ESTCP

EXECUTIVE SUMMARY

Developing a Framework for Monitored Natural Attenuation at PFAS Sites

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ESTCP Project ER21-5198

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ACRONYMS AND ABBREVIATIONS

CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
DoD	Department of Defense
ESTCP	Environmental Security and Technology Certification Program
LOE	line of evidence
MNA	monitored natural attenuation
NAVFAC EXWC	Naval Facilities Engineering and Expeditionary Warfare Center
PACZ	plume assimilative capacity zone
PER	PFAS enhanced retention
PFAS	per- and polyfluoroalkyl substances
PFAA	perfluoroalkyl acid
PI	Principal Investigator
PMR	PFAS monitored retention
RI/FS	remedial investigation/feasibility study
ROD	record of decision
USEPA	U.S. Environmental Protection Agency

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The project team consists of the following organizations and personnel:

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- Stroo Consulting
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- Naval Facilities Engineering and Expeditionary Warfare Center (NAVFAC EXWC)
  - Dr. Ramona Iery (Co-PI)

A draft of the "Framework" document that was the other primary project deliverable was reviewed by an Expert Panel and their feedback was incorporated into the final version. The project team greatly appreciates the time and effort that these panel members volunteered to the project: Chris Higgins (Colorado School of Mines), Jovan Popovic (NAVFAC EXWC), Robert Ford (U.S. Environmental Protection Agency (USEPA)), Michael Singletary (Naval Facilities Engineering Systems Command Southeast), and John Wilson (Scissortail Environmental).

Additional technical support was provided by several current and former employees / interns at GSI Environmental, including:

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1.0 INTRODUCTION

Groundwater sites with per- and polyfluoroalkyl substances (PFAS) present substantial challenges in terms of investigation and remediation, owing to factors such as stringent cleanup objectives, no known degradation processes for certain PFAS, and the apparent mobility and persistence of key PFAS. Given the lack of natural degradation for many PFAS and their persistent behavior in groundwater, the applicability of monitored natural attenuation (MNA) was initially considered limited. However, MNA as a remedy or site management approach has been accepted by environmental regulators for selected non-degrading metals, metalloids, and radionuclides (e.g., chromium, arsenic, and uranium) if site-specific geochemical conditions can *immobilize* (sequester) these chemicals of concern. That means that even though some PFAS do not degrade naturally, attenuation-based management approaches may be possible for PFAS plumes in groundwater because PFAS retention processes can contribute to attenuation.

There is a need for a retention-based PFAS site management approach to provide a wider range of remedial options for several reasons:

- There may be tens of thousands of PFAS groundwater sites that will require some type of management (Newell et al., 2020; 2022). Two estimates suggest there could be 50,000 to 60,000 potential PFAS sites in the U.S. (EBJ, 2022; Salvatore et al., 2022).
- There are no viable technologies at this time (late 2024) to destroy PFAS in situ in groundwater, and it is unlikely that the remediation industry can quickly manage all these PFAS groundwater plumes using only two technologies that are currently viable, groundwater pump-and-treat and in situ sorbents (Newell et al., 2022).
- While there are no confirmed natural processes that permanently sequester PFAS, natural processes can potentially *retain* PFAS in the subsurface for long time frames.
- Therefore, some type of site management approach for managing PFAS plumes based on PFAS retention could be an important factor in reducing the near-term risk associated with PFAS groundwater plumes at some sites (Newell et al., 2021a, b; 2022).
- Even after active remediation, it is likely that low residual concentrations of PFAS will remain in soils and groundwater but may be effectively controlled by retention as well.

As a result, the applicability of using processes that reduce PFAS migration rates and mass discharge rates is of considerable interest to site managers. This includes a variety of chemical and geochemical *retention* processes that can be used for PFAS plume management. As discussed within the full report, the project team introduced a variant of MNA for PFAS called the **PFAS Monitored Retention (PMR)** approach that site owners, their consultants, and environmental regulators can use to prioritize and manage their portfolios of PFAS sites. The PMR technology is implemented by utilizing a separate stand-alone framework document.[1]

[1] Framework for Evaluating PFAS Monitored Retention (PMR) at PFAS Groundwater Sites, ESCTP Project ER21-5198.

