overall effectiveness of PFAS remediation via sorption
743 treatments (Darlington et al., 2018). The pH of the solid media (e.g., soil, biosolid), as well as
744 their concentrations of inorganic and organic ions, significantly control the sorption efficiency.

745 PFAS sorption decreases with increasing pH of the medium. While natural organic matter in
746 the soil does not reduce the PFAS sorption capacity of activated carbon and clays, high ionic
747 strength (i.e., high concentration of inorganic salts) adversely affects the sorption capacity of
748 organoclays through the adsorbents' charge reversal behaviour (Das et al., 2013; Jeon et al.,
749 2011; Sarkar et al., 2012). The presence of sulfonate functional groups can lead to strong
750 sorption of PFAS. Physicochemical characteristics of adsorbents (e.g., SSA, pore size
751 distribution, cation exchange capacity, zeta potential) also have a remarkable effect on PFAS
752 sorption (Du et al., 2014; Jeon et al., 2011). Adsorbents with small pore size and high SSA can
753 lead to high PFAS sorption capacity. Similarly, adsorbents with a basic or positively charged
754 surface tend to show high PFAS sorption capacity through the combined mechanism of
755 hydrophobic interaction and electrostatic attraction (Lu et al., 2016).

756

757 *4.2.2 Stabilization and solidification*

758 Stabilization and solidification (S/S) of contaminants including PFAS can be achieved by
759 applying cementitious binders and additives into the contaminated soil, sediment, and waste
760 media (Bates et al., 2000; Fagerlund et al., 2019; Söregård et al., 2019a; b). The
761 stabilizing/solidifying agents immobilize contaminants via physical and/or chemical
762 protection. In the case of physical protection, contaminant leaching is prevented by reducing
763 the hydraulic conductivity of the system. In the case of chemical protection, contaminants are
764 stabilized by reducing their aqueous solubility through precipitation, redox alteration, and
765 sorption reactions. The S/S technique undertaken *in situ* or *ex situ* can prove efficient in terms
766 of treatment performance and costs when contamination over a large area requires remediation
767 (Fagerlund et al., 2019; Söregård et al., 2019a; b). A limited number of reports are available
768 on PFAS site remediation using the S/S method. Söregård et al. (2019a) examined seven
769 additives, namely pulverised activated carbon (PAC), Rembind[®], powdered zeolite, chitosan,

770 hydrotalcite, bentonite, and CaCl₂ at 2% application rate, for stabilizing a total of 14 PFAS
771 compounds in an aged-contaminated soil. The PAC and Rembind[®] additives performed the
772 best with, respectively, 70 and 94% reduction of leaching achieved for all the concerned PFAS
773 compounds except perfluorobutane sulfonate (PFBA). Highly persistent and bioaccumulative
774 long-chained PFAS (e.g., PFOS) was stabilized by 99.9% by PAC or Rembind[®] application.
775 The chain length and functional groups present in the PFAS compounds influenced the
776 additives' PFAS stabilization efficiency. The stabilization capacity increased by 11–15 % per
777 CF₃-moeity, and the stabilization of perfluorosulfonates (PFSAs) was 49% higher than
778 perfluorocarboxylates (PFCAs). During the soil PFAS stabilization process, PAC and
779 Rembind[®] did not show any significant impact on the physical matrix stability (Orengarda et
780 al., 2019).

781

782 4.3 Destruction of PFAS compounds

783 A complete remediation of PFAS contaminated soils can be achieved by complete destruction
784 of PFAS compounds through biotic (e.g., biodegradation) and/or abiotic (e.g., thermal
785 oxidation, chemo oxidation, ball milling) degradation processes, as discussed below.

786

787 *4.3.1 Bioremediation*

788 Biodegradation via microorganisms can be an effective and efficient method to remediate soils
789 and groundwater contaminated with many organic pollutants (Fahid et al., 2020; Wang et al.,
790 2019). An aerobic environment promotes the production of perfluorinated compounds (PFCs)
791 from relevant precursors through microbial transformation (Dasu and Lee, 2016, Liu and
792 Mejia-Avenidaño, 2013). However, PFOS and PFOA are reported to be strongly resistant to
793 microbial transformation under aerobic environments, and only a few reports are available on

794 this topic (Liu and Mejia-Avendaño, 2013; Pasquini et al., 2013; Ochoa-Herrera et al., 2016;
795 Liu et al., 2010; Chetverikov et al., 2017).

796 Biodegradation of PFAS in soil depends on the nature of microbial composition. For instance,
797 Pasquini et al. (2013) discovered that *E. coli* was not able to biodegrade PFOS and PFOA. In
798 contrast, *P. plecoglossicida* (Chetverikov et al., 2017), *P. parafulva* (Yi et al., 2016),
799 *Acidimicrobium sp.* (Huang and Jaffe, 2019) and *P. aeruginosa* (Kwon et al., 2014) led to a
800 significant reduction of the PFOS concentration. Beskoski et al. (2018) reported that
801 chemoorganoheterotrophic bacteria, as well as yeast and molds, could reduce PFOA and PFOS
802 moderately. A few other studies investigated the degradation potential of various PFCs using
803 microbial cultures of sludges, industrial site sediments, contaminated and uncontaminated
804 soils, and waste-water treatment plants; however, the exact bacterial composition was often not
805 clarified (e.g., Ochoa-Herrera et al., 2016; Liu et al., 2010; Mejia-Avendaño et al., 2016). Apart
806 from the bacterial composition, environmental factors that include pH and soil solution
807 composition are important for the optimal degradation rate of PFAS compounds.

808 Several studies reported that the degradation of PFCs is limited to the non-fluorinated moiety;
809 therefore, defluorination by biodegradation seems not possible (Mejia-Avendaño et al., 2016;
810 Dimitrov et al., 2004). Defluorination is an important process to turn PFC molecules largely
811 harmless. Nevertheless, various studies detected the release of fluorine ions (Beskoski et al.,
812 2018; Chetverikov et al., 2017; Liu et al., 2010; Ochoa-Herrera et al., 2016). Ochoa-Herrera et
813 al. (2016) reported a 3% release of the total fluorine ions of the PFOS concentration with an
814 aerobic microbial treatment. They speculated that the release of fluorine ions was due to the
815 degradation of fluorinated impurities caused by the low purity of PFOS (96%). Another study
816 found a low release of fluorine ions during the degradation of PFOA (96% purity) with
817 anaerobic microorganisms (Liu et al., 2010).

818 To date, the defluorination of PFCs by microbial biodegradation has not been reliably verified
819 or falsified, but this does not imply an entire biodegradation of PFCs. Mejia-Avenidaño et al.
820 (2016) examined the aerobic biotransformation of perfluorooctane sulfonamide quaternary
821 ammonium salt (PFOSAmS) and perfluorooctaneamido quaternary ammonium salt
822 (PFOAAmS), whereby the PFOAAmS concentration was reduced to 43% of the initial amount
823 while PFOA increased. A parallel run without microorganisms, which showed no change in
824 PFOAAmS or PFOA concentration, indicated that these results were related to biodegradation.
825 In the experiment with microorganisms PFOAAmS decreased and PFOA increased and
826 without microorganisms the PFOAAmS and PFOA concentration remained the same. Mejia-
827 Avenidaño (2016) explained this phenomenon with the biodegradation of PFOAAmS to PFOA.
828 This study demonstrated that biodegradation was possible, although this only involved the
829 breakdown of the non-fluorinated moiety. However, to our knowledge, no field or *in situ*
830 experiments have been conducted on PFC biodegradation. Therefore, further research should
831 focus on *in situ* implementation of PFC biodegradation, as previous studies were only
832 conducted at the laboratory level. Furthermore, in order to understand fully the whole process,
833 all degradation pathways and their environmental impact should be investigated in the future.

834

835 4.3.2 Chemical and thermal treatment

836 In general, PFOS and PFOA destruction using common water oxidative/disinfection methods
837 (e.g., chloramination, chlorination, ozonation, chemical oxidation and ultraviolet treatment)
838 was found ineffective in most cases (Higgins and Dickenson, 2016; USEPA, 2016a; b).
839 However, the removal of PFAS compounds from soil, waste, and water sources can be achieved
840 through chemical and thermal redox reactions. For example, laboratory scale destruction of
841 PFAS was achieved through catalytic and electrocatalytic oxidation using anodic mixed metal
842 oxides (e.g., Ti/RuO₂) (Lin et al., 2012). However, the presence of naturally occurring DOM

843 might significantly restrict the degradation rate of PFAS in soil via chemical oxidation (Buxton,
844 1988). The low reduction potential of fluorine ($E < -2.7V$) as such makes the defluorination
845 reaction thermodynamically unfavorable. However, sub-critical reduction of certain elements
846 (e.g., Fe) at high temperature and pressure could lead to PFOS oxidation at least in laboratory
847 and bench scales, but it is not so feasible for *in situ* application.

