

EDITORIAL**TOPICAL COLLECTION ON PFAS ANALYTICS AND TREATMENT**

PFAS are forever? The state of the science and research needs for analyzing and treating PFAS-laden water

1 | INTRODUCTION

As discussed in this topical collection, per- and poly-fluoroalkyl substances (PFAS) have recently become a major issue within the water community and for the public at large. Studies have shown the following:

- Select PFAS has health consequences at very low concentrations.
- Several PFAS have been found in the environment at concentrations that raise concern.
- Treatment options for these select PFAS are few, expensive, and have limitations.
- Non-targeted analytical approaches continue to identify PFAS in drinking water sources, for which health, fate and transport, and treatment data are lacking.

This juxtaposition of factors combined with a lack of standardized analytical methods, health effects data, and treatment options for the vast majority of PFAS identified in the environment have created a complex regulatory and social dynamic. Without a firm regulatory framework, impacted water utilities and other entities can be pressured into making difficult choices. Ideally, any management approach chosen would be flexible enough to account for future health protection requirements along with changes in other factors, such as increased knowledge of additional PFAS because of advances in analytical techniques or changing influent PFAS concentrations. This flexibility ideally would include both the treatment of the water for its intended use, but also for handling any residual streams that would be disposed of in a managed system or via discharge to the environment. However, any built-in flexibility must be economically, managerially, technically, and environmentally sustainable.

To develop a sound management approach, more guidance is needed. This includes general information to set the stage for what PFAS need to be removed, including

- health effects information for all PFAS that have the potential for impacting human health,

- ecological impact information to allow for adequate determinations on discharges,
- occurrence information for these PFAS to determine relative source contribution, and
- PFAS analytical techniques for various relevant matrices.

Furthermore, treatment performance and costs are needed to decide strategies that can economically remove PFAS to levels that will not harm humans or sensitive ecosystems. In lieu of this information, internal and external pressure on a site or community may leave it no choice but to start analyzing for PFAS with current methods and installing treatment for removing these PFAS. Some removal efforts could proceed without a full understanding of the complete suite of PFAS occurrence information and potential health impacts, along with the benefits from the concurrent removal of other contaminants. The research covered within this topical collection includes many of these areas and points to future research directions that can help guide those in decision-making positions.

2 | SAMPLING AND ANALYSIS METHODS

Determining PFAS in water is challenging because

- PFAS may be present in a wide concentration range from parts-per-million (mg/L) to low parts-per-trillion (ng/L) levels;
- each category of PFAS contains a high number of individual compounds, many of which do not have analytical standards and/or isotopically labeled standards;
- PFAS are present in some consumer products and field or laboratory materials such that care is required to eliminate background contamination during sample collection, preparation, and instrumental analysis; and
- PFAS can sorb to sampling and laboratory materials, which can lead to underestimates of measured PFAS concentrations in water.

To determine PFAS levels in drinking water, the US Environmental Protection Agency (EPA) has introduced EPA Methods 533 and 537.1, which together target 29 PFAS. In both methods, water samples are prepared by solid-phase extraction before analysis by liquid chromatography tandem mass spectrometry (LC-MS/MS). To more comprehensively analyze PFAS, alternative approaches are used. These include suspect screening and non-targeted approaches using high-resolution mass spectrometry, the total oxidizable precursor (TOP) assay to determine the presence of unidentified precursors to perfluoroalkyl acids (PFAAs), and adsorbable/extractable organic fluorine methods to determine “total” organic fluorine by combustion ion chromatography.

The unique properties of PFAS can cause problems when conducting basic bench or field studies. For instance, the bottle choice is important not only for assuring accurate analytical measurements, but also for the design of the studies themselves. In this topical collection, Kleiner et al. (2021) assess the practical implications of using various bottle materials in ion exchange (IX) or activated carbon isotherm experiments. Silanized glass, polypropylene, and high-density polyethylene (HDPE) were screened. It is shown that five perfluoroalkyl sulfonic acids (PFSA) sorbed to HDPE bottles, and linear isomers of two PFSA exhibited enhanced adsorption relative to corresponding branched isomers. From these results, guidance for conducting IX and activated carbon isotherm studies are developed.