2.0 OBJECTIVES

The overall objective of this project was to develop and demonstrate how natural retention/ attenuation processes for PFAS can be used to evaluate if a particular site (or set of sites) is amenable to being managed by PMR and/or PFAS enhanced retention (PER).

This objective was addressed through the following project tasks:

1. ***Compile and document key knowledge and data to document technical basis for PMR.*** The overall objective of this task was to compile and integrate available information on PFAS retention and attenuation processes to develop the technical basis for PMR evaluations. This information was used to develop an initial "lines of evidence" approach for PMR, understand what processes need to be incorporated into site assessment, and to compile site data that may be useful for case studies of this approach.
2. ***Develop a field protocol for evaluating PMR.*** A general characterization protocol for demonstrating PMR based on measurements of retention in relevant subsurface compartments was developed. This protocol was based on collecting data that serve as lines of evidence for PMR, with a focus on using methods that are readily available to practitioners. The goal was to develop a three-tiered approach (Tier 1, Tier 2, Tier 3) used understand how to manage the level of effort for a PMR evaluation at a specific site, based on an understanding that the complexity and widely varying characteristics of PFAS sites necessitated differing degrees of effort for their appropriate management.
3. ***Outreach and Tech Transfer.*** The project was designed to have a significant outreach portion. This included a framework document that served as the key project deliverable and laid out the technical basis for PMR, including the lines of evidence required and the field protocol described above. The project team assembled an expert panel consisting of several regulators, academics, consultants, and Department of Defense (DoD) PFAS experts to weigh in on the entire spectrum of technical, regulatory, and social issues regarding using PMR that are laid out within the framework. The feedback from the panel was then incorporated into the final framework document (as well as the full report). Finally, the team prepared numerous articles and gave technical presentations on PMR to a wide variety of audiences via conferences, webinars, and journal articles.

3.0 TECHNOLOGY DESCRIPTION

Overview: The PMR framework developed during this project was based on the key retention processes identified for PFAS, including various sorption mechanisms, geologic matrix diffusion, geochemical conditions that limit the biotransformation of precursors, and dispersion of migrating PFAS plumes. Between sequestration/immobilization and retention, noting that while permanent sequestration of PFAS compounds had not been confirmed, significant long-term retention processes were likely present. The PMR framework incorporated a range of retention-based scientific knowledge describing each retention process and how to quantitively evaluate the impact of retention on PFAS plume migration. The framework could be applied to any PFAS in groundwater using applicable existing environmental criteria and/or using toxicity/risk data from a risk assessment.

The PMR framework was intended to support site managers during remedial decision-making, including developing and evaluating remediation alternatives during a remedial investigation/ feasibility study (RI/FS). A key intended use for this framework was to aid regulators and other parties who managed portfolios of PFAS sites. It was consistent with risk-based approaches that DoD used to support decision-making among their environmental restoration sites (i.e., the relative risk site evaluation process). In that case, the goal was to support the prioritization of PFAS sites to more effectively allocate resources for PFAS cleanup and management.

In a broad sense, PMR and PER processes offer several advantages in managing PFAS plumes, even in the absence of permanent sequestration. Three key advantages include:

1. **Plume Stabilization**: At some sites, retention processes coupled with dispersion could be substantial enough to stabilize the PFAS plume, thereby hindering further expansion. If the source of the plume has been removed or isolated, the PFAS plume will eventually diminish under such conditions.
2. **Plume Slow-Down**: Retention processes have the potential to slow the migration of PFAS plumes over time, thereby extending the period before receptors are affected. This slowing can allow site managers to concentrate on any immediate threats, thus allocating resources to sites necessitating prompt action. Additionally, this slow-down effect can give the site manager additional time to consider the most cost-effective existing or emerging remediation technology for managing potential groundwater impacts.
3. **Peak Dampening**: Some retention processes render the plume susceptible to potential “hysteretic” and rate-limited retention (Brusseau 2019; Stults et al., 2024). While the understanding of these mechanisms is still developing, they are based on observational and modeling studies that show that a portion of the sorbed chemical of concern mass may desorb slowly, which can cause tailing. At sites where retention capacity is substantial, these processes may attenuate the mass discharge of the plume by reducing the peak mass discharge, extending the plume discharge over a longer timeframe. This effect, also known as “Peak Dampening” (or sometimes “Peak Shaving”) decreases peak concentrations of PFAS at downgradient well locations, and in some cases could reduce them below applicable standards, much like how a reservoir manages downstream flood levels. Other processes that may contribute to peak dampening (or plume slow-down) include: (i) salting out that could occur as groundwater approaches coastal discharge zones with elevated salinity (Li et al., 2022) (note that salting out research is also ongoing with mechanisms that are incompletely understood), (ii) matrix diffusion of PFAS through pores (Schaefer et al., 2021; Farhat et al, 2022), and (iii) surface diffusion based on electrostatic interactions of PFAS with mineral surfaces (instead of hydrophobic sorption alone) that may enhance diffusion in and out of clays (Schaefer et al., 2021).

Lines of Evidence: For PFAS sites, the three lines of evidence originally developed for MNA of other chemicals of concern have been heavily modified to develop lines of evidence for PMR. Because there were no known processes that destroyed the fully fluorinated perfluoroalkyl acid (PFAA) class of PFAS in the environment, the demonstrated mass loss requirement for MNA line of evidence 1 has been modified to demonstrate that PFAS plumes have enough retention to pose no near-term risk to receptors.

The **First Line of Evidence** was to determine where a site fit into a PFAS plume management scenario based on two key retention metrics: mass discharge (Line of Evidence-1A) and travel time to potential receptors (Line of Evidence-1B). Sites that were most amenable for PMR would be sites with 1) relatively lower PFAS mass discharge; and 2) stable or shrinking PFAS plumes, or alternatively long travel times before the closest receptors were impacted (e.g., 10 years or more). Note that ongoing groundwater monitoring (and likely land use controls) would be required as an additional safety factor to ensure that receptors are protected.

A qualitative framework for managing PFAS sites was developed to help determine the suitability of PMR at a specific site (Figure ES-1). This framework required quantitative estimates for the two key factors controlling the magnitude of PFAS retention that served as the first line of evidence for PMR. Establishing relevant (site-specific) values for these two factors is discussed further in Section 3.4 of the full report (and in the framework document).

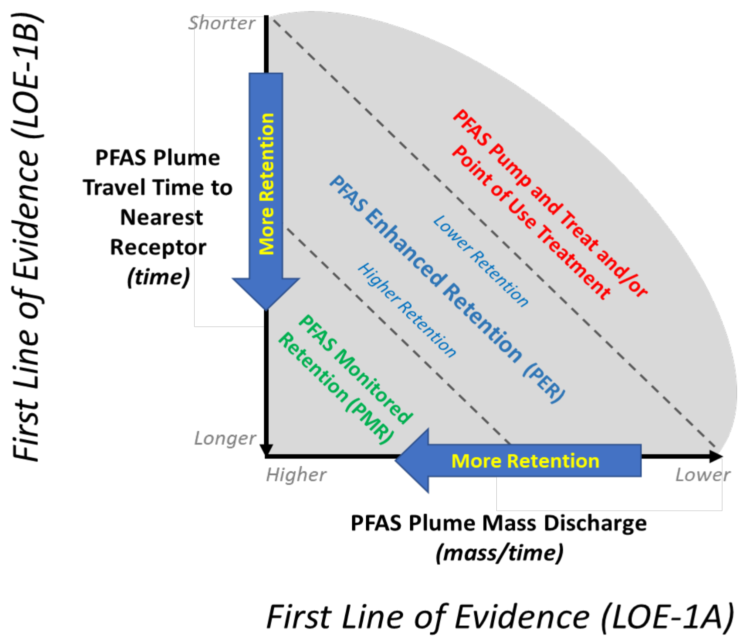

Figure ES-1. Primary Lines of Evidence for a PMR Evaluation.