848 *In-situ* chemical oxidation (ISCO) using a peroxydisulfate process has been previously used
849 for the remediation of pollutants such as chlorinated ethenes and benzenes, oxygenates,
850 benzene, toluene, ethylbenzene, xylenes (BTEXs), and PAHs from soil (Nadim et al., 2006;
851 Tsitonaki et al., 2010). Formation of the activated persulfate radicals can be accomplished
852 through UV exposure, heat, high pH (alkaline conditions), hydrogen peroxide, and a variety of
853 transition metals (Watts and Teel, 2006). Thus, persulfate first can be delivered to the
854 contaminated soil subsurface in an inactive form, and then activated when it comes in contact
855 with the contaminated zone. Activation by heat can be accomplished using steam injection or
856 thermal energy production using electrodes (Heine et al., 1999).

857 Hori et al. (2008) found that persulfate oxidation, activated by hot water, was effective at
858 degrading PFOA to below the detection limit after 6 h of treatment at 80 °C. Lee et al. (2012)
859 were able to achieve a complete persulphate-induced degradation of PFOA at pH = 2.5 after
860 72 h at 40 °C, and 215 h at 30 °C. Hawley et al. (2012) examined the activation of persulfate
861 oxidation with the goal of soil and groundwater treatment. Activators that were able to degrade
862 PFOS by more than 97.5% included the following: Fenton's reagent, peroxide- activated
863 persulfate, and heat-activated persulfate. Strong reducing agents such as sodium dithionite and
864 sodium hypophosphate were also tested, but only partial degradation of PFOS was observed.

865 They attempted a 'Smart Combination *In-situ* Oxidation/Reduction (SCISOR)' technique
866 involving a combination of redox agents and activators, which achieved 60% PFOS removal
867 after one contact phase. However, Place and Field (2012) expressed concern regarding the use

868 of *in-situ* chemical oxidation (ISCO) for the remediation of AFFF-impacted sites, because
869 advanced oxidation techniques have been known to facilitate PFAS and PFCA formation from
870 the more complex precursors present in AFFFs. Pancras et al. (2013) demonstrated that the
871 combination of SCISOR and soil washing techniques was able to achieve > 99% removal of
872 PFOS from contaminated soil. Similarly, electrochemical oxidation involving a specific anode-
873 electrolyte combination was found effective in degrading PFAS at <200 mg/L concentrations
874 in a bench-scale reactor (Niu et al., 2016).

875 Although chemical treatments involving chloramination, chlorination, ozonation and oxidation
876 reactions have been found to be effective in the removal and destruction of PFAS compounds,
877 it may not be applicable to large-scale remediation of PFAS contaminated soils under field
878 conditions. It involves expensive chemicals and results in residual chemicals which may cause
879 environmental degradation and issues with safe disposal (Higgins and Dickenson, 2016).

880 Thermal treatments of PFAS contaminated soil include both complete degradation of PFAS
881 requiring high temperature (900-1100 °C) (Watanabe et al., 2016), and thermal desorption (Lim
882 et al., 2016). The complete thermal degradation is covered under '*Vitrification or incineration*'
883 (Section 4.3.3). The thermal desorption technique involves *ex situ* or *in situ* heating of PFAS
884 contaminated soils, and the subsequent removal of vaporised PFAS compounds through air
885 filtration. For example, Söregård et al. (2020) have been able to achieve 71-99% thermal
886 desorption of PFAS in a field contaminated soil at 550 °C, and >99% desorption in a PFAS
887 fortified soil.

888 4.3.3 *Vitrification or incineration*

889 Temperatures required for vitrification of PFAS compounds range between 1600 °C to 2000
890 °C. An advantage of this process in the context of PFAS is the lack of by-products generated,
891 because all organic contaminants are fully destroyed. For example, Yamada et al. (2005) tested
892 whether fluorotelomer-treated textiles and paper, after being destroyed under municipal

893 incinerator conditions, would form PFOA as a degradation product. There was no PFOA
894 detected in the samples after incineration. Even if PFOA was formed during incineration, it
895 must have also been destroyed in the process, meaning degradation of precursors during
896 incineration would not be a significant PFOA source in the environment. There are several
897 processes available to reach vitrification temperatures: electrical, thermal, and plasma. The
898 electrical process is *in situ* and involves construction of a zone surrounded by graphite
899 electrodes inserted in the ground which pass energy through the soil. The thermal process is *ex*
900 *situ* and is generally carried out in a rotary kiln. Plasma processes are only necessary when
901 temperatures of up to 5000 °C are required. Electrical or thermal processes could be used to
902 target the combustion of PFAS on site without using more extreme temperatures.

903

904 4.3.4 *Ball milling*

905 Mechanochemical destruction (MCD), or high energy ball milling, has recently gained
906 attraction in the soil remediation sector due to its ability to destroy effectively POPs with no
907 requirement for toxic solvents, extreme temperatures, harmful additives, or high pressures
908 (Cagnetta et al., 2016). The mechanisms involved in mechanochemical transformations tend
909 to be complex and dissimilar to reactions observed in other remediation processes, i.e., thermal,
910 photochemical, and chemical oxidation. While heat is generated under ball milling conditions,
911 it was disregarded as the sole reaction initiation mechanism because of the progression of
912 mechanochemical reactions even at very low temperatures (i.e., 77 K) (Beyer et al., 2005).
913 Rather, the formation of free radicals and matrix defects by strong mechanical activation (i.e.,
914 grinding) are the main reaction initiators (Sohma, 1989). These ‘mechanoradicals’ are capable
915 of accelerated chemical transformation and destruction of POPs by high energy ball milling.
916 Centrifugal and planetary ball mills are commonly utilised in laboratory-scale research (Li et
917 al., 2017; Nomura et al., 2012; Zhang et al., 2013a).

918 Although POP degradation pathways by ball milling are complex and not fully understood,
919 most published trials show high rates of destruction between 99-100% for a wide range of POPs
920 and organic contaminants subjected to mechanochemical conditions (Cagnetta et al., 2016).
921 While the focus of MCD has historically been directed toward chlorinated POPs, several papers
922 have recently been published related to PFAS degradation by high energy ball milling. Work
923 by Zhang and colleagues (2013a) revealed degradation efficiencies of 100% for PFOA at 180
924 min and 99.88% for PFOS at 360 min. The destruction of PFOS and PFOA was carried out in
925 a range of supporting matrices including CaO, SiO₂, Fe-Si mix, NaOH, and KOH. The most
926 complete PFAS degradation was obtained with KOH as the support matrix, showing
927 conversion of organic fluoride (C-F) to negatively charged monovalent fluoride (F⁻). Zhang
928 and colleagues (2016a) repeated the same trial with a Chinese PFOS alternative known as F-
929 53B (6:2 chlorinated polyfluoroalkyl ether sulfonate), and they achieved a more rapid rate of
930 degradation due in part to the substitution of a single fluorine with a chlorine on the terminal
931 carbon and the introduction of an ether group. Further work by Lu et al. (2017) successfully
932 destroyed 6:2 fluorotelomer sulfonate within 60 min, again using KOH as the support. Citing
933 the concerns over the amount of KOH required for matrix support, Cagnetta et al. (2017)
934 degraded a range of PFAS compounds and switched the support matrix to La₂O₃. While
935 effective, La₂O₃ cannot be used for soil remediation due to its high cost, and low concentration
936 of PFAS in soil would lead to insignificant amounts of lanthanum oxyfluoride formed.
937 As a non-thermal technology requiring only mechanical energy input, ball milling presents a
938 compelling treatment option for PFAS-impacted soil and hazardous waste. Further research is
939 required to determine its effectiveness as a *bona fide* remediation technology, especially related
940 to PFAS degradation mechanisms, kinetic reaction progression, fluoride fate, and scaling
941 factors.
942

943 **5. Case studies of field application of remediation technologies**

944 In this section, selected 5 selected case studies involving the field application of mobilization
945 (soil washing and phytoremediation), immobilization (stabilization/solidification), and
946 destruction (thermal oxidation and chemical oxidation) techniques of remediating soil
947 contaminated with PFAS compounds are given. The reduction of the mobility and
948 bioavailability, and ultimate removal, of PFAS compounds are discussed.

949

950 **5.1 Case study 1: Mobilization and soil washing**

951 In this method, water is used to extract PFAS from contaminated soil without any use of
952 additional chemicals. The method is being applied in several projects using mobile equipment
953 operated by a Swedish company, Svevia. Contaminated soil is excavated and loaded to a
954 container where it is mixed with water. A high-pressure water stream is used to scrub the fines
955 from coarser soil particles and dissolve some of the contaminants. Fines are then separated
956 from coarser fractions for further management. The coarse (washed) particles are ejected and
957 placed back to the pit after concentration of contaminants is checked through sampling and
958 analysis. The method was tested on a pilot scale (10 tonnes) where 96% separation of PFOS
959 from soil particles was achieved (Swedish EPA, 2018).

