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Evaluation and Management Strategies for Per- and Polyfluoroalkyl Substances (PFASs) in Drinking Water Aquifers: Perspectives from Impacted U.S. Northeast Communities

Jennifer L. Guelfo,[1] Thomas Marlow,[2] David M. Klein,[3] David A. Savitz,[4] Scott Frickel,[2] Michelle Crimi,[5] and Eric M. Suuberg[1]

[1]School of Engineering, Brown University, Providence, Rhode Island, USA
[2]Department of Sociology and Institute at Brown for Environment and Society, Brown University, Providence, Rhode Island, USA
[3]Department of Pathology and Laboratory Medicine, Brown University, Providence, Rhode Island, USA
[4]Departments of Epidemiology and Obstetrics and Gynecology, Brown University, Providence, Rhode Island, USA
[5]Department of Engineering and Management, Clarkson University, Potsdam, New York, USA

BACKGROUND: Multiple Northeast U.S. communities have discovered per- and polyfluoroalkyl substances (PFASs) in drinking water aquifers in excess of health-based regulatory levels or advisories. Regional stakeholders (consultants, regulators, and others) need technical background and tools to mitigate risks associated with exposure to PFAS-affected groundwater.

OBJECTIVES: The aim was to identify challenges faced by stakeholders to extend best practices to other regions experiencing PFAS releases and to establish a framework for research strategies and best management practices.

METHODS AND APPROACH: Management challenges were identified during stakeholder engagement events connecting attendees with PFAS experts in focus areas, including fate/transport, toxicology, and regulation. Review of the literature provided perspective on challenges in all focus areas. Publicly available data were used to characterize sources of PFAS impacts in groundwater and conduct a geospatial case study of potential source locations relative to drinking water aquifers in Rhode Island.

DISCUSSION: Challenges in managing PFAS impacts in drinking water arise from the large number of relevant PFASs, unconsolidated information regarding sources, and limited studies on some PFASs. In particular, there is still considerable uncertainty regarding human health impacts of PFASs. Frameworks sequentially evaluating exposure, persistence, and treatability can prioritize PFASs for evaluation of potential human health impacts. A regional case study illustrates how risk-based, geospatial methods can help address knowledge gaps regarding potential sources of PFASs in drinking water aquifers and evaluate risk of exposure.

CONCLUSION: Lessons learned from stakeholder engagement can assist in developing strategies for management of PFASs in other regions. However, current management practices primarily target a subset of PFASs for which in-depth studies are available. Exposure to less-studied, co-occurring PFASs remains largely unaddressed. Frameworks leveraging the current state of science can be applied toward accelerating this process and reducing exposure to total PFASs in drinking water, even as research regarding health effects continues. https://doi.org/10.1289/EHP2727

Introduction

Per- and polyfluoroalkyl substances (PFASs) exhibit unique chemistry that makes them favorable for use in a wide variety of consumer and industrial products and applications (Kissa 2001). This same chemistry has led to limitations in using traditional environmental chemistry and engineering principles and techniques to understand and manage risks associated with their environmental releases. For example, unlike many neutral organic contaminants, in organisms PFASs are not lipophilic and are known to bind to proteins such as serum albumin (Conder et al. 2008). Additionally, some PFASs are environmentally persistent with no significant natural pathways for complete degradation following release. PFAS chemistry is largely attributable to the strength and low polarizability of the carbon-fluorine covalent bond (Banks et al. 1994; Kissa 2001). PFAS characteristics include thermal stability, chemical stability, surfactant behavior, and stain-resistant properties (Banks et al. 1994; Kissa 2001). Because of these characteristics, PFASs are used in products and applications such as firefighting foams, fluoropolymer manufacturing, stain-resistant coatings, and electroplating. These uses have contributed to their global distribution in organisms and the environment. At the same time, knowledge regarding human health impacts is quite limited, and because of their unique properties, conventional water-treatment techniques do not fully mitigate exposure (DeWitt 2015; Eschauzier et al. 2012; Giesy and Kannan 2001).

Recent studies estimate as many as 3,000 PFASs are now or have been on the global market (Wang et al. 2017). Within this group are perfluoroalkyl substances, which contain an alkyl tail with all carbons bonded to fluorine and which are persistent in the environment (Buck et al. 2011). These perfluoroalkyl substances include PFASs such as perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS), which have been the subject of much of the PFAS research to date. PFOA, PFOS, and their homologues (i.e., shorter and/or longer perfluoroalkyl carboxylates and perfluoroalkyl sulfonates) are often collectively referred to as perfluoroalkyl acids (PFAAs) (Buck et al. 2011). Polyfluoroalkyl substances have at least one perfluoroalkyl moiety (C_nF_{2n+1}) but elsewhere in the structure also contain carbons bonded to hydrogen. These compounds are capable of transformation in the environment (Buck et al. 2011). The terminal degradation products of polyfluoroalkyl substances include PFAAs (e.g., PFOA). So, they are often referred to as precursors and thus still represent a source of recalcitrant PFAAs in the environment (Harding-Marjanovic et al. 2015; Mejia Avendaño and Liu 2015). Examples include fluorotelomer sulfonates, some of which are capable of transforming to PFOA (Harding-Marjanovic et al. 2015).