The **Second Line of Evidence** relied on site-specific field data that helped demonstrate that specific retention processes were active at the site (see Section 3 of the full report). For example, conditions amenable for matrix diffusion (e.g., significant geologic heterogeneity, which can be enhanced by surface diffusion for some PFAAs) could be demonstrated in unconsolidated settings by collecting samples in low-permeability zones (e.g., silts, clays) and analyzing these for PFAS and soil organic carbon. The PFAS concentrations or mass could be compared to the concentrations or mass of PFAS in soils in the transmissive zones containing the PFAS plume. For sorption, co-located groundwater and soil samples could be collected to demonstrate significant ongoing processes in key compartments and to estimate field-based partition coefficients (Kd) that could be used to estimate plume transport retardation factors. For chemical retention, field sampling to establish both the geochemical conditions as well as the total mass present as precursors vs. PFAAs in various portions of the site could be conducted. The second line of evidence could also involve quantifying the mass discharge of PFAS from the unsaturated zone that was entering groundwater.

This could be done using direct porewater measurements (lysimeters), partitioning calculations, or leaching tests. One goal of unsaturated zone mass discharge was to evaluate if the contributions to groundwater were small enough to limit the need for soil cleanup.

The **Third Line of Evidence** used special field measurements or modeling studies to better establish how ongoing retention processes influenced PFAS transport to potential receptors (see Section 3 of the full report).

PFAS Enhanced Retention: PER approaches were intended to help manage those sites where natural retention mechanisms were not sufficient on their own to ensure that the primary lines of evidence (low PFAS mass discharge, long PFAS travel time to nearest receptor) for PMR could be met. At these sites, one of more of the key PFAS retention mechanisms may have been active, but available site-specific data indicated that some type of intervention would be necessary (currently or in the near future). In the context of the framework (**Figure ES-2**), these sites represented a higher priority for an action, but the conditions did not warrant the immediate implementation of an active source remediation (e.g., excavation) or the installation of a pump-and-treat system or point-of-use treatment to protect downgradient receptors.