960 Soil washing was then applied on a full scale to soil from a site in Kalmar, Sweden, where
961 firefighting exercises have been carried out for 10 years. The aim was to wash the soil in order
962 to separate PFAS from soil particles, place cleaned particles back to the site, and clean the
963 washing solution. Because PFAS are readily water soluble, it was expected that most of PFAS
964 from soil could be removed and that the remediation goals could be reached. Fifty tonnes of
965 soil were washed per batch. In the first two batches the achieved PFOS concentration in washed
966 soil was 17µg/kg (remediation goal was set to 29 µg/kg). In the following two batches the
967 remaining concentration in washed soil was above the remediation goal. The washing solution

968 was first cleaned using ozonation, but it was not sufficient to reduce PFAS concentrations and
969 active carbon filters were applied. Only 10% (150 of 1500 tonnes) of the planned soil volume
970 got treated. The main obstacle to proceed with the clean-up was a high fraction of clayey soil
971 particles that hampered the equipment (Johansson, 2019).

972

973 5.2 Case study 2: Mobilization and phytoremediation

974 Gobelius et al. (2017) conducted a major study on the plant uptake of PFAS at a contaminated
975 site of fire training facility at Stockholm Arlanda airport, to assess PFAS phytoremediation
976 potential of a range of plant species under field conditions. The extent of contamination of soil
977 and groundwater and the uptake of 26 PFASs by plants were evaluated. Based on the
978 bioaccumulation factor and biomass production, they proposed three scenarios to estimate the
979 PFAS phytoextraction efficiency and remediation period. The first scenario comprised a shelter
980 wood of mixed silver birch (~66%) and Norway spruce (~33%) stands. Considering this
981 system with frequent harvest of shoot and birch sap, and an understory of ground elder, they
982 estimated that it was possible to remove annually 1.4 g of $\sum 26$ PFASs/ha. The second scenario
983 was the regular coppicing of birches, in which the tree trunks were left in the field but the twigs
984 and leaves were harvested in every 3–5 years of rotation. Annually 5 tonnes/ha of biomass
985 composed of twigs and leaves could be generated, leading to annually 0.65 g of extractable
986 $\sum 26$ PFASs for birches. The third scenario was the preservation of a meadow composed of plant
987 species with high PFAS uptake potential. Accordingly, the long beech fern (*Phegopteris*
988 *connectilis*) and ground elder (*Aegopodium podagraria*) are practical choices, extracting
989 annually 0.55 g/ha of $\sum 26$ PFASs if mowed regularly.

990 Results of this field study suggested that, while plant uptake thresholds for plant species
991 investigated in this study remained uncertain, the first scenario with a shelter wood appeared
992 an ideal remediation option with the highest PFASs uptake. Taking into consideration the

993 decreasing concentrations of PFOS over time and the threshold values for PFOS in soils (i.e.,
994 0.003 µg/kg for sensitive land use and 0.02 µg/kg for non-sensitive land use), the time period
995 required for remediating the soil at the contaminated site was estimated. It was estimated that
996 the uptake of PFOS by spruces and birches would require 48,000 years and 160,000 years,
997 respectively, to reach the threshold value for sensitive land use or 18,000 years and 58,000
998 years, respectively, for the non-sensitive land use. Assuming similar toxicity of the other 25
999 PFASs as for PFOS, the threshold values for $\Sigma 26$ PFASs would be 0.078 µg/kg for sensitive
1000 land use and 0.52 µg/kg for non-sensitive land use. Thus, the uptake of $\Sigma 26$ PFASs by birches
1001 and spruces would require <45 years for the remediation of the site due to the greater uptake
1002 potential, in particular for PFBA, PFNA, PFHxS, and 6:2 FTSA.

1003

1004 5.3 Case study 3: Immobilization and stabilisation/solidification

1005 The method was tested in Sweden for stabilization of PFAS in a soil matrix using various
1006 binders, such as combination of Portland cement, fly ash, and ground granulated blast-furnace
1007 base slag (9:1 soil to binder ratio), as well as commercially available stabilizers such as
1008 pulverised activated carbon, Rembind®, powdered zeolite, chitosan, hydrotalcite, bentonite,
1009 and calcium chloride (Söregård et al., 2019a). The leaching of various PFAS compounds
1010 decreased on average by 70% and was larger for longer carbon chains. For PFOS the decrease
1011 was 99.9%. Six tonnes of PFAS contaminated soil ($\Sigma 11$ PFAS=160 µg/kg) from an industrial
1012 site were then treated on a pilot scale using 10% Portland-fly ash cement with an addition of
1013 2% granular activated carbon. Solidified soil monoliths (1.2 m³) are being leached with water
1014 simulating 15 years precipitation. According to the preliminary results, the leaching of PFOS
1015 and PFOA decreased by 98% (Kleja et al., 2020).

1016 In a recent study, two soils affected by AFFF training activities were treated with a combination
1017 of no-treatment, soil plus granular activated carbon (GAC), or soil plus GAC and general

1018 purpose Portland cement and aged 30 days before being subjected to simulated acidic leaching
1019 by a Synthetic Precipitation Leaching Procedure (SPLP) (USEPA, 2019). A total of 24 analytes
1020 were monitored in a soil leachability testing, including perfluoroalkyl acids (PFAAs), such as
1021 perfluorinated carboxylates (PFCAs) and perfluorinated sulfonates (PFSAs), and
1022 precursors/intermediates such as perfluorooctyl sulfonamide (FOSA), N-methyl/ethyl
1023 perfluorooctyl sulfonamidoacetic acids (N-MeFOSAA, N-EtFOSAA), and fluorotelomer
1024 sulfonates (FTS) (4:2 FTS, 6:2 FTS and 8:2 FTS). For most of the PFAS analytes, addition of
1025 GAC to the soils dramatically decreased the leachability of the PFAS compounds present in
1026 the soils (stabilization), and further reduced the leachability of PFAS compounds with the
1027 addition of cement as a binder (solidification) for about one third of the PFAS present in these
1028 contaminated soils. Overall percent immobilization of PFAS that were detectable in the
1029 leachate from treated soils ranged from 87.1% to 99.9%.

1030 In another case study in 2015, RemBind Pty Ltd ([https:// https://rembind.com/](https://rembind.com/)) investigated
1031 the potential value of stabilization techniques to remediate 1,000 tonnes of PFAS contaminated
1032 soil originated from two airport sites. In this work, RemBind[®] material was applied to stabilize
1033 PFAS in soil, thereby reducing its leachability to allow for safe disposal of the contaminated
1034 soil to landfill with regulatory approval. The lab-scale tests demonstrated that 5% (w/w)
1035 RemBind[®] addition was effective in reducing the leachate PFAS concentrations to below the
1036 target criteria of <0.2 µg/L.

1037 After treatment of the contaminated soil with RemBind[®], the approval for safe disposal
1038 validation results indicated that the leachate PFAS concentrations reduced to the level of
1039 reporting (LOR; 0.01 µg/L). Based on these results, permission was obtained for the safe
1040 disposal of treated soil to a lined landfill without any requirements for remediation or
1041 management. For the safe disposal of the PFAS contaminated soil, a 100 cm layer of pure
1042 RemBind[®] was spread in the bottom as an additional level of risk mitigation (SI Figure 5). The

1043 RemBind[®] amended soil was spread on a layer of the RemBind[®] bottom liner and covered with
1044 another 100 cm layer of pure RemBind[®]. This is one of the major large scale (1,000 tonnes)
1045 PFAS soil disposal projects completed in Australia with EPA regulatory approval. This field-
1046 based study provided a proof of concept for the application of immobilization agents as a cost-
1047 effective remediation technology for the sustainable management of PFAS contaminated soil.
1048 Since this initial work, RemBind[®] has been used at full commercial scale to treat 1,000's of
1049 tonnes of PFAS contaminated soil in Sweden ([https://rembind.com/projects/remediation-of-](https://rembind.com/projects/remediation-of-pfas-impacted-soil-at-a-fire-station-in-sweden/)
1050 [pfas-impacted-soil-at-a-fire-station-in-sweden/](https://rembind.com/projects/remediation-of-pfas-impacted-soil-at-a-fire-station-in-sweden/)) and , more recently, at the Townsville RAAF
1051 Base in Queensland, Australia (<https://rembind.com/projects/full-scale-pfas-remediation/>).

