



Enhanced attenuation (EA) to manage PFAS plumes in groundwater

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Abstract

Remediation of per- and polyfluoroalkyl substances (PFAS) in groundwater is particularly challenging because of their unique chemical and fate and transport properties. Many conventional in-situ remediation technologies, commonly applied to address other groundwater contaminants, have proven ineffective for treatment of PFAS. Given their stability, destruction of PFAS in-situ has remained elusive as an in-situ treatment option. Consequently, new approaches to manage PFAS groundwater plumes are of great interest to environmental practitioners. We propose that enhancing PFAS retention can play an important role in reducing PFAS mass flux and providing long-term protection of downgradient groundwater receptors. Enhanced retention of PFAS fits directly into the enhanced attenuation (EA) framework, an established groundwater remediation strategy that was developed in the first decade of the 2000s for other groundwater contaminants. In this paper, we propose eight EA approaches for PFAS in groundwater, including technologies that are currently being implemented at PFAS sites (e.g., injection of particulate carbon amendments), applications of conventional remediation technologies to PFAS sites (e.g., capping to retain PFAS in the vadose zone), and novel, innovative approaches (e.g., intentional food grade LNAPL emplacement to retain PFAS) for enhanced PFAS retention. These EA approaches leverage the properties of PFAS to (i) facilitate sorption to conventional and novel sorbents, (ii) concentrate PFAS at air/water interface via gas sparging, and/or (iii) encourage retention via tidal pumping and PFAS salting out processes. For each of the proposed EA approaches, we describe the methodology or concept and discuss the key processes, potential applications, anticipated increases in PFAS retention compared to natural systems, potential challenges, alternate designs, and current likelihood of large-scale adoption.

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1 | INTRODUCTION

Remediation of polyfluoroalkyl substances (PFAS)-impacted soils and groundwater poses a number of challenges, including PFAS being released in the environment as complex mixtures, the relative persistence of some PFAS, and evolving stringent regulatory criteria. PFAS as a chemical class include thousands of different chemical compounds that can have varying physical, chemical, and fate and transport processes. Figure 1 provides a high-level summary of classes, nomenclature, charge, and degradation potential. This figure highlights several key concepts: (1) perfluoroalkyl acids (PFAAs) have not yet been shown to degrade in groundwater under natural conditions and (2) some precursors ("PFAA precursors") can transform into PFAAs but have not yet been shown to completely degrade. Additionally, anionic PFAS are generally more mobile than cationic or zwitterionic PFAS and only a few key PFAAs (e.g., PFOS, PFOA) are the current focus of the regulatory community. Note that these are observed characteristics as of 2022 and may change as the PFAS science and remediation field progresses.

Importantly, currently demonstrated in-situ remediation technologies are not known to destroy PFAAs. As a result, at present, the most common way to manage PFAS groundwater plumes is the oldest groundwater remediation technology—groundwater pumping and ex-situ treatment. A newer approach that is being employed at some sites is the injection of particulate sorbents directly into PFAS plumes to form barriers to retain the PFAS in-situ for an indefinite time. While there are several other in-situ remediation technologies in development, some of which attempt to destroy PFAS in-situ, it is still unknown which of these technologies may emerge as practical, proven methods that could be rapidly adopted by the groundwater community.

Based on the PFAS Experts Symposium II meeting, Newell et al. (2022a) outlined three broad scenarios for how remediation of PFAS plumes may evolve over the next several years:

1. Under Scenario 1, the most common groundwater remediation technology, pump and treat, and an emerging technology, in-situ injection or emplacement of sorbents, could be applied at every PFAS groundwater site where some form of plume control is required.
2. Under Scenario 2, the current investment in PFAS research yields one or more "silver bullet" remediation technologies that destroy PFAS in-situ and are widely adopted, similar to the rapid adoption of chemical oxidation, chemical reduction, biodegradation, and thermal remediation at chlorinated solvent sites in the early 2000s.
3. Under Scenario 3, a triage approach is applied, whereby (i) very large sites are managed with point-of-use treatment, (ii) pump and treat, and sorbent technologies are improved and used at some fraction of sites, and (iii) some type of retention-based monitored natural attenuation (MNA) is employed at sites where no receptors are present in the immediate proximity of the current and future plume footprint.

Overall, Scenario 3 was suggested to be the most likely path forward for groundwater remediation in Newell et al. (2022a). While some PFAS sites may be managed with natural retention processes alone, other sites may require near-term, higher energy response measures to protect potential receptors. Therefore, developing improved methods to enhance natural processes that contribute to retention-based MNA is a critical need. The objective of this paper is to identify and describe several options for enhancing these

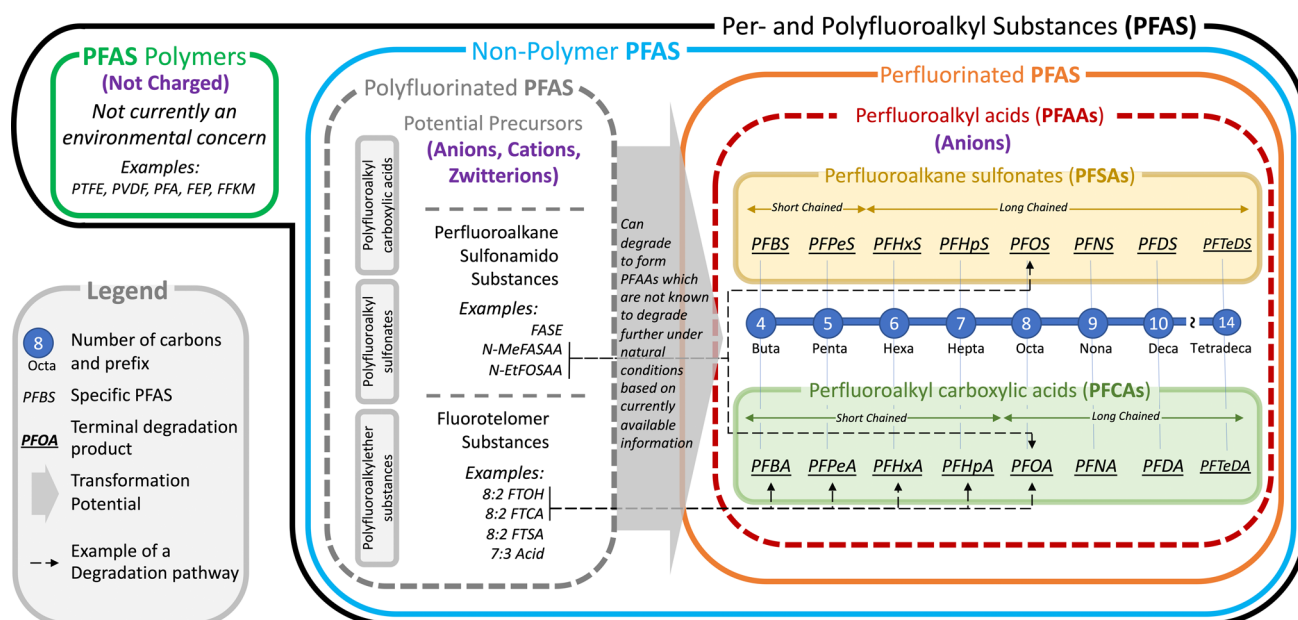


FIGURE 1 PFAS classes with examples of individual PFAS. PFAS, per- and polyfluoroalkyl substances

processes that could be used as the basis for site-specific PFAS remediation strategies.

2 | ENHANCED ATTENUATION (EA)

Although some PFAS sites may be able to rely on natural retention to help manage a PFAS plume, other sites may benefit from interventions that enhance MNA retention based processes. The Interstate Technology and Regulatory Council (ITRC, 2008) defined EA as:

“Any type of intervention that might be implemented in a source-plume system to increase the magnitude of attenuation by natural processes beyond that which occurs without intervention. Enhanced attenuation is the result of applying an enhancement that sustainably manipulates a natural attenuation process, leading to an increased reduction in mass flux of contaminants.”

This guidance was developed to provide a bridge between active source remediation and MNA at chlorinated solvent sites, with the goal of reducing mass flux via potential enhancements in either the source zone, the plume, or plume discharge to surface water areas. The guidance suggested that the source zone provides the greatest number of options for enhancements at chlorinated solvent sites.

For chlorinated solvent sites, Truex et al. (2006) identified two basic categories of EA approaches:

- *Reduced source loading* by hydraulic manipulation, diversion of surface water or groundwater, and injection of sustainable enhancements, such as long-lasting materials that serve as diffusion barriers or promote the degradation of the source.
- *Increased attenuation capacity* by biological or abiotic enhancements that are long-lasting, such as long-lived permeable reactive or sorbent barriers and phytoremediation. These enhancements can be within the plume itself or at plume discharge points to surface water.

For inorganic sites (including certain heavy metals and radionuclides), Truex et al. (2006) identified the same two basic categories of EA approaches but with some differences compared to chlorinated solvents:

- *Reduced source loading* including hydraulic manipulation, reactive barriers, geochemical manipulation, and bioremediation.
- *Increased attenuation capacity* including hydraulic manipulation, reactive barriers, and phytoremediation.