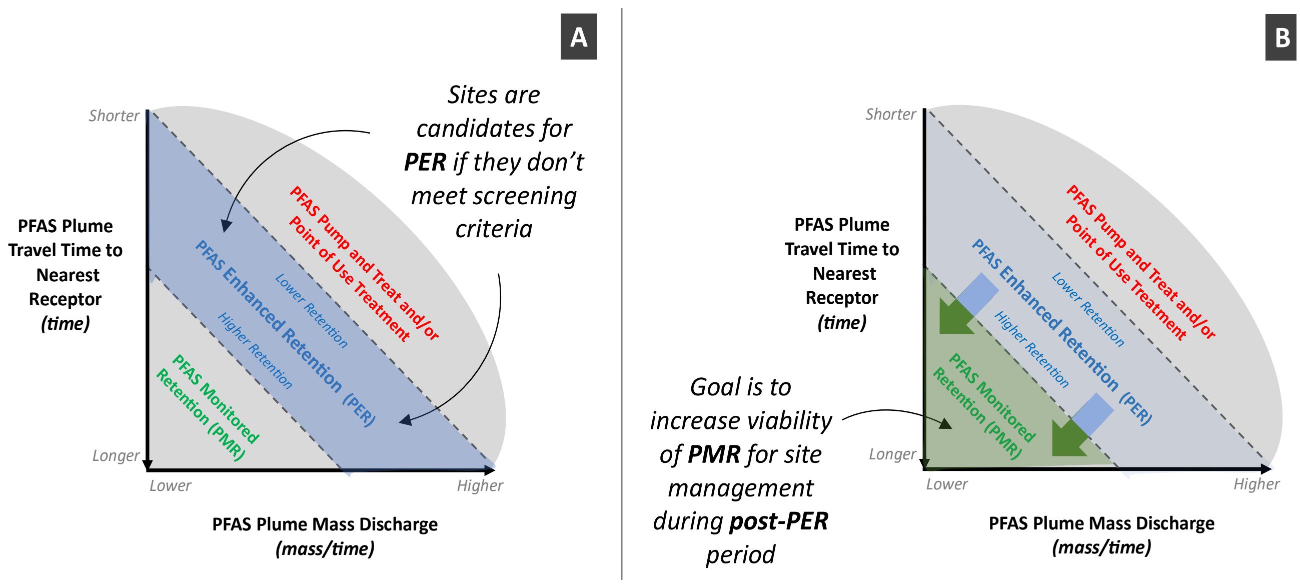

Figure ES-2. PER Reduces Mass Discharge and/or Increases Travel Time so that Site Can Potentially Be Managed Using PMR.

Evaluation Tiers: The PMR framework relied heavily on site characterization data, with fate and transport modeling as a complementary option to support the various lines of evidence. The complexity and widely varying characteristics of PFAS sites, as well as the resources that could be involved in obtaining data, necessitated differing degrees of effort for their appropriate management. Factors such as the source and plume size, proximity to receptors, and groundwater concentrations would markedly influence the required actions. As such, a stratified, three-tiered approach (Tier 1, Tier 2, Tier 3) based on the lines of evidence described above was suggested for potential PMR sites, according to their respective level of effort and complexity:

- **Tier 1 Evaluation:** A PMR study involving a limited number of conventional groundwater samples and soil samples may be sufficient for assessing PMR efficacy at less complex PFAS sites and/or sites with no or low immediate risk. It may also be appropriate as an initial screening step in evaluating a portfolio of sites, as a basis for making decisions about performing further (higher tier) investigations. The data gathered for the first and second lines of evidence could be based on this limited sampling. Simple data analysis techniques, such as extrapolating the PFAS plume length to estimate the length of the plume in the future, could be employed to get a conservative estimate of potential plume growth.
- **Tier 2 Evaluation:** Sites with increased complexity and risk, or where the existing data are limited or less certain, may necessitate a more detailed evaluation. This could include a more rigorous documentation of the first and second lines of evidence to better understand the critical site-specific retention processes. Furthermore, a basic Tier 2 groundwater modeling study could be conducted, incorporating matrix diffusion effects within a straightforward groundwater flow regime to estimate how much further plume expansion might be expected (e.g., the Environmental Security and Technology Certification Program (ESTCP) REMChlor-MD model or the ESTCP REMFluor model that is now being developed as part of ESTCP ER24-8200).
- **Tier 3 Evaluation:** The most complex sites with more immediate risk may warrant high-resolution site characterization techniques akin to those utilized by Adamson et al. (2020, 2022). These techniques augment the first and second lines of evidence, enhancing comprehension of mass distribution across different compartments, the extent of chemical retention on-site, and the mass flux relative to the distance from the source. A sophisticated three-dimensional model might be employed to better manage complex groundwater flow patterns and geological heterogeneity.

Defining whether a site is a Tier 1, Tier 2, or Tier 3 by complexity and risk is relatively subjective, but some guidance is provided in the framework. The key metrics of mass discharge and travel time to receptors could play a role in determining which Tier is best suited for a particular site, as well as other metrics.

4.0 PERFORMANCE ASSESSMENT

To evaluate the project performance, the following performance objectives were developed (Table ES-1). These objectives were primarily related to Task 1 of the project *(compile and document key knowledge and data to document the technical basis for PMR)*.

Table ES-1. Overview of Performance Objectives.