For sampling in the field, a demonstrated passive sampling technique would be extremely important because of the vagaries of PFAS concentrations seen in natural environments. Hartmann et al. (2021) demonstrate a passive sampler that can be used for PFAS, including C₄-C₉ perfluorocarboxylates (PFCAs) and C₄, C₆, and C₈ perfluorosulfonates (PFSAs) under a variety of solution conditions. The sampler uses a PFAS-specific adsorbent tailored for both hydrophobic and IX interactions. The adsorbent is an organosilica resin modified with cross-linked amine polymer with the addition of amine groups in combination with Cu²⁺. The samplers show an integrated linear response to concentration with time with all PFAS analytes tested except for short-chain PFAS (i.e., C₄ and C₅ PFCAs), and relatively low variability in sampling rate is demonstrated over a range of conditions including water matrices with elevated ionic strength and natural organic matter concentrations. Sampling times as short as 3 days are necessary to reach detection limits <70 ng/L for PFOA and PFOS.

Determination of PFAS sources is vital to protect drinking water sources. In this topical collection, Andrews et al. (2021) analyze the potential for environmental PFAS releases from contaminated sites, currently active industrial

Article Impact Statement

Readers will benefit from learning about the current status of PFAS sampling, analysis, and treatment techniques.

sites, and waste disposal sites. EPA's Enforcement and Compliance History Online (ECHO) database and specific case studies from Michigan and California demonstrate how PFAS source identification programs can provide crucial information for determining PFAS sources and therefore aid in developing effluent guidelines and pretreatment programs.

With regard to analytic methods, at the time we started curating this topical issue, the only available EPA Methods were Methods 537.1 and 533 developed for drinking water. These methods have also been widely used for source water assessments because of the lack of ambient water methods. To date, few studies have been published to support expanding the methods to other water matrices. Whitaker et al. (2021) evaluate the performance of these two methods for use with both potable and nonpotable waters. Both methods are found to be generally applicable for the analysis of PFAS in pristine nonpotable water matrices. However, low biased recoveries are notable issues for long-chain PFAS or those that elute late in the reverse-phase column because of surface adsorption loss and the water matrix. Also, there are extraction concerns for those PFAS for which matched isotopically labeled standards are not available. These challenges are successfully resolved or reduced by enhancing post-extraction bottle rinsing and elution procedures. The results of Whitaker et al. (2021) illustrate the challenge of simultaneously analyzing an extensive suite of PFAS with diverse properties. EPA recently published Draft Method 1633 targeting 40 PFAS and applicable to aqueous solids and tissue samples. The method is undoubtedly of great interest to many, albeit beyond the scope of the current collection.

Combustion ion chromatography (CIC) has been a valuable tool to determine total fluorine or total organic fluorine of environmental samples, but such a system is not commonly available. In this topical collection, Wang, Abusallout, et al. (2021), Wang, Yan, et al. (2021) tested the idea of modifying a total organic carbon analyzer—an instrument common in laboratories across disciplines—as an alternative approach. The modified system can serve as a screening technique or allow for a fluorine mass balance evaluation to help determine overall treatment or percent mineralization for destruction technologies. The hydrogen fluoride evolving from combusted samples is captured in

the impinger and analyzed by ion chromatography. The method detection limit based on PFOA spiked samples is 36 µg-F/L (52 µg PFOA/L). River water and wastewater have similar recoveries to those spiked in deionized water. Wang, Abusallout, et al. (2021), Wang, Yan, et al. (2021) identified some previously unknown limitations of CIC methods: possible variable recoveries across PFAS of different chain lengths or structures, and inorganic fluorine loss during combustion. The study emphasizes that suitable extraction and inorganic fluoride removal techniques are essential in this analysis to best define whether it is total, organic, or adsorbable organic fluorine.

3 | TREATMENT

Treatment decisions are generally complex because they have to account for a host of issues, including:

- the presence of other contaminants
- background water quality
- existing water treatment infrastructure
- available utility space
- hydraulic gradients
- compatibility with waters from other treatment works that may be blended in the distribution system
- discharge capabilities (e.g., sewer discharge)
- distribution system materials and potential corrosion concern

Additional issues are the utility's or managing entity's technical, financial, and managerial resources and capabilities, and how they match up with a proposed management strategy.

A great deal of work has been published on PFAS treatment as referenced in the articles in this topical collection. However, the basic technologies that are commonly applied for PFAS removal from water have not changed since PFAS became a concern. These include granular activated carbon (GAC) adsorption, IX, and filtration through high-pressure membranes—that is, nanofiltration (NF) and reverse osmosis (RO) membranes. This is understandable knowing the length of time needed to move a novel technology into the marketplace, especially if it includes potable water production given the potential health consequences that may occur if the system is designed poorly. Also, there is a need to factor in all aspects of certifying and permitting a new technology through state primacy agencies. This is not insignificant for drinking water applications; however, for those dealing with remediation waters, industrial streams, landfill leachates, or residual streams from a drinking water treatment process (e.g., activated carbon, IX resins, and NF/RO membranes), novel technologies may

find a niche from which they could expand to potable applications in the future.