The EA concept has been incorporated into a qualitative framework (Figure 2) where two key factors (the mass discharge (ITRC, 2010) in units of mass per time of a PFAS plume and the

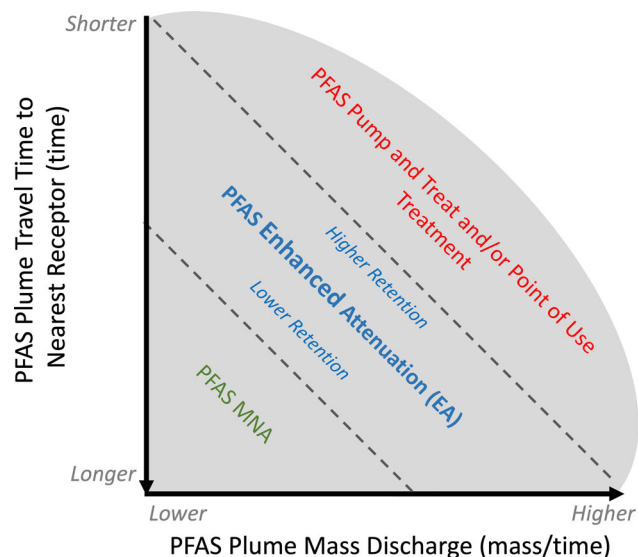


FIGURE 2 Qualitative framework for application of PFAS MNA, EA, enhanced attenuation; PFAS, per- and polyfluoroalkyl substances

travel time to the nearest receptors) are used to define the site scenarios where EA may be most beneficial. The general roles for application of PFAS MNA versus more intensive treatment approaches such as groundwater pump and treat and/or point of use treatment were discussed by Simon et al. (2019). The relative areas of the three zones in Figure 2 are conceptual, and do not reflect estimates of the number of PFAS plumes that might be managed by the three approaches.

3 | GENERAL BENEFITS AND LIMITATIONS OF ENHANCED RETENTION

There are a number of retention processes that are relevant for PFAS in the subsurface, some of which are similar to those for other groundwater contaminants (e.g., TCE, benzene) while others are distinctly different. Newell et al. (2021a) summarized PFAS retention processes that provide a scientific basis for PFAS MNA (Figure 2). A key concept is that all of the processes below are *retention* processes rather than permanent *sequestration* processes, based upon the following definitions:

- *Sequestration*: The permanent trapping and isolation of a chemical in the environment in a natural or artificial storage compartment, such that the chemical does not impact potential receptors; and
- *Retention*: The storage of a chemical in the environment so that the chemical is isolated from potential receptors for a certain time period.

At this time, the PFAAs in a PFAS groundwater plume have not been shown to degrade in groundwater under natural conditions; consequently, there is an expectation that more PFAA plumes will be

expanding as opposed to being stable or shrinking compared to contaminants that are known to be readily biodegradable in groundwater (e.g., benzene). In addition, the retention processes described above are generally reversible, whereby the PFAS are not permanently sequestered but are retained and move much more slowly compared to PFAS that are migrating in transmissive units. Therefore, the primary benefit of these retention processes is to slow the migration of PFAS plumes and reduce the mass discharge of PFAS at potential receptor locations or points of compliance. Some PFAS retention processes may also serve as subsurface *storage reservoirs* that can provide significant benefits by blunting short-term, high mass discharge plumes and converting them to long-term but low mass discharge plumes.

For PFAS, the key retention processes that have the potential to impact fate and transport in the subsurface include air/water interfacial sorption, hydrophobic interactions, NAPL/water sorption, electrostatic interactions, "salting out," and matrix diffusion (Costanza et al., 2019; Higgins & Luthy, 2006; Guelfo et al., 2021; Newell et al., 2021a; Sharifan et al., 2021; Van Glubt & Brusseau, 2021). These concepts were used to develop some potential guidelines on how retention based MNA could be applied at actual PFAS field sites (Newell et al., 2021b) and will be further developed as part of a Department of Defense-sponsored (DoD) project on PFAS MNA (ESTCP, 2021a).

Overall, retention-based MNA processes can provide the following plume management benefits at PFAS groundwater sites even if permanent sequestration is not achieved:

- **Plume Stabilization:** At some sites, retention processes and dispersion may be significant enough to stabilize the PFAS plume and prevent further expansion. If the plume source has been removed or depleted, the PFAS plume may shrink under this condition.

- **Plume Slow-Down:** Retention processes can slow the development and migration of PFAS plumes, extending the time before receptors are impacted. Site managers can then focus on those site-specific conditions, if any, that pose near-term threats, thereby directing resources to the sites requiring more urgent action. This slow-down effect also provides the site manager the time to consider the most cost-effective existing or emerging remediation technology for management of the groundwater impact.
- **Peak Shaving:** For some retention processes, the plume will be subject to "hysteretic" retention processes—wherein desorption occurs more slowly than sorption. At sites where retention capacity is sufficiently large, this fast sorption/slow desorption process decreases the mass discharge of the plume by spreading it out over a longer time period. This phenomenon, also called "Peak Shaving" is illustrated in Figure 3 and, as shown, is analogous to the behavior of a flood control reservoir in that the peak flows are intercepted and spread out over time. For protection of groundwater users, this means that the peak concentrations of PFAS at downgradient well locations could be held at levels below applicable water quality criteria—in much the same way that a reservoir controls downstream flood levels. This same effect could apply to discharge of PFAS from unsaturated soil to underlying groundwater.

Even completely reversible retention processes, wherein the rates of sorption and desorption are equal, may also provide peak shaving benefits, depending on the overall retention capacity at a site, the specific PFAS mass loading pattern over time, and the amount of source attenuation.

However, there are important limitations to retention as an attenuation process for PFAS plumes:

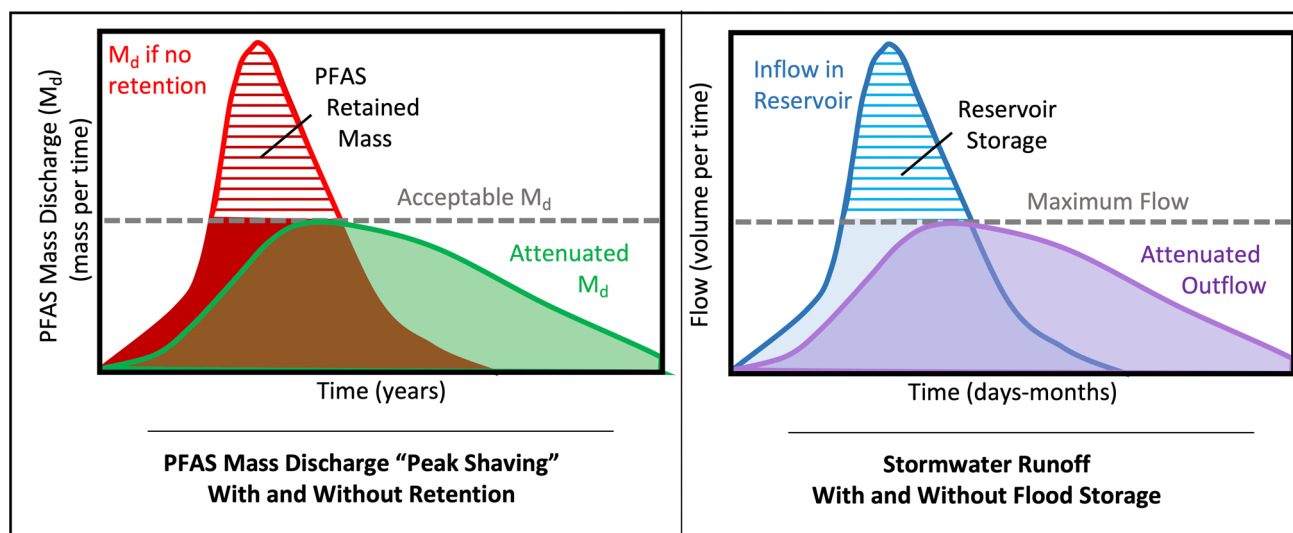


FIGURE 3 PFAS peak shaving with some retention processes versus retention of stormwater in a flood control reservoir. PFAS, per- and polyfluoroalkyl substances

- Retention slows down the plume migration but does not destroy PFAS mass. At this time, PFAA plumes are not known to degrade under natural conditions, and therefore, if the plume source is not diminished over time, some PFAA plumes may continue to expand for relatively long time frames (Farhat et al., 2022).
- Mass retention, when not permanent, may be less favored by some regulatory authorities compared to remediation that entails permanent removal or destruction of the PFAS mass (e.g., see USEPA, 1999a).
- While retention slows the plume migration, if the plume source does not attenuate significantly over time and no other corrective measures are taken, the PFAS concentration at some closed groundwater discharge points (e.g., a lake receiving groundwater flow) will ultimately reach the same level regardless of retention effects, posing the same risk to water users, but at a later time.

4 | EA APPROACHES FOR PFAS PLUMES

This paper describes a suite of EA approaches for PFAS groundwater plumes, ranging from those that are already being implemented at field sites to others that are early-stage concepts. Once subjected to a detailed process review or laboratory experimentation, some of the early-stage ideas discussed herein may be found to be impractical or unfeasible. Nevertheless, this discussion develops a framework for considering the potential applicability of retention-based technologies under different site conditions. In total, we propose eight potential EA approaches for retention-based MNA of PFAS in groundwater. Figure 4 and Table 1 summarize these approaches, which are then each described in more detail with a narrative and conceptual figure.