1052

1053 5.4 Case study 4: Destruction by thermal oxidation

1054 Endpoint (2017) introduced a new technology named as Vapor Energy Generator (VEG) and
1055 applied it practically on a small scale for thermal treatment of PFAS contaminated soil. The
1056 instrument delivered thermal energy via recycled water and propane to produce steam at 1100
1057 °C for PFAS treatment (700 °C for other chemicals). Endpoint Consulting Inc.
1058 (http://www.endpoint-inc.com/contact_us.htm) in collaboration with the Colorado School of
1059 Mines conducted a VEG trial on PFAS contaminated spiked soil to examine the treatment
1060 effectiveness at various temperature ranges. Operation at 950 °C for 30 min removed several
1061 groups of PFAS together with PFOS with 99% efficiency (Endpoint, 2017). In response to the
1062 increasing PFAS concentration in the environment, Clean Earth
1063 (<https://cleanearthinc.com/locations>) has started and applied thermal desorption as a feasible
1064 method to eliminate PFAS from soils. This method has been applied by Clean Earth since 1992
1065 to remove diverse groups of organic contaminants from the environment. Under the guidance
1066 of the New York State Department of Environmental Conservation (NYSDEC), 22.6 tonnes of
1067 contaminated soil at Clean Earth's Fort Edward, New York facility were decontaminated in

1068 December 2018 and February 2019. Based on this opening trial of thermal desorption
1069 technology for treatment of PFAS-contaminated soil, Clean Earth is continuously working with
1070 state agencies to launch standards to establish the effectiveness of this technology and its
1071 optimization (<https://cleanearthinc.com/what-we-handle/pfas-contaminated-soils>).
1072 Enviropacific is an Australian (<https://enviropacific.com.au/about-us/>) owned industry and a
1073 leader in providing waste management services, which have wide-ranging thermal treatment
1074 abilities for PFAS-contaminated soil. Recently thermal treatment trials by Enviropacific
1075 showed an effective PFAS removal in 20 soil samples, with >99.9% reduction in PFOS (from
1076 172 µg/kg to 0.004 µg/kg) and PFOA (from 2.73 µg/kg to <0.0005 µg/kg)
1077 ([https://www.enviropacific.com.au/wp-content/uploads/2016/09/Enviropacific_Treatment-of-](https://www.enviropacific.com.au/wp-content/uploads/2016/09/Enviropacific_Treatment-of-PFAS.pdf)
1078 [PFAS.pdf](https://www.enviropacific.com.au/wp-content/uploads/2016/09/Enviropacific_Treatment-of-PFAS.pdf)) (SI Figure 6). In addition, Ventia (<https://www.ventia.com/projects>) has established
1079 a joint venture with Suez (<https://www.suez.com.au/en-au>) to construct an efficient soil
1080 decontamination facility in Dandenong South, Victoria, Australia to provide a clean, consistent,
1081 and economical PFAS treatment service. This service has been functional since November
1082 2018 and is situated next to the only dumping site licensed to collect Category B waste (i.e.,
1083 industrial wastes with medium levels of contamination) in Victoria (Ventia, 2018). According
1084 to recent report, Ventia has treated more than 525,000 tonnes of polluted soil (including PFAS-
1085 contaminated soil) using thermal treatment technologies. This figure of 525,000 tonnes
1086 represents more than 72% of all soils that have been thermally treated in Australia.

1087

1088 5.5 Case study 5: Destruction by chemical oxidation

1089 In Canada, a bench-scale treatability trial on the removal of PFOS in AFFF-impacted water
1090 and soil systems was directed, with an emphasis on the application of oxidation processes.
1091 Results of the bench-scale treatability trial revealed that 84-97% PFAS removal occurred in 30
1092 days from soil/tap water systems. The technology involved a cautious balance of free radicals,

1093 rate of reaction, and radical scavengers (Mahinroosta and Senevirathna, 2020). However, this
1094 is considered as a relatively non-economical technique, due to the high cost and greater
1095 quantities of chemical consumption (Australian Defense, 2019). Apart from Environment and
1096 Climate Change Canada (ECCC) and Emergencies Science and Technology Section (ESTS)
1097 of Canada reports, there are only a few reports available and only on the laboratory scale for
1098 removal of PFAS from contaminated water systems via chemical oxidation or advanced
1099 oxidation processes. Recently AECOM (<https://www.aecom.com/au/>) launched a new
1100 electrochemical oxidation technology DE-FLUORO™ for permanent removal of PFAS from
1101 the environment in Adelaide; this trial was based on removal of PFAS from contaminated water
1102 ([https://www.aecom.com/press-releases/aecom-to-launch-pfas-solution-de-fluoro-at-cleanup-
1103 2019-in-adelaide/](https://www.aecom.com/press-releases/aecom-to-launch-pfas-solution-de-fluoro-at-cleanup-2019-in-adelaide/)).

1104

1105 **6. Summary and conclusions**

1106 This review examined mobilization, immobilization, and destruction techniques for the
1107 remediation of soil contaminated with PFAS from various sources. Soil amendments which are
1108 effective in the mobilization of PFAS compounds via desorption and complexation reactions
1109 can be applied to enhance the mobility and bioavailability, and subsequent removal through
1110 plant uptake and soil washing. Anionic surfactants can be effective in the mobilization of PFAS
1111 compounds, and their subsequent removal through soil washing. However, one of the
1112 environmental issues with the application of mobilization techniques is that the mobilized
1113 PFAS compounds are subject to leaching, particularly in the absence of plant uptake and soil
1114 washing, thereby leading to ground-water contamination. Mobilization techniques can be used
1115 for the complete removal of PFAS compounds through abiotic and biotic degradation.
1116 Although most of the PFAS compounds are recalcitrant to undergo biotic degradation, the

1117 potential value of abiotic degradation through thermal and chemical redox reactions for the
1118 removal of PFAS compounds has been demonstrated.

1119 Stabilization and solidification remediation treatment has been found to be an effective
1120 immobilization technology aimed at reducing leaching and bioavailability of PFAS
1121 compounds. However, a major intrinsic issue linked to immobilization techniques is that,
1122 although the PFAS compounds become less mobile and bioavailable, their total mass in soils
1123 remains unaffected. The immobilized PFAS compounds may be solubilized and become
1124 bioavailable with time through breakdown of organic-PFAS compounds complexes. Complete
1125 removal of PFAS compounds from solid media, including soil and biosolid waste, can be
1126 achieved through biotic and abiotic degradation and decomposition. Biodegradation of PFAS
1127 is slow and not readily applicable under field conditions to treat large volumes of contaminated
1128 matrix. However, abiotic degradation involving thermal and chemical oxidation and ball
1129 milling can be applied under field conditions to treat large volumes of contaminated matrix.

1130 Given the present understanding on the mobilization, immobilization, and destruction
1131 techniques with regard to managing the mobility and bioavailability of PFAS compounds, and
1132 subsequent remediation of contaminated soils, we propose the following future research
1133 priorities:

1134 – Remediation technologies are primarily focused on PFOS and PFOA, which are the
1135 major PFAS compounds detected in environmental matrices; however, many other PFAS may
1136 be present in environmental matrices including soil, wastewater, and biosolids, and
1137 technologies that are able to treat PFOS and PFOA effectively may not be appropriate for other
1138 PFAS with different properties.

1139 – More *in situ* field studies are necessary to validate the beneficial effect of a wide range
1140 of mobilizing and immobilizing agents in remediating soil contaminated with PFAS.

1141 – Co-contaminants including hydrocarbons and chlorinated solvents may also be present
1142 in PFAS contaminated soil. Oxidative technologies including *in situ* chemical oxidation that
1143 are often applied to these co-contaminants may transform PFAS compounds into PFOS and
1144 PFOA. Hence field studies are necessary to evaluate the impact of soil amendments on the
1145 mobilization of associated co-contaminants.

1146 – It is necessary to develop methods to demonstrate effectiveness of mobilization and
1147 immobilization techniques to manage the mobility and bioavailability of PFAS. For example,
1148 advanced spectroscopic methods can be applied to examine the *in-situ* long-term stability and
1149 effectiveness of immobilization.

1150 – *In situ* field studies are also necessary to determine bioavailability, phytotoxicity, and
1151 ecoreceptor endpoints to demonstrate risk reduction derived from the application of soil
1152 amendments to manage PFAS.

1153 – It is important to enhance regulatory acceptance of these methods through sound
1154 scientific advances and demonstrations.

1155

1156 **Acknowledgment**

1157 We would like to thank Dr Richard Stewart, Managing Director, RemBind Pty Ltd, Australia
1158 for providing information for the Case Study 3 covering PFAS immobilization. We thank the
1159 Federal State North Rhine-Westphalia, Germany, and the European Fond for Regional
1160 Development (EFRE) 2014-2020 “Investitionen in Wachstum und Beschäftigung” for the
1161 funding of the project: “*In situ Eliminierung von Per- und Polyfluorierten Verbindungen in*
1162 *belasteten Grundwässern unter Einsatz von Adsorptiven Komposit-Nanopartikeln auf Basis*
1163 *Oxidativ-Reaktiver Aktivkohle-Polysaccharid-Lipid-Ferrat-Micellen-Aggregate*
1164 *(PANORAMA)*.” Förderkennzeichen: EFRE-0801149. Aktenzeichen LeitmarktAgentur.
1165 NRW: EU-2-1-016B. Although EPA contributed to this article, the research presented was not

1166 performed by or funded by EPA and was subject to EPA's quality system
1167 requirements. Consequently, the views, interpretations, and conclusions expressed in this
1168 article are solely those of the authors and do not necessarily reflect or represent EPA's views
1169 or policies.