Technologies besides GAC, IX resins, and NF/RO membranes include foam separation techniques that take advantage of PFAS's propensity to accumulate at air/water interfaces or adsorption onto clays or biosolids; these may be less expensive and could also have properties that result in the ultimate disposal or destruction of PFAS. Also, destructive technologies have shown the potential for PFAS removal, as discussed in this topical issues, perhaps best suited for high concentration streams. As demonstrated in the articles of this topical collection, recent research is working to advance the application of specific PFAS treatments.

3.1 | GAC and IX treatments

Communities have struggled with determining treatment choices that are both robust and economical. This is especially true for communities dealing with additional contaminants besides PFAS. For very small communities or individual rural homes affected by groundwater contamination, home treatment systems can be an option to reduce exposure. As with other aspects of PFAS treatment, PFAS removal by household point-of-entry (POE) treatment systems is not well-established. In this topical collection, Natarajan et al. (2021) test a POE adsorption system to remove PFAS and their precursors in a groundwater contaminated with aqueous film-forming foam. As expected from the literature, the long-chain PFAS are more effectively removed than short-chain PFAS, and PFCAs break through before PFSAs of the same chain length. The TOP assay highlights that precursors of short-chain PFCAs break through the filter system earlier than the measured PFCAs and PFSAs, but at the same general time as the dissolved organic carbon (DOC). Overall, under these conditions, the studied POE filters show 5 months of effective removal for all the PFAS measured, and over 7 months for PFAS other than select shorter-chain PFCAs. The results of Natarajan et al. (2021) therefore show that the studied POE system can be effective in managing PFAS contamination.

Also at the home system scale, Mulhern et al. (2021) show that in a study of 18 private wells in North Carolina, a commercially-available point of use (POU) carbon-block filter certified under NSF's P-473 protocol for PFOA and PFOS was successful in removing 17 PFAS detected in the filter influent, including short-chain perfluoroalkyl ether acids (PFEAs). The eight-month study covered the manufacturer's recommended lifetime for the device. Variations in the influent water quality (pH range of 4.4–7.1, DOC range of 0.6–1.2 mg/L) did not

have a significant effect on performance. This study demonstrates that POU devices operated under manufacturer's guidelines can be an effective approach for treating PFAS contaminated water, although additional data are needed to determine whether a wider range of site-specific conditions such as PFAS concentration or background water quality may affect performance.

IX treatment can have higher capacities as compared to GAC; however, higher media costs result in an unclear situation as to which media is the most cost-effective to achieve a final target concentration in any given water. As with GAC, optimization of IX resins is a research need, including evaluation of regenerable resins. To date, most systems have employed single-use resins. In this topical collection, Dietz et al. (2021) evaluate the regeneration efficiency of strong-base anion exchange resins for various PFAS concentrations, polymer compositions, and regeneration solutions. Generally, it is shown that a higher PFAS concentration in the influent resulted in higher PFAS loading on the resin and higher regeneration efficiency, and this was especially true for the polystyrene resin. Polystyrene resins showed greater removal of PFASs than PFCAs, and greater removal of long-chain PFAAs than short-chain analogs. The presence of organic cosolvents in the regenerant brine also improved regeneration efficiency.

3.2 | Scale-up and modeling of GAC and IX systems

Treatment performance models are useful in anticipating how a full-scale design will function when conditions or situations change, such as varying influent PFAS concentrations, flow rates, and future treatment goals. Compared with GAC models, there are few IX treatment models freely available. In this topical collection, Hauptert et al. (2021) discuss important theoretical and practical considerations for modeling PFAS removal from drinking water using gel-type, strong base anion exchange resin in batch and column processes. It is shown that several important limitations found in the literature preclude movement toward model development, including the use of inappropriate isotherms and not accounting for non-equilibrium conditions. Theoretical considerations are presented that will help future researchers develop models based on basic ion-exchange theory, and for designing and interpreting the results of both batch and column experiments.

Similarly, Kempisty et al. (2022) evaluate adsorption of 10 PFAAs onto two sub-bituminous, coal-based GACs at the bench-, pilot-, and full-scales to evaluate GAC scale-up procedures and to determine the effects of PFAA

characteristics and background organic matter on carbon use rates. Batch tests with pulverized GAC show the GACs perform similarly in groundwater, but the GAC with the larger mesopore volume is more effective for PFAA removal in surface water. The proportional diffusivity rapid small-scale column tests (RSSCTs) over-predicted full-scale capacity. The column tests show that carbon use rates decrease with increasing PFAS chain length and are lower for groundwater than for surface water. No differences are observed for empty bed contact times of 13 and 26 min in the full-scale adsorbers treating groundwater or in the proportional diffusivity RSSCT experiments. Based on a carbon use rate of $<25 \text{ mg}_{\text{GAC}} / \text{L}_{\text{water treated}}$ as a criterion for feasibility, the field-scale GAC adsorber data suggest GAC is a viable treatment option for long-chain and some short-chain PFAAs.