Where available, an estimated increase in PFAS mass retention per treatment area is compared to the retention under unaltered subsurface conditions (e.g., a relative retardation factor). The increase is based on process knowledge, experimental results from representative research, and other factors (see Supporting Information). Several of the calculations assume retention based on aqueous phase concentration of 1000 ng/L PFOS. A range of 10X was used for some retention values and a “less than” range is used for others to reflect uncertainty. Retention enhancements <10X, 10–100X and >100X were categorized as Low, Moderate and High, respectively.

5 | PFAS EA APPROACH 1: INJECTION OF PARTICULATE SORBENTS TO ENHANCE RETENTION

An emerging approach to enhance natural attenuation of PFAS is injection of particulate carbon amendments (PCAs) into the subsurface to create an in-situ permeable sorption barrier (PSB) or source treatment zone (Figure 5). There are several commercially available PCAs on the market to address a variety of contaminants including (but not limited to): PlumeStop® (Regenesis), Trap & Treat BOS-100® (RPI, Inc.), COGAC™ (Remington), and FluxSorb (Cascade). While each product is different, in general each is designed to: (1) disperse sorbent over some distance from the injection point with little or no impact to groundwater flow (no “clogging”), (2) draw mobile contaminants out of bulk groundwater by serving as a sink via contaminant sorption, and (3) achieve long-term contaminant retention. For example, PlumeStop® (Regenesis) installed via direct push injection into a shallow silty sand aquifer has been reported to reduce PFOA and PFOS concentrations in groundwater from parts per billion levels to below method detection limits (20 ppt for PFOA and 30 ppt for PFOS in 2018) over an 18-month

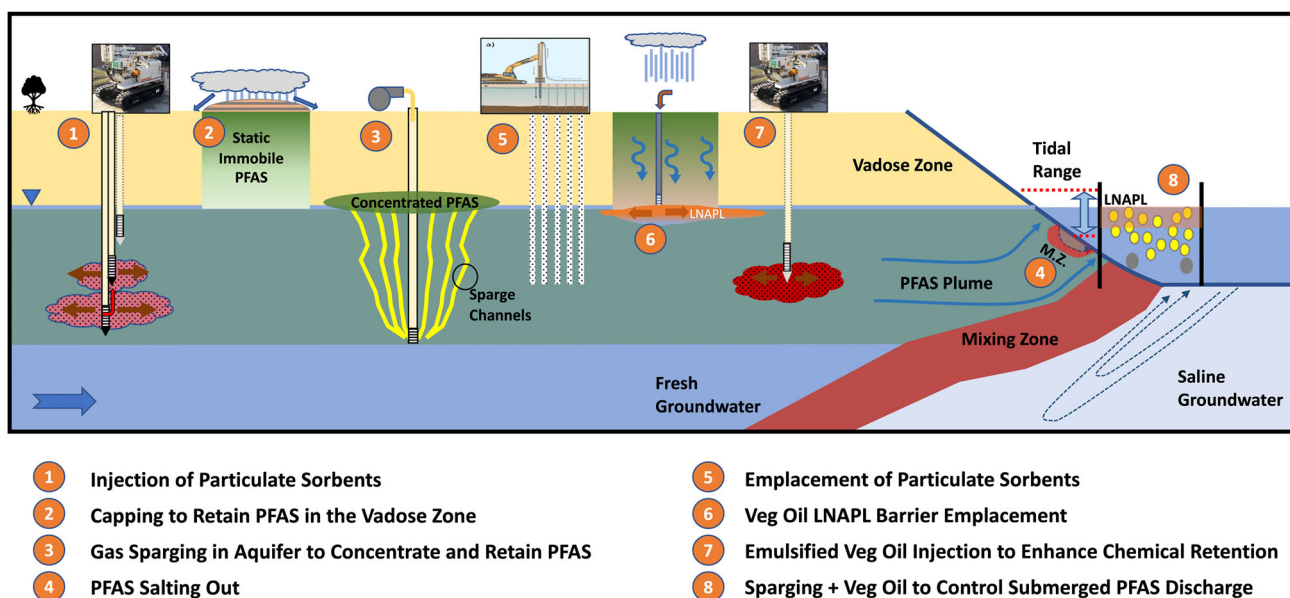


FIGURE 4 Eight potential EA approaches for PFAS plumes. EA, enhanced attenuation; PFAS, per- and polyfluoroalkyl substances

TABLE 1 Summary of eight enhanced attenuation approaches for PFAS Plumes

	1.	2.	3.	4.	5.	6.	7.	8.
Key Concept	Inject particulate sorbents into pore spaces	Stop infiltration and leaching to groundwater	Sparge gas to concentrate and/or retain PFAS around water table	Natural mixing of fresh and saline water increases PFAS sorption	Emplace sorbents into aquifer using geotech equipment	Release neat food-grade LNAPL to form PFAS barrier at water table	Enhances chemical retention (Newell et al., 2021a, 2021b) by slowing or stopping precursor transformation	Construct walls around discharge zone, then sparge gas through water and into oil layer
Location of EA	Primarily PSB in Saturated Zone	Unsaturated Zone	Plume or Source in Saturated Zone	Marine Discharge Points	Primarily PSB or source in Saturated Zone	Horizontal PSB at Source at Water Table	Plume or Source, primarily in Saturated Zone	Discharge Points
Key Processes	Hydrophobic interactions	Restricting infiltration	Interfacial sorption	Salting out	Hydrophobic interactions	NAPL/Water partitioning + Interfacial Sorption	"Chemical Retention"	Air/water + NAPL/Water partitioning
Key Applications	Low to moderate mass flux plumes or sources	Strong vadose zone sources currently leaching PFAS to groundwater	Unconfined aquifers with shallow PFAS plumes with no confining units	Freshwater PFAS plumes in coastal areas with submarine discharge	Flat, open sites less than 80 ft deep	PFAS sources in vadose zone	Precursor sources in aerobic aquifers	Near-shore groundwater discharge points
Example of Increase in Retention in Treatment Area Compared to Natural Systems (see Supporting Info)	High For PFOS 300–3000X for in situ colloidal activated carbon (based on Carey et al., 2019 data) and 1000 ng/L PFOS plume.	High Potentially orders of magnitude reduction in mass flux from PFAS above groundwater fluctuation zone.	Moderate-High For PFOS up to 180X for unsaturated soil, air/water partitioning. (Estimated using data from Guo et al., 2020).	Moderate For PFOS Up to 15X (Hong et al., 2013) (Compared to Freshwater Systems).	High For PFOS 400–4000X for 75% GAC columns with 2-foot spacing and 1000 ng/L PFOS plume.	Low (Saturated Zone) PFOS up to approximately 4X in sat. zone; (Van Glubt & Brusseau, 2021).	Low-Moderate (Chemical Retention) Could reduce PFAA formation from precursors by 6–60X (Butt et al., 2013) (potential additional sorption).	Unknown
Potential Challenges	Short lifetimes for high mass flux? Potential	Does not control saturated	Groundwater fluctuations could reduce retention?	Retention alone may not	Lens effect for closely spaced	Potential for infiltration to	Precursors may come under	Construction may be difficult and

TABLE 1 (Continued)

	1.	2.	3.	4.	5.	6.	7.	8.
	Injection of particulate sorbents to enhance retention	Capping to retain PFAS in the vadose zone	Gas sparging in aquifers to concentrate, retain PFAS	Retention Via PFAS salting out processes	Emplacement of particulate sorbents with geotech equipment	Intentional LNAPL barrier emplacement to retain PFAS	Inject emulsified Veg. Oil (EVO) to enhance chemical retention	Capture of PFAS in groundwater discharging to surface water
	for rerelease over time? Potential to plug aquifer?	sources. Reduction in vertical mass flux difficult to measure. Timeframe for flux reduction uncertain.	directly in heterogeneous aquifers could spread PFAS?	meet stringent marine aquatic criteria. Desorption during high recharge years may be a factor?	columns not yet verified in the field. Potential for rerelease over time? Short lifetimes for high mass flux?	be diverted and not migrate through LNAPL zone?	more stringent regulatory scrutiny and may need to be controlled as well. If EVO is depleted, PFAS released.	process efficiency is unknown.
Potential Alternate Designs	Used for source treatment vs. plume barriers. Use of nonactivated carbon sorbents.	Changing site drainage patterns as opposed to capping to reduce infiltration.	Sparging in trenches (ESTCP, 2021b); Sparging + phytoremediation (Newell et al., 2021d); use of aphrons in situ.	Injection into deep saline aquifers via deep well disposal. Injection of divalent cations (e.g., Ca ²⁺) to increase sorption.	Used for source treatment vs. plume barriers. Use of alternate sorbents such as poly-DADMAC.	Use of thixotropic gels (Riha and Looney, 2015). Use of neat LNAPLs that are amended with suspended sorbents to increase retention.	Use of EVO formulations that do not ferment to CH ₄ , increasing longevity (Borden, 2022).	On-shore application rather than off-shore to intercept PFAS plume.
Development Status as of Early 2022	Applied or planned at 100 PFAS sites (Regenesis, 2021).	As of 2019, used at 12% of PFAS Sites (EBJ, 2019).	Some unintentional cases at PFAS sites (e.g., McGuire et al., 2014).	Likely occurring to some degree at most PFAS plumes close to coasts.	No laboratory or field trials.	No laboratory or field trials.	No intentional cases but may have occurred at EVO CVOC sites.	No laboratory or field trials.
Related R&D Projects	(SERDP, 2021a; ESTCP, 2020a, 2020b)	(ESTCP, 2022a)	(SERDP, 2021b; ESTCP, 2021b)	(SERDP, 2021c) (ESTCP, 2022b) one Navy project	(ESTCP, 2022b)	-	-	-
Likelihood of Large-Scale Adoption	Very High	High	Moderate	Moderate	Moderate-High	Low-Moderate	Low-Moderate	Low

Note: Permeable Sorption Barriers (e.g., Roehl et al., 2001) are also sometimes referred to as permeable reactive barriers and are typically constructed in the plume and/or at a plume discharge point. Abbreviations: EVO, emulsified vegetable oil; PSB, permeable sorption barrier.