Performance Objective	Data Requirements	Success Criteria	Performance Objective Met?
Qualitative Performance Objectives			
Develop a comprehensive PFAS attenuation/retention library	Review all relevant SERDP/ESTCP projects, key journal articles, and databases	Expert Panel and ESTCP indicate all relevant PFAS transport/attenuation/retention papers/reports/databases have been integrated into key deliverables for this project	**Yes**
Quantitative Performance Objectives			
Obtain key hydrogeologic and PFAS characterization data at 3 to 5 DoD sites	Obtain site characterization reports, technical papers, and/or databases that can be used to evaluate attenuation/retention of PFAS plumes downgradient of PFAS source zones	Hydrogeologic and PFAS sampling data from at least 3 sites with PFAS data including PFAAs and Precursors from 4 or more groundwater points along a plume centerline	**Yes**
Positive evidence for attenuation at one or more sites	Hydrogeologic and PFAS data in both vadose zone and saturated zone	PFAS plume length is at least 25% shorter than what would be expected if no attenuation/retention in vadose zone and saturated zone	**Yes**
Demonstrate that PFAS plume attenuation rates (Ln concentration) vs. distance slope (k) at one or more sites are correlated to PFAS retention processes	PFAS monitoring data down plume centerlines and PFAS retention data in vadose zone and saturated zone	A difference in k of least a factor of two between the site with the highest and lowest retention	**Partially** • Zero-order rate constants for mass discharge did differ by more than a factor of 2 between the sites, with the highest rate constant observed for Site 2 (0.011 $kg \cdot yr^{-1} \cdot m^{-1}$) and the lowest rate constant observed for Site 3 (0.0013 $kg \cdot yr^{-1} \cdot m^{-1}$) • First-order rate constants based on total PFAS concentration were relatively similar for all three sites, differing by less than a factor of 2, The highest attenuation rate was observed for Site 1 (0.32 yr-1), while Site 2 (0.28 yr-1) and Site 3 (0.27 yr-1)

5.0 COST ASSESSMENT

There is an opportunity for cost-savings if using PMR is justified vs. implementing more aggressive remedial options (e.g., pump-and-treat). The project cost model did not attempt to project the cost savings associated with this type of outcome, but instead focused on the cost of evaluating PMR as part of the RI/FS stage. However, it should be understood that PMR was generally a cost-effective technology in terms of capital and operation and maintenance costs, and it also could reduce the environmental impact at these sites. As a result, the framework should promote the acceptance of retention-based strategies and increase the number of sites where it could be used as part of the remedial strategy.

The cost model identified and incorporated the following key cost drivers:

- The cost for collecting data on specific parameters or matrices that are not normally collected as part of a standard site assessment / remedial investigation.
- The PMR framework described a stratified, three-tiered approach (Tier 1, Tier 2, Tier 3) for PMR sites, such that the tier selected by site managers for initial characterization would be a key cost driver. The tiers assumed that the complexity and widely varying characteristics of PFAS sites necessitated differing degrees of effort for their appropriate management. Since these tiers are representative of the level of effort associated with collecting data, they could also be thought of as a step-wise process where the key metrics of mass discharge and travel time to receptors could play a role in determining which tier was best suited for a site and/or if further data should be collected.
- The labor cost associated with learning how to implement the PMR approach.

The cost model was applied for a single site at each of the Tier 1, Tier 2, and Tier 3 levels. Based on the assumptions used to develop the model (see Section 5 of the full report), the total cost for the PMR evaluation of a PFAS site ranged from $60K for Tier 1 evaluation, $164K for a Tier 2 evaluation, and $257K for a Tier 3 evaluation. These included all costs (minus any contingencies) that were associated with planning, implementation, sample analysis, data evaluation (including modeling options), and reporting.

6.0 IMPLEMENTATION ISSUES

6.1 REGULATORY

PMR can be evaluated as a remediation technology under almost any regulatory jurisdiction (including the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)) because regulatory frameworks generally allow for site-specific, risk-based approaches that consider non-destructive attenuation mechanisms such as sorption and matrix diffusion, provided the remedy is protective of human health and the environment and includes appropriate monitoring and contingency measures. Under CERCLA, PMR may be considered as part of a site-specific remedial action—particularly as an interim remedy, polishing step, or component of a phased MNA cleanup strategy—consistent with the U.S. Environmental Protection Agency (USEPA) guidance, which states that MNA approaches are acceptable if they meet the statutory and regulatory requirements for protectiveness, permanence, and long-term reliability (USEPA, 1999; Ford et al., 2007).