3.3 | Novel adsorbents

GAC and IX resins are the standard adsorbents generally accepted for PFAS treatment; however, there are numerous formulations for these adsorbents. Also, there are other more novel media that are known to remove organic contaminants. A media's acceptability is a function of many issues such as performance, cost, operational requirements, impacts on general water quality, and regulatory acceptance. In this topical collection, Li et al. (2021) review and discuss the potential of using functional bioadsorbents as low-cost alternatives to traditional adsorbents, and of particular interest is their potential removal of short-chain PFAS. The authors state that nano-scale biosorbents can be derived and properly functionalized in a cost-effective manner from raw biomass. The functionalized biomass sorbents were shown to effectively remove short-chain PFAS. Najm et al. (2021) presented RSSCT results for three commercially available bituminous coal-based GACs and one clay-based adsorbent containing quaternary ammonium groups. The three GACs are shown to perform similarly with initial breakthrough starting around 100,000 bed volumes except for PFBS (C_4 PFSA) and PFHxA (C_6 PFCA), which showed initial breakthrough at around 50,000 bed volumes. For the clay-based adsorbent, PFHxA was the only studied PFAS that appeared in the column effluent after treating 300,000 bed volumes.

Similar to IX resins, the clay-based sorbent could be operated at a shorter empty bed contact time and higher hydraulic loading rate compared to GAC. Grieco et al. (2021) expand on this theme by presenting batch equilibrium and RSSCT results for activated carbon and an emerging modified clay sorbent using drinking water with similar PFAS composition but varying dissolved

organic carbon concentrations. Under the conditions tested, both batch equilibrium and flow-through column experiments demonstrate that the modified clay has a greater adsorptive capacity and faster adsorption kinetics compared to activated carbon for select long- (PFOA) and short-chain PFAS (PFBS).

Adsorbent treatment schemes are employed in treatment plants to produce potable water, but they are also used in the subsurface to control the migration of contaminants. Wang, Yan, et al. (2021) show through batch experiments that modified bentonite clays and GAC can help minimize PFAS leaching from contaminated soils, which are long-term PFAS sources to groundwater and surface water. The modified clay is shown to perform the best in immobilizing many anionic PFAS whereas GAC is demonstrated to be most effective in preventing leaching of select cationic PFAS. Hardwood biochar, despite good performance in deionized water, is shown to have minor or negligible effects for all studied PFAS classes when present in soil slurry. Overall, the study shows potential options for reducing leaching from contaminated soils to minimize the continuous release of PFAS into water.

3.4 | Managing hard to treat water matrices

It is known that certain PFAS are poorly removed by GAC and IX resins, even in clean water matrices. There are no clear treatment choices for difficult to treat water. RO and NF membranes generally reject PFAS to a great degree, but because these separation processes do not destroy PFAS, they produce a concentrated waste stream. Therefore, there is a need to develop treatment technologies that can treat waste streams to prevent environmental and potential human exposure, possibly at geographically distant locations. In this topical collection, Tow et al. (2021) review a wide range of possible destruction, concentration, and sequestration strategies for the treatment of RO and NF concentrate streams. Cost issues are paramount, given the costs of both the initial membrane separation of PFAS and any subsequent concentrate management strategy.

The issues with membrane retentate streams also apply to other hard-to-treat waters such as landfill leachates, remediation waters, and wastewaters. To identify approaches for treating residuals from non-destructive water treatment processes, Ryan et al. (2021) review electrochemical technologies such as electrocoagulation and electrooxidation. It is found that electrochemical treatment is also promising for residual streams of IX, NF, and RO processes. However, much work must be

completed to account for environmentally relevant PFAS concentrations, high conductivity, and natural organic matter, limitations in treating short-chain PFAS, transformation products, and a community's technical and financial limitations.