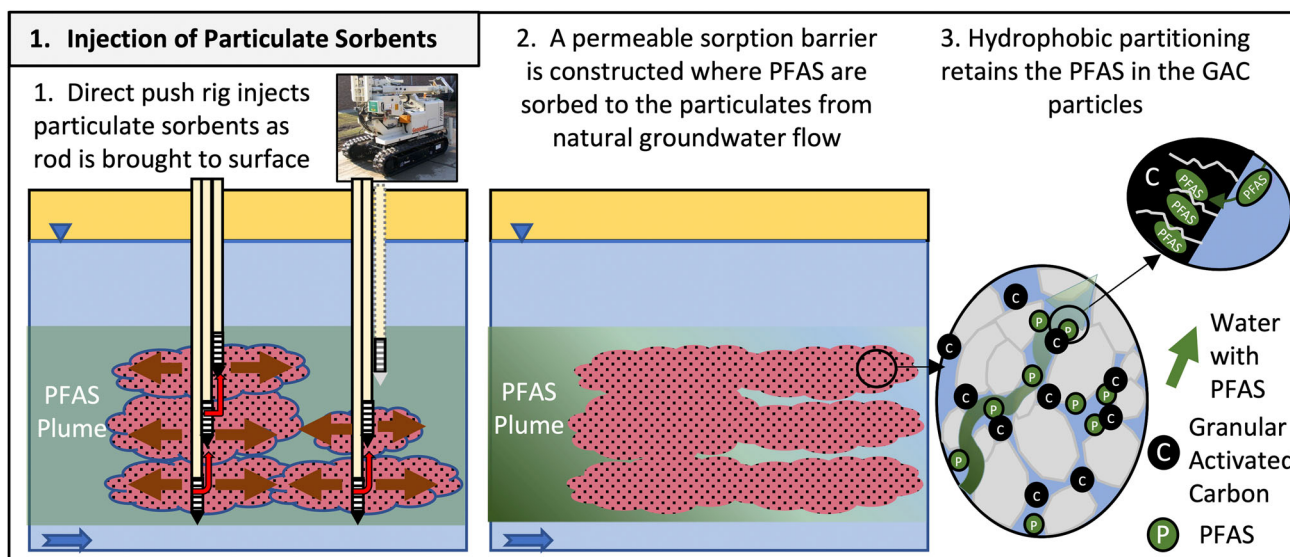


FIGURE 5 Injection of particulate sorbents to enhance attenuation of PFAS plumes. PFAS, per- and polyfluoroalkyl substances

monitoring period (McGregor et al., 2018). McGregor et al. (2018) found that the activated carbon was well distributed within the source zone and was detectable up to 5 m away from the injection point. Furthermore, Carey et al. (2019) used a numerical model to simulate the longevity of colloidal activated carbon injection based on several factors including the fraction of colloidal activated carbon in soil (0.01%–0.1% of the soil mass in the treatment zone). For the various simulated scenarios, breakthrough of the plume (PFOS above a screening value of 0.6 $\mu\text{g/L}$) in a downgradient monitoring well ranged from about approximately 5 years to no breakthrough for at least 100 years (Carey et al., 2019). The use of powdered activated carbon with a coagulant (poly-DADMAC) as a subsurface injected sorbent has also been researched (Liu et al., 2020; SERDP, 2020; Simcik et al., 2019). Polymer stabilized ion-exchange resins have also shown promise as sorbent candidates for subsurface injection in lab-scale column studies (Liu et al., 2022). Overall retention from this approach may be negatively impacted by co-occurring dissolve organic carbon if not taken into consideration for activated carbon dosing.

Under the SERDP program, the DoD funded eight research projects in 2021 to better understand the processes that influence the effectiveness and fate of particulate amendments (SERDP, 2019; SERDP, 2021a; SERDP, 2021d). The specific points of inquiry for these research projects vary, but include sorption/biodegradation processes; behavior of particulate sorbents in fractures; efficacy in arresting downgradient plume migration; mechanisms and processes involved in injected particulate sorption; new methods to evaluate synergistic sorption/degradation effects; use of mineral-based soil amendments; and the long-term performance of these injected barriers in coastal groundwater. In addition, the DoD's ESTCP program has funded research projects evaluating the injection of sorbents to control PFAS, such as methods to validate performance (ESTCP, 2020a) and methods to quantify the spatial delivery of particulates in the subsurface (ESTCP, 2020b).

6 | PFAS EA APPROACH 2: SURFACE CAPPING TO INCREASE RETENTION IN THE VADOSE ZONE

Construction of impermeable covers and caps over PFAS sources in the vadose zone have the potential to greatly reduce mass flux of PFAS to groundwater by (1) reducing PFAS leaching due to infiltration and (2) potentially reducing oxygen influx to suppress the biotransformation PFAS precursors and retain the PFAS in less-mobile precursor forms, compared to key, mobile PFAAs. The surface covers or caps do not change the overall mass of PFAS in the vadose zone, but could help reduce leaching and retain the source in place until other remedial measures and/or institutional controls can be implemented, if needed. Figure 6 shows the conceptual design of this retention measure. A 2019 survey of remediation professionals performed by the Environmental Business Journal indicated that capping was used 12% of time for PFAS remediation projects (EBJ, 2019).

Candidate covers or caps include temporary covers such as plastic sheeting, semi-permanent or permanent covers including evapotranspiration covers, compacted clay caps, asphalt/concrete caps, caps with geosynthetic liners, and engineered RCRA subtitle C Caps (USEPA, 1989; ITRC, 2003). For source areas that will eventually require active remediation, such as treatment or removal, temporary covers could serve as an interim measure if adequate treatment or disposal capacity is unavailable. For other PFAS sources, permanent covers/caps can be used to retain (or potentially sequester) the PFAS in the vadose zone, which is a commonly used approach at many waste sites and landfills. Drainage designs must ensure that any runoff from the cap does not percolate through soils containing significant PFAS concentrations. Computer models such as HELP (Hydrologic Evaluation of Landfill Performance; USEPA, 1984) and PFAS vadose zone transport models (e.g., Guo

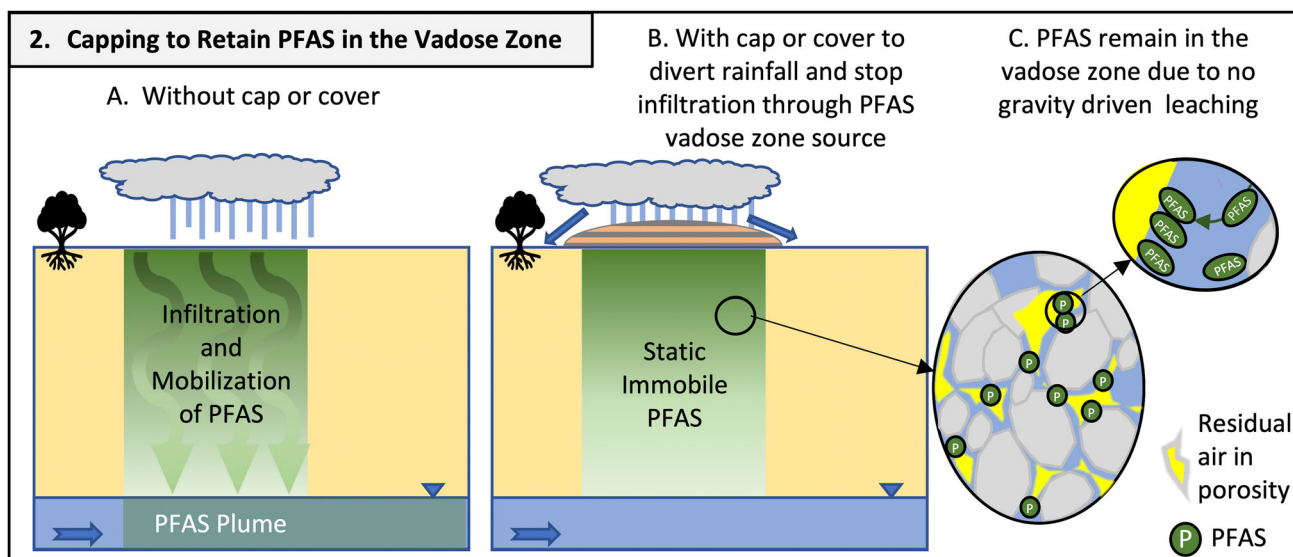


FIGURE 6 Covers/caps to increase the retention of PFAS in the vadose zone. PFAS, per- and polyfluoroalkyl substances