By adapting the framework to the specific requirements of each site, project managers can develop targeted and effective strategies for managing PFAS impact. In terms of its broader applications within the CERCLA process, the concept of a PFAS plume expanding indefinitely might preclude the use of PMR as a single final remedy in a final record of decision (ROD). Mass retention, when temporary, may be less favored by some regulatory authorities compared to remediation that involves permanent removal, destruction, or sequestration of the PFAS mass (for example, refer to USEPA MNA Directive, 1999). However, the PMR framework can be utilized in various other ways to effectively manage and remediate PFAS in groundwater:

1. ***Interim ROD with Contingency Remedy:*** This approach allows for the immediate implementation of PMR while providing a fallback option (e.g., pump-and-treat) if the plume continues to expand beyond acceptable limits.
2. ***Polishing Step after Initial Remediation:*** PMR can be employed after an initial remediation technology, such as source control, enhanced retention, or pump-and-treat, to ensure any residual PFAS impact can be effectively managed.
3. ***Resource Allocation Tool for PFAS Site Characterization:*** The PMR framework can serve as a tool to guide resource allocation for PFAS site characterization. By providing project managers with a process to classify each site, informed decisions can be made regarding resource allocation and timing of remedial actions.

6.2 PROCUREMENT

There are no significant procurement issues with using PMR (or PER) at PFAS-impacted groundwater sites. All methods can be implemented using standard equipment and analyses are either currently offered by commercial labs or expected to be offered through future lab developments (e.g., expanded PFAS analyte lists that include more precursors).

6.3 POTENTIAL LIMITATIONS

Nevertheless, there are several potential limitations to using retention as an attenuation process for PFAS plumes:

- *Processes are nondestructive.* Retention decelerates plume migration but does not eliminate PFAS mass. Presently, PFAA plumes are not known to degrade naturally, so unless the plume source decreases over time, some PFAA plumes may expand for extended periods (Farhat et al., 2022). To implement PMR, a plume assimilative capacity zone (PACZ) (Newell et al, 2021a) may be needed to accommodate plume expansion if there are no nearby receptors that would be impacted by this plume expansion.
- *Regulatory approval may be more challengin*g. Mass retention, when temporary, may be less favored by some regulatory authorities compared to remediation that involves permanent removal, destruction, or sequestration of the PFAS mass (for example, refer to USEPA MNA Directive, 1999).
- *Delayed impacts may be observed.* Although retention decelerates plume migration, if the plume source does not significantly attenuate over time and no other corrective measures are implemented, the PFAS concentration at some groundwater discharge points close to surface water bodies (for instance, a lake receiving groundwater flow) will ultimately reach the same level irrespective of retention effects, eventually posing the same risk to water users at a later time.

6.4 END-USER CONCERNS

Since PFAS retention processes slow the rate of PFAS plume migration but do not eliminate PFAS mass, the implementation of PMR (or PER) as a management strategy may require that a site have a PACZ downgradient of the source to accommodate plume expansion. PFAA plumes are not known to degrade naturally, so unless the plume source decreases over time, some PFAA plumes may expand for extended periods The use of a PACZ assumes that there are no nearby receptors that would be impacted by this plume expansion, and that the rate of plume expansion can be adequately estimated (using the methods outlined in the framework).

Measures should be taken to ensure the long-term effectiveness and reliability of PMR during the post-implementation period. PMR requires the implementation and use of appropriate monitoring to detect the rate of migration of PFAS over time. Sites may also require land use restrictions or institutional controls to prevent exposure and further reduce mass loading. Managing the uncertainties associated with retained PFAS mass is a critical long-term concern. PMR involves the long-term storage of PFAS-affected materials on site, which may create uncertainties and liabilities for site managers.

Finally, compared to other PFAS remedies, the application of PMR may require increased engagement and communication of risks with the stakeholders and the public. These concerns and questions may include the rationale and justification for PMR, the effectiveness and reliability of the approach, the potential impacts and risks of PFAS retention, and the future plans and actions for PFAS management. This means it is important to engage and communicate with the stakeholders and the public in a transparent and proactive manner, and to ensure that their concerns and questions are sufficiently addressed.

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