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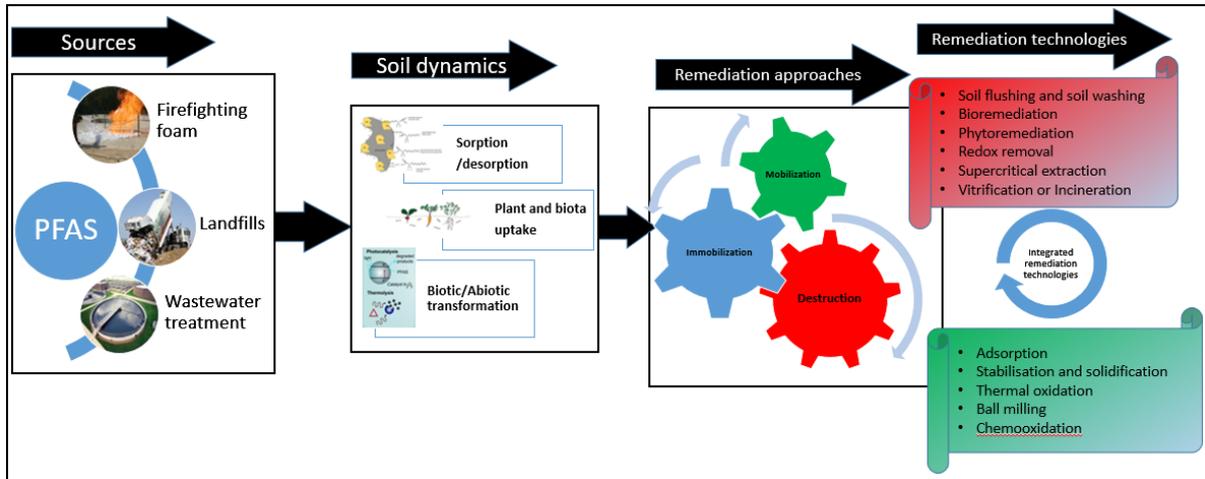
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2031 **Graphical abstract**



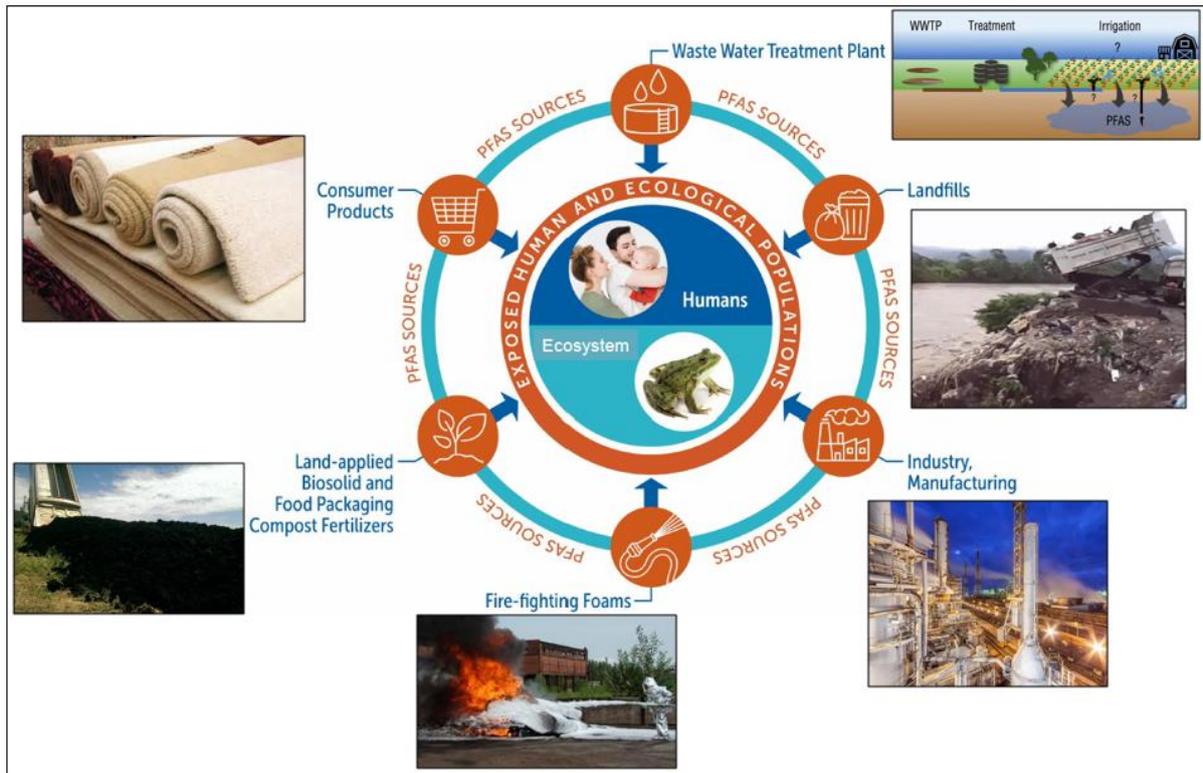
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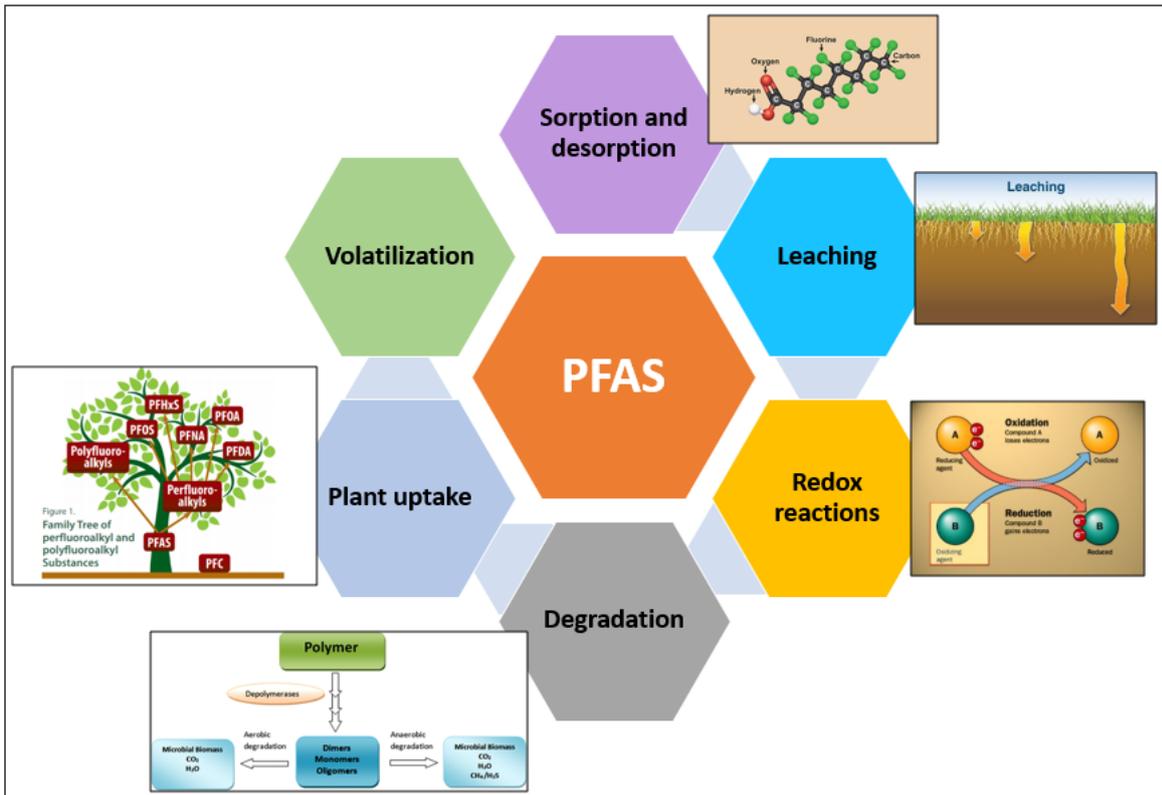
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2038 Figure 1. Schematic diagram on the sources of PFAS compounds