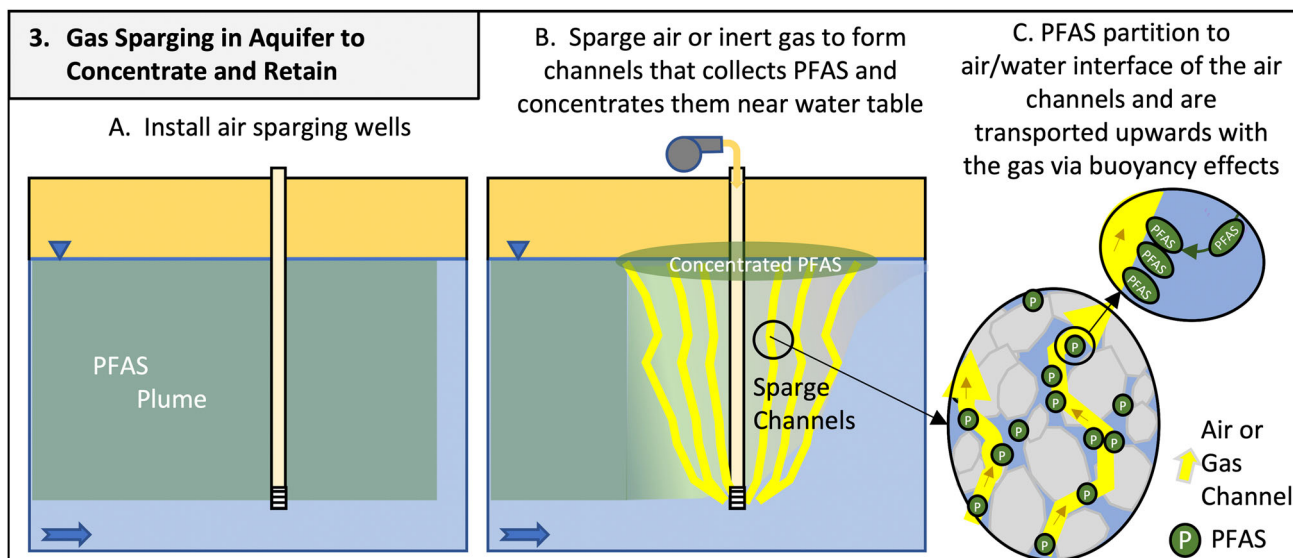


FIGURE 7 Gas sparging directly in aquifers to enhance retention of PFAS. PFAS, per- and polyfluoroalkyl substances

et al., 2020; Silva et al., 2020) could be used to design caps and evaluate the reduction in PFAS mass flux to groundwater.

7 | PFAS EA APPROACH 3: GAS SPARGING IN AQUIFERS TO CONCENTRATE AND RETAIN PFAS

Newell et al. (2020; 2021d) proposed that, for PFAS plumes in unconfined aquifers, gas sparging could be utilized to enhance long term retention of PFAS in the geologic media. Under this process, gas sparging can serve to transport diffuse levels of PFAS in groundwater upward to the capillary fringe, where the PFAS mass is concentrated

and naturally retained due the high propensity of PFAS to adsorb at the air-water interface (Figure 7). In theory, the sparging process reduces the overall mass discharge of PFAS leaving the sparging zone by: (1) concentrating the PFAS in a much thinner aquifer horizon near the water table; (2) increasing the effective retardation factor of the PFAS due to air/water interfacial sorption effects; and (3) reducing the relative permeability and therefore reducing the groundwater flow through the saturated portion of the concentrated PFAS zone as long as sparging is ongoing.

To prevent remobilization of PFAS mass from the capillary retention zone, a semi-permanent system could be operated periodically to reestablish the retention of PFAS that are either leached back into migrating groundwater by recharge events or water table fluctuations.

By increasing oxygen concentrations in groundwater, air sparging has the potential to stimulate the aerobic biodegradation of PFAS precursors to form PFAAs, which are the more mobile forms of PFAS in groundwater. However, the negative effects of this biodegradation transformation process could be mitigated by either (1) sparging with an inert gas such as nitrogen; or (2) sparging in zones with relatively few precursors (e.g., the leading edge of the PFAS plume as explained by Adamson et al., 2020).

Potential alternative designs include sparging with horizontal wells; sparging in trenches (ESTCP, 2021b; Newell et al., 2021d); combining sparging with phytoremediation to remove some of the retained PFAS near the water table (Newell et al., 2021d), or potentially injecting colloidal gas apheres for in-situ remediation of PFAS (Kulkarni et al., 2022a; Newell et al., 2021e) instead of a sparge gas.

8 | PFAS EA APPROACH 4: RETENTION VIA PFAS SALTING OUT

Newell et al. (2021a) observed that the salting-out effects of PFAS may be an important natural attenuation process in environments where fresh groundwater plumes mix with saline groundwater, a condition which occurs in “subterranean estuaries” (Moore, 1999). We have designated this process as EA because of (1) limited recognition of this process and (2) niche circumstances where salting out can be enhanced by increasing the salinity in freshwater plumes containing PFAS. The salting-out process is driven by two separate natural phenomena, one hydrologic and one chemical:

1. *Tidal pumping and wave-driven circulation, and density driven seawater circulation:* This is a natural hydrologic process that can result in additional mixing of fresh groundwater with saline

groundwater compared to estuarine environments (Figure 8) (Robinson et al., 2006; Taniguchi et al., 2019). Variable density groundwater flow models such as SEAWAT (Langevin, date unknown) can be used to simulate this phenomenon.

2. *Salting out:* This is a chemical process wherein the solubility of a dissolved constituent in a solution is decreased (and in some cases, sorption is increased in the presence of solid phases) by increasing the ionic strength of the solution (Chen et al., 2013; Hong et al., 2013; Jeon et al., 2011; Milinovic et al., 2015; Munoz et al., 2017; Wang et al., 2013; Wang et al., 2020; You et al., 2010).

Together, these two processes may result in naturally enhanced retention of PFAS for fresh groundwater plumes that discharge to saline systems (e.g., estuaries, bays, and the open ocean) (Figure 8). The “salting-out” effect for PFAS has been extensively studied in surface water systems, where retention has been observed to increase by over 10X for PFOS in rivers that mix with salt water in estuaries. Hong et al. (2013) investigated the fate and partitioning of 13 PFAAs in river estuaries, and reported that “ K_d values increased exponentially as a function of salinity. Due to the ‘salting-out’ effect, perfluoroalkyl acids (PFAAs) were largely scavenged by adsorption onto suspended sediment and/or sediments in estuarine environments.” Wang et al. (2020) analyzed 24 PFAAs in surface water and sediment at estuarine areas, and the data exhibited “an exponential relationship between salinity and the K_d values of PFBA, PFHxA and PFOS, indicating a ‘salting-out’ effect that promotes the transport of these compounds from water to sediment,” and also a strong positive linear correlation between the K_d values and the chain lengths of PFCAs.

Other key laboratory findings on the retention of PFAS on the solid phase in saturated saline systems include: (1) the organic carbon content in the solid phase was positively correlated to K_d values of PFAS (Chen et al., 2013; Milinovic et al., 2015; Wang et al., 2013;

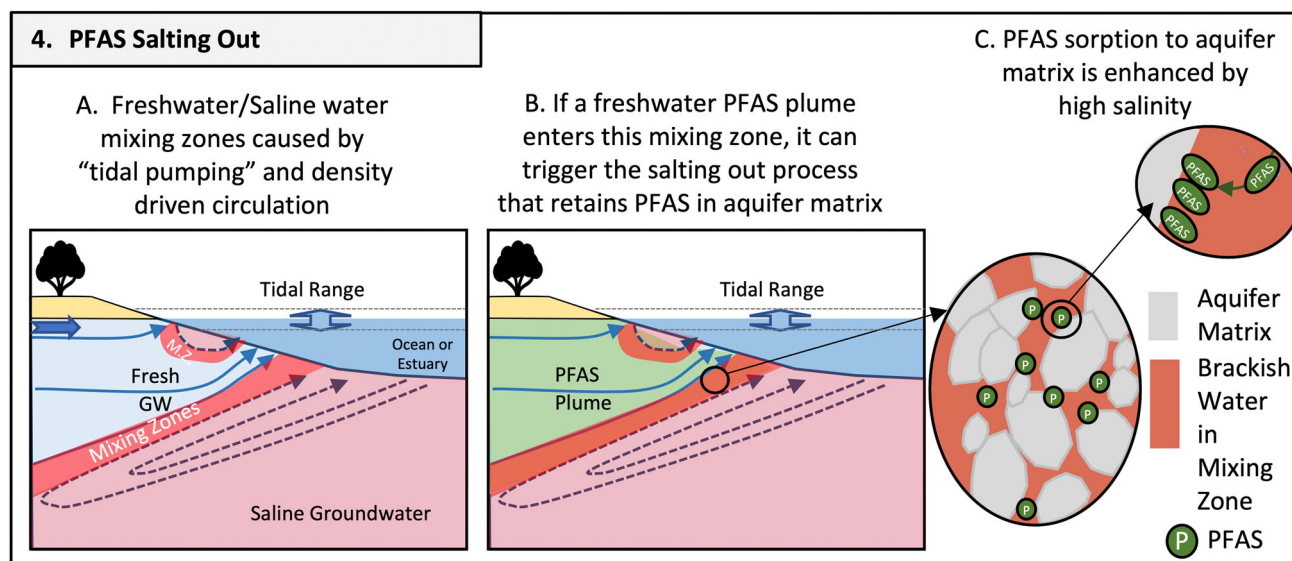


FIGURE 8 Salting out processes that enhance the retention of PFAS. PFAS, per- and polyfluoroalkyl substances

You et al., 2010) and (2) divalent cations exhibited stronger salting-out effects on PFAS than monovalent cations (Chen et al., 2013; Lei et al., 2022; You et al., 2010).