McCleaf et al. (2021) examine foam fractionation treatment of landfill leachate. Foam fractionation is shown to give removal efficiencies of greater than 90% for PFOA, PFOS, PFHxS, and PFHpA, and 6:2 FTSA; between 50% and 80% removal for other PFAS such as PFNA, PFPeS, PFHxA, PFHpS, and PFBS; and 20%–50% removal for EtFOSAA, PFBA, PFDA, FOSA, PFPeA, and MeFOSAA. Increased removal efficiencies for the majority of the PFAS are demonstrated when the flowrate is increased, iron (III) oxide coagulant is added, conductivity is increased, and higher influent PFAS concentrations are applied.

Wastewaters can be challenging to treat and present possible health effects when considering reuse applications. Kumar et al. (2021) present data from the City of Altamonte Springs' potable reuse demonstration facility. The combination of ozonation, biological activated carbon filtration, ultrafiltration, GAC adsorption, and UV advanced oxidation shows the removal of long-chain PFAS (greater than five carbons for PFSA and greater than six carbons for PFCA) when the GAC adsorbers were operated to the point that would keep effluent TOC levels to below 3 mg/L, which is the regulatory limit in Florida for indirect potable reuse. Ozone followed by biological filtration increased PFAS concentrations due to oxidation of PFAS precursor material. GAC was determined to be the critical process for removing both PFAS and TOC, and the study shows the treatment trade-offs in a water reuse operation. Likewise, Gonzalez et al. (2021) present a project at Hampton Roads Sanitation District (HRSD), which recently initiated an indirect potable reuse project to recharge the Potomac Aquifer. The results show source control and GAC treatment maintained low concentrations of long-chain PFAAs (<8 ppt sum of PFOA, PFNA, PFDA, PFHxS, PFOS). Early breakthrough in the GAC column for short-chain PFAS (i.e., PFBA, PFPeA, PFHxA) and the high mobility of PFAS in the Potomac Aquifer support strict source control measures and the use of TOC to guide GAC operation.

4 | REGULATIONS AND PUBLIC COMMUNICATION

In the United States, the Safe Drinking Water Act authorizes EPA to set national health-based standards for drinking water to protect against both naturally-occurring and man-made contaminants found in drinking water. In this

topical collection, Alfredo et al. (2021) use a Relative Health Index (RHI) metric to quantify ranges of potential health risk reductions from state and federal PFAS threshold concentrations, and compare them with previous regulatory determinations—for example, arsenic and uranium—as well as with several currently unregulated contaminants. Alfredo et al. (2021) propose that given certain assumptions, the threshold concentrations for PFOA and PFOS derived from a number of references are not expected to achieve meaningful health risk reduction nationally (in the United States) compared with historically established benchmarks given known occurrence data. This study emphasizes the balance in regulatory decision-making between public health protection and the associated costs in the face of data uncertainty.

Due to the absence of, or differing, PFAS regulations, strategies are needed to communicate with the public clearly and effectively. Such communication can be challenging because of existing public opinion as well as the complexity of PFAS and their behaviors in the environment and through treatment. Harclerode et al. (2021) demonstrate a community-specific adaptive risk communication strategy that can allow municipalities to perform assessment and mitigation activities while addressing community concerns. This is demonstrated by two outreach strategies that addressed the respective municipality's planned communication messages following PFAS assessment actions. The two approaches demonstrate that risk perception factors differed, and hence a community-specific approach is likely needed for other cases. The authors point to the value of community engagement in developing risk management solutions.

5 | SUMMARY

As shown in this topical collection, a great deal of research is being conducted on the analysis and treatment of PFAS. The question remains as to whether PFAS can be mineralized or put into a state that will cause no further harm to the environment or the public. Analytical methods are improving in several areas, such as the ability to cover additional analytes in a wider range of matrices such as those found in treatment process residual streams, and standardizing techniques that bring down the cost and level of effort needed for analysis. Measurement parameters such as total adsorbable fluorine will be helpful as a screening technique, and offer the potential for helping close the fluorine mass balance for destructive processes.

For treatment, GAC, IX, and NF/RO membranes remain proven processes for water treatment that will be

considered by contaminated sites and utilities, especially for water sources that are not highly contaminated. GAC and IX are effective treatment processes for long-chain PFAS, but the specifics of treatment are shaped by site-specific factors. Each of these processes, however, have residual streams that need to be managed, and much is still to be learned regarding GAC reactivation, IX resin regeneration or incineration, and NF/RO concentrate treatment. Other novel technologies continue to be developed, and there is hope they will be up to the challenge of treating process residual streams and other hard-to-treat waters such as landfill leachates, wastewaters, membrane concentrates, and highly contaminated waters. Further research is needed.

AUTHOR CONTRIBUTIONS

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DATA AVAILABILITY STATEMENT

Data sharing not applicable to this article as no datasets were generated or analyzed for this editorial.

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