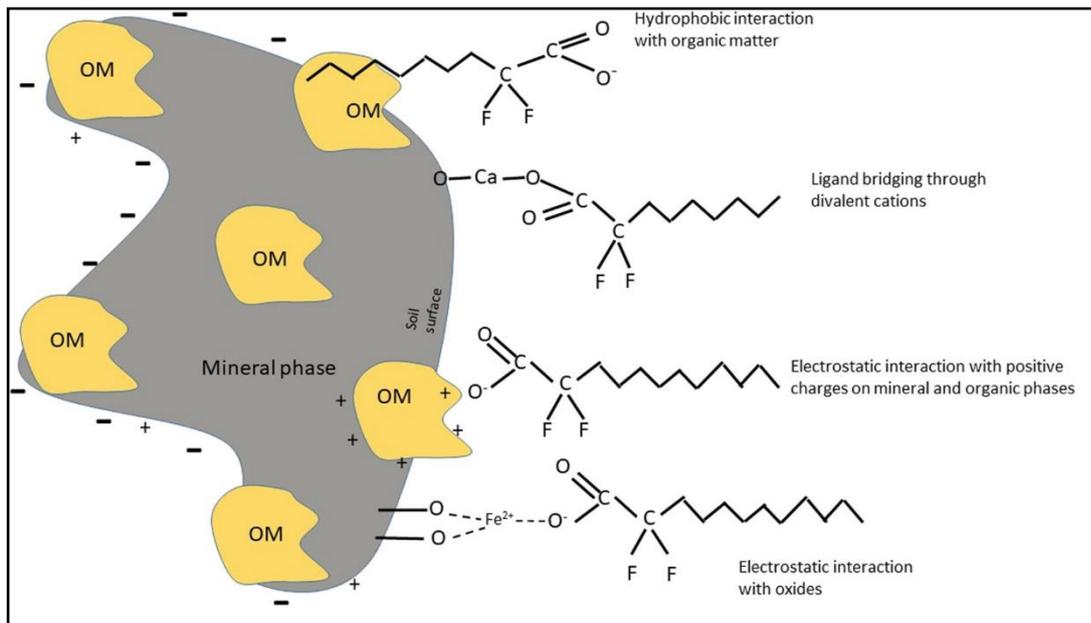
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2041 Figure 2. Schematic diagram on the dynamics of PFAS in soil

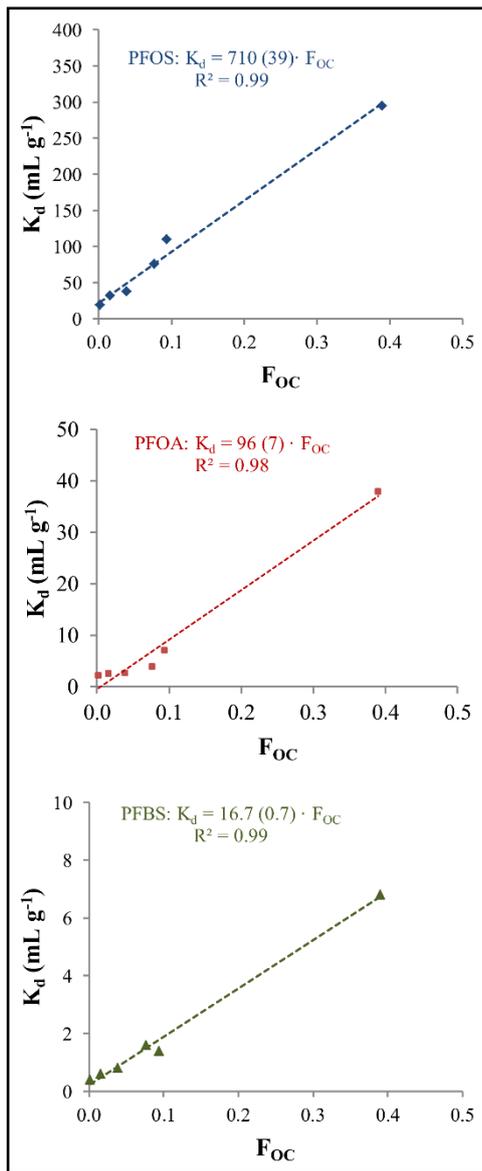
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2044 Figure 3. Adsorption interactions of PFAS in soil (Ross et al., 2018; Li et al., 2018b)

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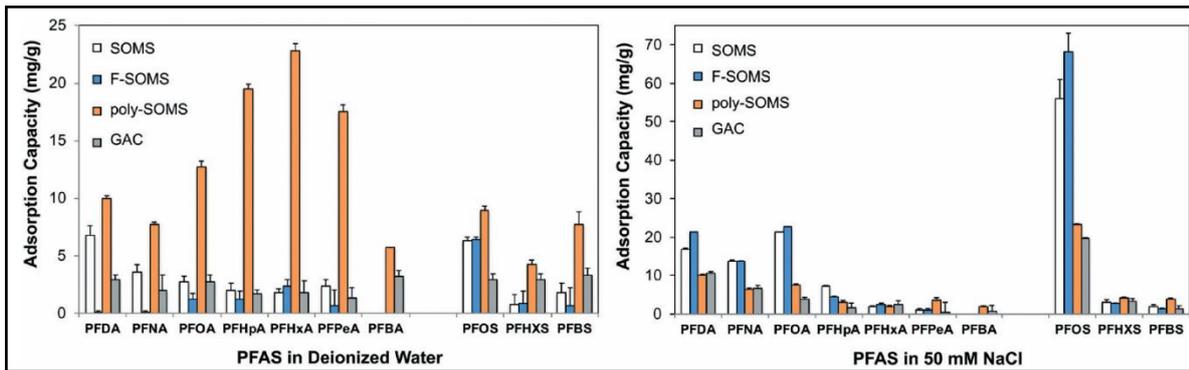


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2047 Figure 4. Relationship between organic matter and PFAS adsorption by soils (Milinovic et

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2058 Table 4. Selected references on the potential value of soil amendments in the immobilization of PFAS compounds in soils

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2062 **Table 1.** Selected references on PFAS contamination in water and wastewater resources.

Source	Country	PFAS content	Reference
Surface and well water from areas associated with application of fluorochemical industry impacted biosolids	Alabama, USA	<p>PFOA ranged from <LOQ (i.e., 10 ng/L) to 11,000 ng/L in well and surface water</p> <p>PFOA ranged from 594 to 2,070 ng/L in drinking water samples</p> <p>PFOS ranged from <LOQ to 151 ng/L in well and surface water</p> <p>PFNA ranged from 12.4 to 286 ng/L in well and surface water</p> <p>PFDA ranged from 54.2 to 838 ng/L in well and surface water</p>	(Lindstrom et al., 2011)
Groundwater, surface water, sewage treatment plant effluents and landfill leachates	Länsstyrelser, Sweden	<p>Drinking water source area - average \sum_{26} PFAS 8.4 ng/L</p> <p>Landfill leachates - average \sum_{26} PFAS 487 ng/L</p> <p>Surface water - average \sum_{26} PFAS 112 ng/L</p> <p>Groundwater - \sum_{26} PFAS 49 ng/L</p> <p>Sewage treatment plant effluents - \sum_{26} PFAS 35 ng/L</p> <p>Background screening lakes - \sum_{26} PFAS 3.4 ng/L</p>	(Ahrens et al., 2016)

Effluent and sludge in wastewater treatment plants	New York, USA	<p>PFOA ranged from 58 to 1050 ng/L in effluent</p> <p>PFOS ranged from 3 to 68 ng/L in effluent</p> <p>PFOA ranged from 18 to 241 ng/g oven dry weight in sludge</p> <p>PFOS ranged from <10 to 65 ng/g oven dry weight in sludge</p>	(Sinclair and Kannan 2006)
Biosolids/ soil mixture exposed to ambient outdoor conditions	Maryland, USA	<p>PFOA showed 24.1 ng/g dry weight</p> <p>PFUnDA showed 18.4 ng/g dry weight</p> <p>PFDA showed 17.4 ng/g dry weight</p> <p>Increased PFDA, PFDoDA and PFOSA over time from unidentified precursors</p>	(Venkatesan and Halden 2014)
Biosolids	Columbia and 32 Sates, USA	<p>PFOS showed 403 ng/g dry weight in biosolids</p> <p>PFOA showed 34 ng/g dry weight in biosolids</p> <p>PFDA showed 26 ng/g dry weight in biosolids</p> <p>Mean load of ΣPFAS in biosolids estimated 2,749–3,450 kg/year</p>	(Venkatesan and Halden 2013)
Water, suspended particulate matter and sediment	Tokyo, Japan	<p>ΣPFAS ranged from 16.7 to 42.3 ng/L in water column</p> <p>ΣPFAS ranged from 6.4 to 15.1 ng/g dry weight in suspended particulate matter</p>	(Ahrens, Taniyasu et al. 2010)