In theory, salting-out processes could enhance retention of PFAS fluids when being injected into deep waste disposal wells. The receiving geologic layers of deep injection wells are typically several thousands of feet deep and saline, hence are not potential sources or threats to drinking water. Therefore, deep well injection might serve as an effective disposal method for PFAS contaminated aqueous matrices. Alternately, building on the work by Higgins and Luthy (2006), Xing et al. (2021), Ebrahimi et al. (2021), and Cai et al. (2022), a potential variant of the salting out approach is to inject aqueous divalent calcium into the subsurface as an engineered salting out process. Finally, it may be possible to engineer increased recirculation of saline groundwater with fresh groundwater above tidal pumping mixing zones to increase the PFAS salting out process near marine shorelines.

9 | PFAS EA APPROACH 5: EMPLACING PARTICULATE SORBENTS USING SPECIALIZED GEOTECHNICAL EQUIPMENT

Specialized geotechnical equipment can be used to emplace sorbents into the subsurface, as an alternative to sorbent injection. The speed and efficiency of these techniques have been successfully exploited to emplace zerovalent iron at a low permeability chlorinated solvent site (ESTCP, 2021c). Recently, a second ESTCP project (ESTCP, 2022b) has been funded to test emplacing sorbents such as activated carbon (or a mixture of activated carbon with poly-DADMAC, as proposed by Liu et al. (2020) as a source remediation and a PSB technology for PFAS sites (Figure 9). Like sorbent injection,

various sorbent formulations including activated carbon and/or ion exchange resins can be emplaced in the subsurface to treat PFAS plumes.

Two types of existing specialized geotechnical equipment can be used to emplace particulate sorbents in the subsurface. The “Grout bomber” (Keller Group) can install hundreds of closely spaced (2- to 4-ft spacing), 3- to 4-inch diameter vertical columns per day, up to 50 ft in depth and filled with sorbent amendments using an in-line grout delivery system. Similarly, the “VibroCat” can emplace sorbents in tens of vertical columns per day with each column having diameter up to 3 ft.

Two potential applications of this technology have been identified, one for source zones and one for permeable sorption barriers. For source zones, a network of these closely spaced sorbent columns can be placed to sorb and immobilize PFAS. Because of the close spacing, removal of PFAS that has diffused into low permeability strata can be greatly accelerated by shortening diffusion lengths and establishing a concentration gradient between the sorbent column and the native material. To control plume migration, a series of high permeability vertical sorbent columns can be placed at downgradient transects in the plume, perpendicular to ground water flow, to act as an in-situ permeable sorption barrier. Because of the column's high permeability relative to that of the geologic formation, a groundwater flow “lens effect” can be established where groundwater streamlines are drawn into and traverse the sorbent columns even though the columns do not span the entire width of the barrier.

During an ESTCP field demonstration in 2018, the Grout Bomber successfully emplaced total of approximately 77,000 lbs of zerovalent iron in 800 closely spaced reaction columns at an average rate of 119 vertical columns per day up to a depth of 30 ft (ESTCP, 2021c; Richardson et al., 2020).

A related technology is to emplace sorbents in permeable sorption barriers using trenching equipment. One ESTCP project is

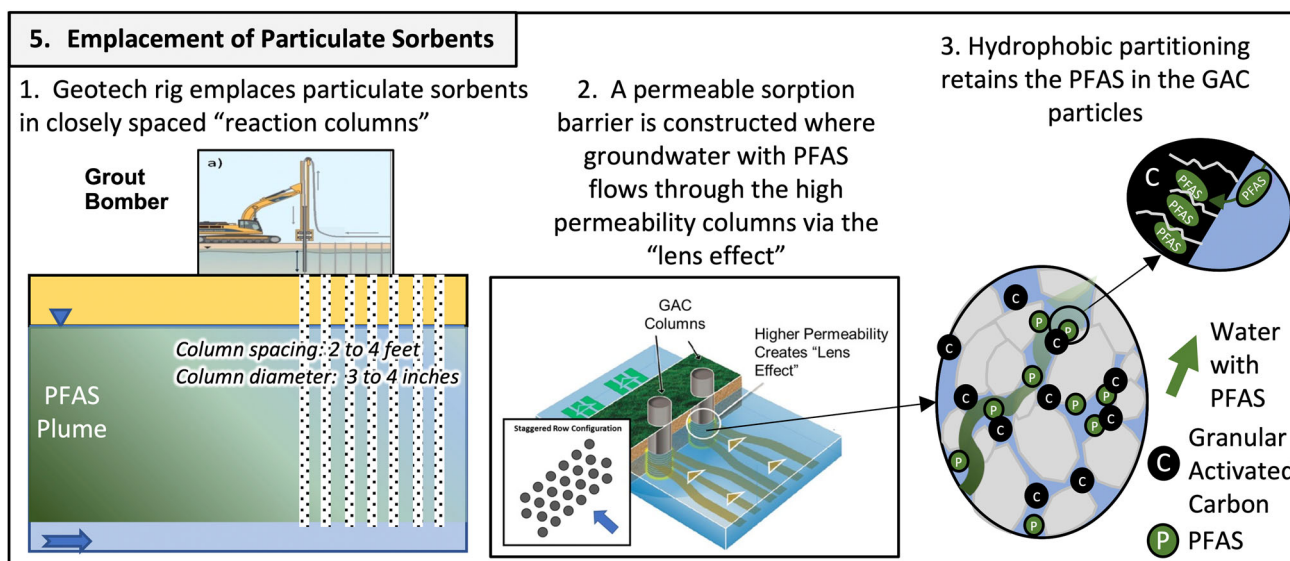


FIGURE 9 Emplacement of solid sorbents with specialized geotechnical equipment to enhance PFAS retention. PFAS, per- and polyfluoroalkyl substances

demonstrating this approach with ion exchange sorbents (ESTCP, 2020c). The use of powered activated carbon with a coagulant (poly-DADMAC) as a subsurface injected sorbent has also been researched (Liu et al., 2020; SERDP, 2020; Simcik et al., 2019).

10 | PFAS EA APPROACH 6: INTENTIONAL FOOD GRADE LNAPL BARRIER EMPLACEMENT TO RETAIN PFAS

PFAS can be significantly retained at the water-NAPL (non-aqueous phase liquid) interface due to the amphiphilic nature of PFAS (Van Glubt & Brusseau, 2021). Of note is that it has been demonstrated that the process of interfacial adsorption drives this PFAS/NAPL retention process (Van Glubt & Brusseau, 2021). Therefore, there can be some degrees of existing enhanced retention at sites that had historical combined releases of petroleum hydrocarbons and aqueous film-forming foam (AFFF) due to PFAS/NAPL interfacial adsorption.

This phenomenon could be used to retain PFAS in the subsurface by constructing engineered food-grade LNAPL (light non-aqueous phase liquid) bodies (e.g., vegetable oil) on or at the water table to intercept PFAS that are being transported downward by percolating water and thereby prevent PFAS from entering the underlying groundwater (Figure 10). The LNAPL emplacement process could be as simple as injecting LNAPL near the water table with a single injection well and letting the LNAPL spread laterally. In such a case, the natural spreading of the LNAPL pool could allow the use of a single injection point instead of a network of multiple injection points, with significant cost savings. While several researchers have suggested that hydrophobic retention of PFAAs (e.g., PFOA and PFOS) may be relatively low at the NAPL-water interface in the saturated zone (retardation factors on the order of 1–4), retention of

precursors may prove to be orders of magnitude greater (Brusseau, 2018; Silva et al., 2019; Van Glubt & Brusseau, 2021).

The Department of Energy has used neat oil injection and spreading as an EA approach to intercept and reduce chlorinated solvent loadings to groundwater at two sites (DOE, 2020; Hooten et al., 2016; Looney, 2022). In addition, LNAPL migration through the unsaturated zone and its spreading at the water table is a well understood phenomenon (CL:AIRE, 2014; Sale et al., 2018) based on empirical observations at thousands of LNAPL sites and the availability of a wide variety of LNAPL release models. Because retention occurs at the NAPL-water interface, interfacial area becomes important. Researchers have suggested that NAPL interfacial area in the vadose zone becomes larger than in the saturated zone due to intermediate phase wetting, wherein NAPL acts as a film between water in the solid media pore spaces and the air phase (Figure 11; Fagerlund, 2006; Sale et al., 2018). Additionally, in the bottom of the LNAPL body, ebullition (bubbling) of methanogenic carbon dioxide/methane bubbles via Natural Source Zone Depletion processes and the resulting PFAS gas-water partitioning could also supplement the direct PFAS retention on the LNAPL-water interfaces (Garg et al., 2017). Finally, the introduction of LNAPL could also make groundwater more anaerobic, thereby slowing the transformation of precursors to PFAAs (see PFAS EA Approach 7).

Key tools to model LNAPL migration include the USEPA's Hydrocarbon Spill Screening Model, a one-dimensional transport model simulates LNAPL transport through the unsaturated zone to the water table (Charbeneau & Weaver, 1995). The UTCHEM model (University of Texas, 2000, 2013); or see summary and video in the Concawe LNAPL Toolbox, Newell et al., 2021c) can be used for more complex sites. These modeling tools could be used to design and intentionally create stable, innocuous neat LNAPL emplacements that then would retain PFAS mobilized by infiltration events. An indirect method for LNAPL-based EA is to forgo near-term active removal of

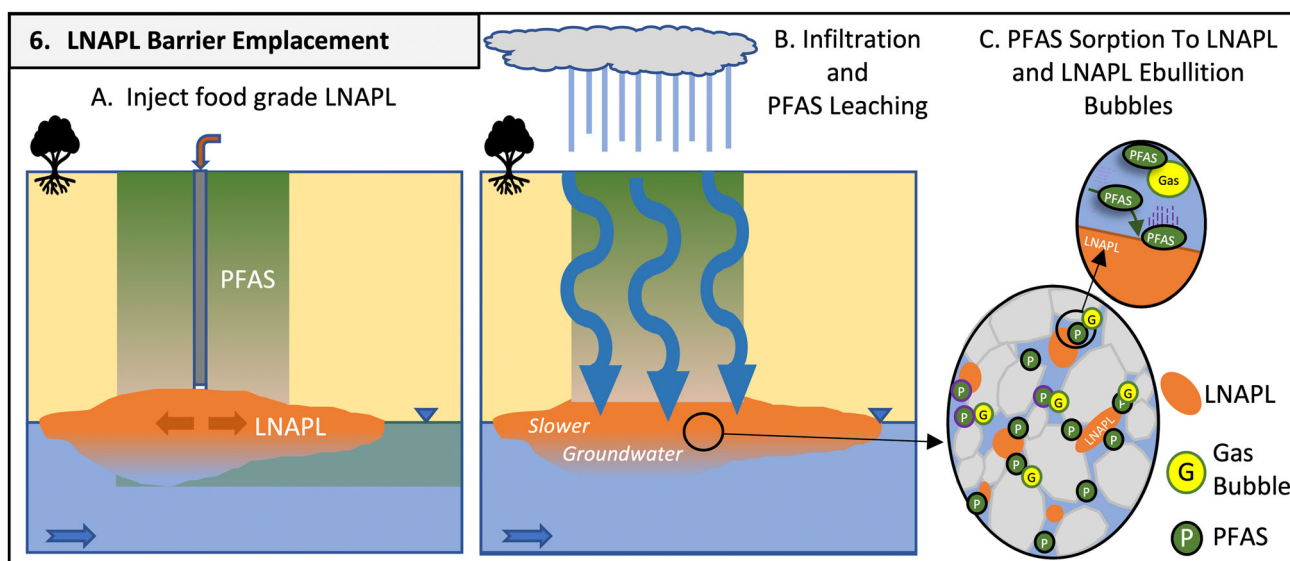


FIGURE 10 PFAS retention by intentionally emplaced LNAPL body. PFAS, per- and polyfluoroalkyl substances

LNAPL at PFAS sites where historical LNAPL is still present, if acceptable to environmental regulators.

11 | PFAS EA APPROACH 7: INJECT EVO TO ENHANCE CHEMICAL RETENTION

PFAS precursors can transform to PFAAs that do not degrade any further in groundwater under natural conditions. Maintaining precursors in the untransformed state is described as “chemical retention” by Newell et al. (2021a). If PFAS are kept in the precursor form, the environmental risk profile may be lower, because the greatest potential health risks are currently focused on PFAAs. Therefore, chemical retention is not a physical form of retention, but a manipulation of the chemical reduction-oxidation state of the PFAS.

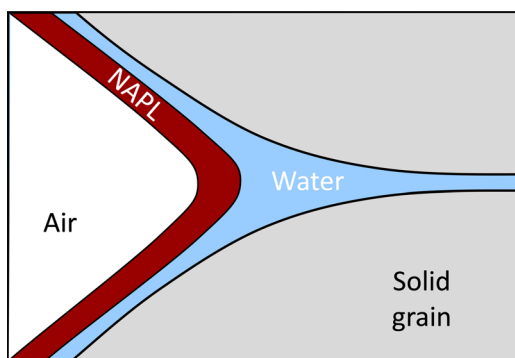


FIGURE 11 Unsaturated zone NAPL-water interfacial area becomes larger than that of saturated zone NAPL due to NAPL intermediate phase wetting (Adapted from Fagerlund, 2006)

Most of the dominant precursor reactions appear to be microbiological reactions facilitated by naturally occurring aerobic bacteria (Sharifan et al., 2021). While anaerobic biodegradation of some PFAA precursors has been documented, available data suggest that aerobic reactions are generally 1–2 orders of magnitude faster (Butt et al., 2013). A recent study conducted by Nickerson et al. (2021) demonstrated that injecting oxygen-sparged, simulated groundwater into laboratory columns containing AFFF-impacted soil resulted in a higher mass flux of PFAAs compared with injection of nitrogen gas-sparged groundwater. The authors attribute the difference to the availability of oxygen that could serve as a terminal electron acceptor for the biotransformation of precursors into PFAAs. Therefore, one potential way to sustain the chemical retention process at PFAS sites is to reduce the aerobic biodegradation rate by reducing the flux of dissolved oxygen into groundwater or into the vadose zone where precursors are present (Figure 12). This can be achieved by injecting organic substrate (e.g., neat oil or more likely emulsified vegetable oil or “EVO”) into source zones and plumes to consume available oxygen and create anaerobic conditions. EVO is a well-established technology for remediation of chlorinated solvents in groundwater. At chlorinated sites, EVO is used to consume oxygen and other electron acceptors (e.g., nitrate, sulfate, ferric iron) and facilitate the production of dissolved hydrogen, which is in turn used by dechlorinating bacteria for reductive dechlorination reactions (Wiedemeier et al., 1999). For PFAS sites, the EVO would be injected primarily to remove the dissolved oxygen and thereby prevent aerobic bacteria from converting PFAA precursors into PFAAs (see Figure 12). Since dissolved oxygen is easily consumed in the subsurface (i.e., the most energetically favorable electron acceptor), the mass or volume of EVO required at PFAA precursor zones may be much less than chlorinated solvent sites because the goal is only to remove the

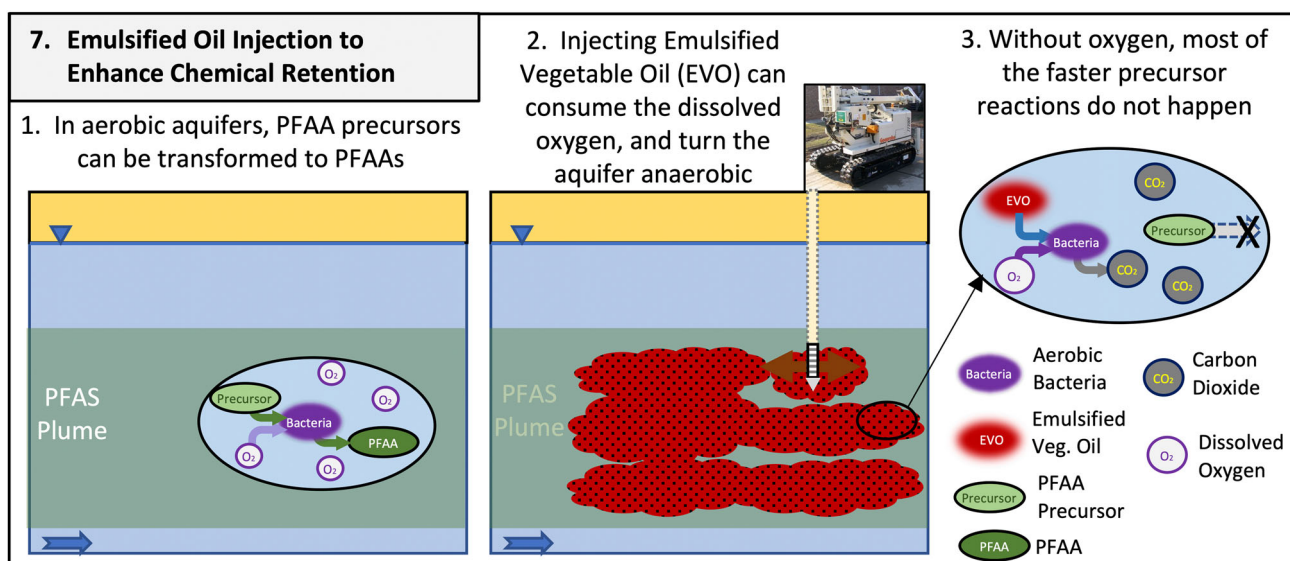


FIGURE 12 Conceptual depiction of PFAA precursor transformation in aerobic aquifers (left panel) and EVO injection to sustain chemical retention of PFAA precursors (right panel). EVO, emulsified vegetable oil; PFAA, perfluoroalkyl acid