		Σ PFAS ranged from 0.29 to 0.36 ng/g dry weight in surface sediment	
Direct sources - Landfill leachate, water draining from a nearby military base/urban area	Central part, The Netherland	Groundwater within landfill leachate plume- L-PFOA showed 1.8 μ g/L PFBA showed 1.2 μ g/L Σ PFAA showed 4.4 μ g/L Maximum concentration of Σ PFAA in groundwater originating military base showed ~17 ng/L	(Eschauzier, Raat et al. 2013)
Indirect source – Infiltrated rainwater		Maximum concentrations of L-PFOA and PFBA in groundwater halfway the landfill and public supply well field showed 29 and 160 ng/L, respectively L-PFOA and PFBA showed 0.96 and 3.5 ng/L, respectively in groundwater pumping wells	
Raw and treated landfill leachate	China	Σ PFAA ranged from 7280 to 292,000 ng/L in raw leachate Σ PFAA ranged from 98.4 to 282,000 ng/L in treated leachate	(Yan, Cousins et al. 2015)

Surface sediment and sediment core samples	Lake Ontario, Canada	<p>PFOS ranged from 0.684 to 51.8 ng/g dry weight in sediment samples</p> <p>PFDA ranged from 0.044 to 4.06 ng/g dry weight in sediment samples</p> <p>PFOS ranged from 0.492 to 30.1 ng/g dry weight in sediment core samples</p> <p>PFDA ranged from 0.024 to 1.676 ng/g dry weight in sediment core samples</p>	(Yeung, De Silva et al. 2013)
Landfill leachates	USA	<p>PFAA precursors (i.e., PFOSA and FTCA) ranged from ~4 to 36 $\mu\text{g/L}$ ΣPFAS</p> <p>ΣPFAS estimated to leave from 8.5 to 25 kg/yr the landfill via leachate for subsequent treatment at a wastewater treatment plant</p>	(Benskin, Li et al. 2012)
WWTP derived sludge-applied soils	Alabama, USA	<p>PFDA showed ≤ 990 ng/g</p> <p>PFDDA showed ≤ 530 ng/g</p> <p>PFOA showed ≤ 320 ng/g</p> <p>PFOS showed ≤ 410 ng/g</p>	(Washington, Yoo et al. 2010)

Biosolids amended soils	Beijing, China	\sum_9 PFCA ranged from 18 to 113 ng/g dry weight \sum_3 PFSA ranged from 23.4 to 107 ng/g dry weight \sum PFAA ranged from 41.4 to 220 ng/g dry weight	(Wen, Li et al. 2014)
Soil improver produced by industrial waste	Sauerland, Germany	PFOA showed > 0.5 μ g/L in drinking water \sum PFOS+PFOA showed <160 μ g/L in creek water	(Wilhelm, Kraft et al. 2008)
Landfill leachate, biosolids	Australia	PFHxA ranged from 12 to 5,700 ng/L in landfill leachate Decabromodiphenyl ether (BDE-209) ranged from <0.4 to 2300 ng/g in biosolids PFOS ranged from <LOD to 380 ng/g in biosolids	(Gallen, Drage et al. 2016)
Primary sludge, waste biological sludge, treated biosolids	Canada	PBDE ranged from 230 to 82,000 ng/g in primary sludge PBDE ranged from 530 to 8,800 ng/g in waste biological sludge PBDE ranged from 420 to 6,000 ng/g in treated biosolids	(Kim, Guerra et al. 2013)
Lime stabilised biosolids	Mid-Atlantic region, USA	PFNA showed 25.1 ng/g dry weight in sewage sludge PFOA 23.5 ng/g dry weight in sewage sludge PFOS 22.5 ng/g dry weight in sewage sludge	(Armstrong, Lozano et al. 2016)

Sewage sludge	Shanghai, China	Σ PFAs ranged from 126 to 809 ng/g dry weight PFOA ranged from 23.2 to 298 ng/g dry weight	(Yan, Zhang et al. 2012)
Sludge	Nigeria	PFCA ranged from 10 to 597 pg/g in sludge PFSA ranged from 14 to 540 pg/g in sludge PFOS showed 539.6 pg/g in hospital sewage sludge	(Sindik, Orata et al. 2013)
Wastewater, sludge	Korea	PFOS ranged from 3.3 to 54.1 ng/g in sludge PFOA ranged from 2.3 to 615 ng/L in wastewater PFOA ranged from 3.4 to 591 ng/L in influent and effluent wastewater	(Guo, Sim et al. 2010)
Influent, effluent, sludge	Spain	PFHxA showed 5.60 μ g/L in water L-PFBS showed 0.31 μ g/L L-PFOS showed 1.79 μ g/g dry weight in sludge PFBA showed 1.88 μ g/g dry weight in sludge	(Campo, Masia et al. 2014)
Biosolids	Canada	diPAP ranged from 4 to 83 ng/g dry weight PFCA ranged from 0.1 to 19 ng/g dry weight	(Lee, Tevlin et al., 2014)

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2065 **Table 2.** Selected references on the sources of PFAS compounds in soil.

Sources	Country	Observations	Reference
Fluoropolymer manufacturing facility	USA	Both atmospheric deposition and groundwater recharge have contributed to the sources of the site contamination	(Zhu and Kannan 2019)
Sulfluramid use in agriculture	Brazil	Soil contained \sum PFAS concentrations of up to 5,400 pg/g with PFAS profiles generally dominated by PFOS and FOSA	(Nascimento et al. 2018)
AFFF	Australia	PFOS and PFHxS were the most dominant PFAA in all soil samples, with concentrations of PFOS reaching 13,400 ng/g	(Bräunig et al. 2019)
Mega-fluorochemical industrial park	China	Hotspots of contamination by PFAA were found near the mega-fluorochemical industrial park with the maximum concentrations of 641 ng/g in agricultural soil	(Liu et al. 2017)
PFSA manufacturing facility	China	total concentrations of PFAA ranged from 1.30 to 913 ng/g on a dry weight basis	(Gao et al. 2019)

AFFF	Sweden	PFOS and PFOA concentrations in the soil were ranging from 2.18 to 8520 ng/g and from <0.12 to 287 ng/g dry weight, respectively	(Filipovic et al. 2015)
PFAS-production facility	USA	12.2 ng PFOS/g dry weight and 8.0 ng PFOA/g dry weight	(Xiao et al. 2015)
3 M fluorochemical plant	Belgium	The mean concentrations of PFOA and PFOS in soil were 0.89 and 6.77 ng/g respectively	(Groffen et al.2019)
Sludges generated from WWTP	USA	Highest concentrations were PFDA (≤ 990 ng/g), PFDDA (≤ 530 ng/g), PFOA (≤ 320 ng/g), and PFOS (≤ 410 ng/g)	(Washington et al.2010)
AFFF	USA	PFOS was the PFAS species detected at the highest concentration on nearly every soil (median 2400 $\mu\text{g}/\text{kg}$) and aquifer solid (median 270 $\mu\text{g}/\text{kg}$) sample	(Houtz et al.2013)
Biosolids generated from WWTP	Canada	Biosolid-amended soil exhibited increased concentrations of PFCA (0.1–19 ng/g dw)	(Lee et al.2013)
AFFF	Norway	PFOS accounted for 96% of the total PFAS concentration in the soil with concentrations ranging from <0.3 $\mu\text{g}/\text{kg}$ to 6,500 $\mu\text{g}/\text{kg}$	(Høisæter et al.2019)

AFFF	Australia	PFOS is the dominant PFAS measured, with concentration varying from 10 to 200 µg/g	(Baduel et al.2015)
AFFF-impacted fire fighter training area	USA	PFOS surface soil levels reached 36,000 µg/kg around the burn pit	(McGuire et al.2014)
Biosolids	Spain	Biosolid amendment increased concentrations 1.5–14-fold for PFAS	(Navarro et al.2016)
Wastewater	Kampala, Uganda	∑PFAS ranged from 1,700 to 7,900 pg/g dry weight in soil	(Dalahmeh, Tirgani et al., 2018)
Firefighting foams	France	Median value for ∑PFAS showed 12,112 ng/g in area #2 Median value for ∑PFAS showed 8701 ng/g in area #6 Mean value for ∑PFAS ranged from 1110 to 2237 ng/g and the maximum concentration observed <4300 ng/g in area #3 and #4	(Dauchy, Boiteux et al., 2019)
Biosolids	Australia	Annual load of PFOA in agricultural soils estimated 2.2 kg Annual load of PFOS in agricultural soils estimated 13 kg	(Gallen, Drage et al. 2016)
Biosolids	Columbia and 32 Sates, USA	Mean load of ∑PFAS 1,375–2,070 kg applied to agricultural land Estimated PFOA of 85.8–129 kg/year applied to agricultural land	(Venkatesan and Halden 2013)