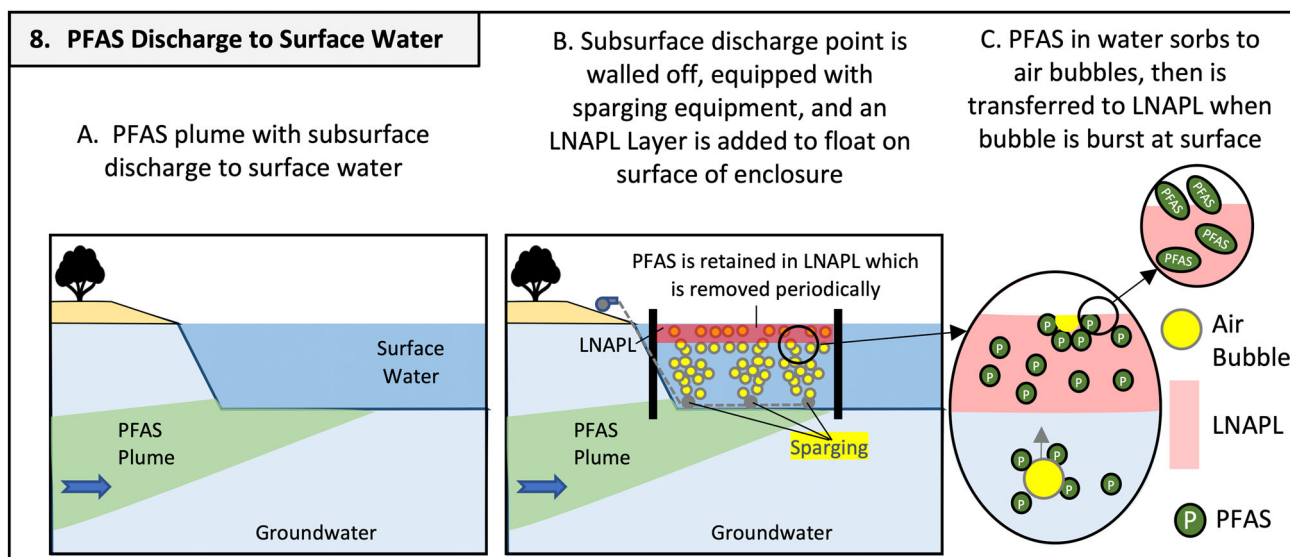


FIGURE 13 Retaining PFAS in submerged groundwater discharge, air sparging, and LNAPL. PFAS, per- and polyfluoroalkyl substances

dissolved oxygen but not generating dissolved hydrogen. Finally, additional retention of PFAAs and other PFAS may also be realized via the PFAS interfacial adsorption process at the EVO-water interface as discussed previously in EA Approach 7.

The EVO injection process is well understood, described extensively in a number of guidance documents and design tools (e.g., Borden et al., 2008; ESTCP, 2010; Suthersan et al., 2002), and has a robust supply chain supported by several vendors. A potential alternative design is to employ a thixotropic gel (a mixture of edible oil, water, and fumed silica) instead of EVO (Riha & Looney, 2015).

12 | PFAS EA APPROACH 8: CAPTURE OF PFAS GROUNDWATER DISCHARGES TO SURFACE WATER USING SPARGING AND LIQUID SORBENTS

Some PFAS plumes that discharge from the subsurface into surface water bodies (streams, lakes, etc.) may pose a special consideration with regard to PFAS retention. In this case, intercepting the plume using a groundwater pump and treat technology may be impractical as the presence of the surface water body can make it difficult to achieve hydraulic control of groundwater.

To address this challenge, the groundwater plume could be diverted to a control zone, where PFAS are directed to the water surface by air sparging and subsequently sorbed onto a floating oil layer that is periodically skimmed from the water surface (Figure 13). Potential design features include:

1. Constructing an open-top/open-bottom enclosure (e.g., caissons or sheet pile enclosures) around the perimeter of the groundwater-to-surface water discharge zone to collect the PFAS impacted groundwater that flows vertically upward;

2. Installing a sparging system (potentially solar powered) with sparge points or diffusers located at the base of the enclosure;
3. Emplacing a floating oil layer comprised of food-grade vegetable oil inside the enclosure;
4. Allowing the submerged groundwater discharge to flow into the caisson/enclosure and then exit out to the surface water a short distance below the floating oil layer;
5. Sparging bubbles through the surface water, which collect the PFAS at the air/water interface on the bubbles and migrate upwards where they travel through the oil layer into the atmosphere, transferring the PFAS to the oil where the PFAS are retained;
6. Removing the PFAS-containing oil periodically by a vacuum truck, potentially on the frequency of months or years, for treatment/disposal.

The retention and only periodic removal of the oil may provide operational advantages compared to a continually operated on-shore pump and treat system for hydraulic control. The technology may also be useful for ex-situ treatment applications. However, the technical feasibility of this approach has not been demonstrated either in laboratory experiments or in the field, and may prove to be impractical.

13 | CONCLUSIONS

PFAS-impacted soil and groundwater sites pose new challenges for remediation due to the unique physical and chemical characteristics of these compounds. In some cases retention-based MNA can serve to retain PFAS mass in the subsurface such that unacceptable levels of PFAS-impacted groundwater do not quickly reach the location of downgradient receptors. At other sites, however, natural retention

processes alone may not be considered sufficient to manage PFAS plumes. In these cases, EA, which has been developed and used for other contaminants, could prove beneficial for managing PFAS plumes as well.

In this paper, eight different retention-based EA approaches for PFAS plumes have been identified. Methods to enhance one or more particular PFAS attenuation processes in the subsurface are presented with a focus on retaining PFAS in the subsurface. The eight EA methods and their salient characteristics are summarized below:

1. Injection of Particulate Sorbents to Enhance Retention: *High retention, being used now.*
2. Capping to Retain PFAS in the Vadose Zone: *Simple way to reduce vadose zone mass flux.*
3. PFAS Sparging in Aquifers to Concentrate and Retain PFAS: *Repurposing sparging for retention.*
4. Retention Via PFAS Salting Out Processes: *Important for plumes crossing coastal shorelines.*
5. Emplacing Particulate Sorbents With Geotechnical Equipment: *Potential method for high retention.*
6. Intentional Food Grade LNAPL Barriers to Retain PFAS: *Inexpensive subsurface delivery.*
7. Inject EVO for Chemical Retention: *Preventing precursor transformation.*
8. Capture of PFAS Groundwater Discharges to Surface Water: *Combine sparging and oil.*

The eight proposed EA approaches leverage key retention processes that have the potential to impact PFAS fate and transport such as air/water interfacial sorption, hydrophobic interactions, NAPL/water interfacial sorption, electrostatic interactions, and "salting out" (Table 1). These approaches range from technologies that are currently being applied at PFAS sites (Methods 1 and 2) to early-stage concepts that have yet to undergo laboratory or field verification (Methods 3–8). Once subjected to a detailed process review or laboratory experimentation, some of the early-stage ideas presented in this paper may ultimately be deemed impractical or unfeasible. Injecting or emplacing activated carbon (Methods 1 and 5) would likely increase the retention more than other methods (Table 1), although the benefits from this retention only accrue in the actively treated area. Based on process knowledge, Capping (Method 2) will also likely provide a high level of retention of PFAS in the vadose zone.

Overall, the eight approaches have been compared in terms of their applicability, the potential increases in PFAS mass retention within the treatment area, their possible disadvantages, alternate designs, development status, and the likelihood of large-scale adoption. A qualitative framework based on travel time and mass discharge identifies general conditions that may drive the selection between PFAS MNA, EA, or more intensive plume management approaches. The information presented in the paper will be useful for PFAS site managers and regulators looking for alternatives to control plumes, and to researchers and practitioners who are interested in the development of early-stage technologies.

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CONFLICTS OF INTEREST

The authors believe that they have no conflict of interest that would compromise the findings of this paper. GSI Environmental is currently working on research projects involving related remediation processes (SERDP Projects ER21-1130, ER22-3221, ER22-3275; ESTCP Projects ER22-7381; ER22-7363). GSI Environmental has patent applications related to the use of sparging and colloidal gas aphanes (Newell et al., 2020; Newell et al., 2021e). One of the authors (Newell) is on the Scientific Advisory board of a company which provides segmented horizontal well drilling services.

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