		Estimated PFOS of 1,026–1,545 kg/year applied to agricultural land	
Biosolids/ soil mixture exposed to ambient outdoor conditions	Maryland, USA	<p>PFOA showed 24.1 ng/g dry weight</p> <p>PFUnDA showed 18.4 ng/g dry weight</p> <p>PFDA showed 17.4 ng/g dry weight</p> <p>Increased PFDA, PFDoDA and PFOSA over time from unidentified precursors</p>	(Venkatesan and Halden 2014)
WWTP derived sludge-applied soils	Alabama, USA	<p>PFDA showed ≤ 990 ng/g</p> <p>PFDDA showed ≤ 530 ng/g</p> <p>PFOA showed ≤ 320 ng/g</p> <p>PFOS showed ≤ 410 ng/g</p>	(Washington, Yoo et al. 2010)
Biosolids amended soils	Beijing, China	<p>\sum_9PFCA ranged from 18 to 113 ng/g dry weight</p> <p>\sum_3PFSA ranged from 23.4 to 107 ng/g dry weight</p> <p>\sumPFAA ranged from 41.4 to 220 ng/g dry weight</p> <p>\sumPFAA in wheat root, straw, husk and grain ranged from 140 to 472, 36.2 to 178, 6.15 to 37.8, and 7.32 to 35.6 ng/g, respectively</p>	(Wen, Li et al. 2014)

Sewage sludge	Shanghai, China	Predicted PFOA in agricultural land was 1.08 ng/g dry weight Predicted PFOA in agricultural land was 7.53 ng/g dry weight	(Yan, Zhang et al. 2012)
Biosolids	Chicago, USA	PFOS ranged from 2 to 483 ng/g in biosolids amended soil	(Sepulvado, Blaine et al. 2011)
Biosolids	Canada	Total diPAP showed up to 300 ng/ g dry weight in soil Total PFCA showed up to 50 ng/g dry weight in soil	Lee, Tevlin et al., 2013
Biosolids	China	PFOS ranged from 1.44 to 43.2 ng/g PFOA ranged from 1.21 to 28.5 ng/g	(Wen, Zhang et al. 2015)

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2068 **Table 3.** Selected references on the potential value of soil amendments in the mobilization of PFAS compounds in soils.

Amendments	PFAS compounds	Observations	Reference
0.01 mol/L CaCl ₂ and 0.03 g of NaN ₃	PFOA, PFBS and PFOS	Desorption yields of PFOA, PFBS and PFOS were 15-19%, 18-27% and <4%, respectively	Milinovic et al. (2016)
Acetic acid	PFCAs and PFSA	Desorption experiments indicated desorption became difficult as the chain length increased, and PFSA were harder to be desorbed than the corresponding PFCAs	Zhao et al. (2012)
Rainwater	PFOA and PFOS	Of the 360 g of PFOA and 367.5 g of PFOS applied to the soil, loss from the soil plot through leachate amounted to 3.12% for PFOA and 0.013% for PFOS. Short-chain PFASs and PFOA pass through the soil much more quickly than PFOS	Stahl et al. (2013)
Water	PFCs	The short chain PFC could pass through the soil without retention and were likely to be carried away easily with surface runoff	Gillrich et al. (2012)

Cationic surfactant, CTAB, and an anionic surfactant, SDBS	PFOS	While CTAB remarkably enhanced the sorption of PFOS on the sediment, SDBS increased the desorption of PFOS	Pan et al. (2009)
Oxalate and root exudates	PFOS	Oxalate increased PFOS desorption by 1.43- to 17.14-fold, effects of root exudates were similar to those of oxalate	Tang et al. (2017)
Methanol with ammonium acetate	86 PFASs	Methanol with hydrochloric acid provided excellent recoveries for most cationic and zwitterionic PFASs	Munoz et al. (2018)
100 mM of CaCl ₂	6 PFCs	The desorption was lower than Adsorption, the soil matrix may act as a protective barrier towards extensive groundwater contamination	Enevoldsen and Juhler (2010)
Supercritical carbon dioxide (Sc-CO ₂)	PFOS and PFOA	The extraction efficiencies (with double extractions) were approximately 77%-100% for PFOA and 59%-80% for PFOS	Chen et al. (2012)
Ethanol	PFOS	The regeneration percent of PFOS from spent activated carbon was 84% after 0.5 h and 98% after 23 h using 50% ethanol solution at 45 °C	Deng et al. (2015)

Ethanol and NaCl in methanol solution	PFCAs	50% ethanol at 45 °C and 1% NaCl in 70% methanol solution	Du et al.
		were suitable for the desorption of PFCAs from the bamboo-derived activated carbon and resin IRA67, respectively	(2015)
Ethyl acetate–dimethylformamide and methanol–phosphoric acid	PAEO, PFOS and PFOA	The sequential use of ethyl acetate–dimethylformamide and methanol–phosphoric acid in combination with pressurised liquid extraction resulted in exhaustive extraction of fluorinated anionic and non-ionic surfactants in sewage sludge	Schröder (2003)
NAPL, anionic surfactant sodium decyl sulfate (SDS) and <i>n,n</i> -dimethyldodecylamine <i>n</i> -oxide (AO)	PFAAs	NAPL, SDS and AO led to decrease in the sorption of PFOS at lower PFOS concentrations (1 µg/L)	Guelfo and Higgins (2013)

2070 **Table 4.** Selected references on the potential value of soil amendments in the immobilization of PFAS compounds in soils.

Amendments	PFAS compounds	Observations	Environmental media	Reference
Activated carbon Montmorillonite clay Compost soil	PFOS	Leaching was reduced by 94-9.9% for AC, 29-34% for compost soil and 28-40% for the montmorillonite amended samples	Soil	Hale et al. (2017)
Powdered activated carbon (PAC), Rembind [®] , pulverized zeolite, chitosan, hydrotalcite, bentonite, and calcium chloride	14 PFAS	Standardized leaching tests on S/S-treated soil revealed that leaching of 13 out of 14 target PFASs (excluding perfluorobutane sulfonate (PFBA)) was reduced by, on average, 70% and 94% by adding PAC and Rembind [®]		Söregård et al. (2019a)
Poly(diallyldimethylammonium) chloride, poly-(dimethylamine-co-epichlorohydrin)	PFOS	below the EPA health advisory level of 0.1 nM using poly(diallyldimethylammonium) chloride concentrations as low as 0.16 g/L	Soil	Anderson et al. (2019)

Palygorskite-based material modified with oleylamine	PFOS	a negligible release (water extractable) of PFOS (only 0.5 to 0.6 %) with a year incubating with amendment (10 % w/w)	Soil	Das et al. (2013)
Fine powdered, coal-based, activated carbon (AC)	PFOS, PFOA and PFHxS	4% (w/w) of the AC to contaminated soils resulted in almost complete removal of PFCs from the water phase	Soil	Kupryianchyk et al. (2016)
Modified clay adsorbent	PFOS	Soil treated with Clay adsorbent (10 % w/w) for a year, a negligible release of the contaminant (only 0.5 to 0.6 %)	Soil	Das et al. (2013)
Cationic polymers polydiallyldimethyl ammonium chloride and polyamine	PFBS, PFHxS, PFOS, PFBA, PFOA and PFNA	For all PFAS the retention on the column showed hysteresis where only 1 to 20% of the PFAS was recovered from the column after flushing with 30 pore volumes of simulated groundwater	Soil	Aly et al. (2019)
Colloidal activated Carbon	PFOS, PFOA	all PFASs were below their respective method detection limits in all post-injection samples	groundwater	McGregor, (2018)

Colloidal activated carbon (PlumeStop ®)	14 PFAS	1% (w/w) colloidal AC treatment, PFAS leachability decreased by up to 81% for i.e. PFOA	Soil	Sorengard et al. (2019b)
Laccase	PFOA	PFOA was degraded 24% in water after 36 d, 40% in soil slurry after 140 d	soil slurry	Luo et al. (2018)
Soil organic matter (SOM)	5 PFAS	peat soil with high fraction of organic carbon (foc, 59%), the PFAAs were completely sequestered in the soil	Soil	Zhao et al. (2016)
SOM	PFOS	SOM in soils promoted sorption of PFOS; ferric oxides had the opposite effect	Soil	Qian et al. (2017)
Iron oxides, alumina and SOM	PFOS	Iron oxides, alumina and SOM were positively correlated to sorption KF	Soil	Wei et al. (2017)
Poly(dimethylamine-co-epichlorohydrin), poly-(diallyldimethylammonium)	PFOS	PFOS concentrations can be reduced below the EPA health advisory level of 0.1 nM using	Soil	Anderson et al. (2019)

		poly(diallyldimethylammonium) chloride		
		concentrations as low as 0.16 g/L		
Corn straw biochar	PFOS	The mobility of PFOS was significantly reduced after the addition of 5% biochar	sediments	Guo et al. (2019)
TiO ₂	PFOS	The addition of TiO ₂ NPs increased PFOS sorption by altering the sediment surface properties	sediments	Li et al. (2018)

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