Address correspondence to J.L. Guelfo, Brown University School of Engineering, 184 Hope St., Box D, Providence, RI 02912. USA. Telephone: (401) 680-0835. Email: jennifer_guelfo@brown.edu

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There are concerns about the human health impacts of some PFASs, particularly PFOA, PFOS, and other perfluoroalkyl substances. Briefly, in rodent studies, these compounds are known to affect lipid metabolism (e.g., Das et al. 2017) and liver weight (e.g., Loveless et al. 2006), decrease birth weight/increased resorptions (e.g., Lau et al. 2006), delay hind/fore limb phalanges ossification sites in offspring (Lau et al. 2006), delay mammary gland development in offspring (e.g., Tucker et al. 2015), and induce immunosuppression (e.g., DeWitt et al. 2008). Notably, these effects have provided the basis for regulation of PFOA/PFOS. In epidemiological studies conducted by the C8 Science Panel (C8 Science Panel 2017) and others, PFASs have shown positive associations with adverse outcomes, including elevated cholesterol (Frisbee et al. 2010; Nelson et al. 2010), ulcerative colitis (Steenland et al. 2013), thyroid disease (Lopez-Espinosa et al. 2012), testicular and kidney cancer (Barry et al. 2013), childhood adiposity (Braun et al. 2016), decreased duration of breast feeding in infants (Romano et al. 2016), and possibly preeclampsia (Savitz et al. 2012b, 2012a).

Routes of exposure to PFASs include diet (Fromme et al. 2007), dust (Shoeib et al. 2005), and drinking water (Hu et al. 2016). This exposure has led to development of a U.S. Environmental Protection Agency (U.S. EPA) drinking water lifetime health advisory (LHA) for the sum of PFOA and PFOS of 70 ng/L [70 parts per trillion (ppt)] (U.S. EPA 2016b, 2016a), and also regional drinking water standards in the low ppt range (NJDWQI 2016; VTDOH 2016). The U.S. EPA recently completed a national survey of six PFASs in U.S. drinking water that was targeted primarily at large, public, drinking water systems serving more than 10,000 people (U.S. EPA 2012). Studies analyzing these publicly available data (under the Third Unregulated Contaminant Monitoring Rule or UCMR3) have concluded that 6 million U.S. residents are served by systems exceeding the LHA (Hu et al. 2016; U.S. EPA 2012). The survey sampled equal numbers of systems sourced from surface water and groundwater, but approximately 72% of PFAS detections occurred in groundwater (Guelfo and Adamson 2018). Groundwater is the water source for 33% of public supplies in the U.S., and 90% of supplies in rural regions that rely on smaller (i.e., private) wells (USGS 2016). Collectively, health concerns and rates of occurrence highlight the important role of groundwater in human health risks associated with PFAS releases.

Numerous communities in the Northeast U.S. are currently assessing and managing risks due to PFAS-affected groundwater used as drinking water at the private, community, and public scales (e.g., Cape Cod, Massachusetts; Merrimack, New Hampshire; Portsmouth, New Hampshire; Hoosick Falls, New York; Paulsboro, New Jersey; and Bennington, Vermont). In many cases, discovery of these impacts occurred almost simultaneously in a period beginning in 2015, as a result of investigations initiated when residents and regulators learned of potential PFAS sources proximal to drinking water wells. Discovery of PFAS impacts led to a large group of regulators, consultants, analytical laboratories, and responsible parties (herein referred to collectively as stakeholders) with an immediate need to understand and manage risks associated with PFAS-affected groundwater and associated exposure. To determine risks associated with affected groundwater aquifers, stakeholders need information regarding sources, fate and transport pathways, affected receptors, sampling/analytical tools, health-based regulations, and water treatment technologies.

The Brown University Superfund Research Program (SRP) actively engaged with over a thousand stakeholders through PFAS workshops, analytical guidance, and other research translation efforts targeted at communicating the state of the science in the areas of PFAS chemistry, uses, sources, sampling/analysis, fate/transport, remediation, toxicology, regulation, and case studies. These efforts also provided an opportunity to hear diverse perspectives and learn of the key challenges faced by the broader scientific community in managing PFAS impacts. The objectives of this commentary are to (1) compile, critically evaluate, and share research translation findings to rationalize extrapolation of findings to other regions experiencing PFAS releases and (2) provide a framework to guide research and management strategies by prioritizing those PFASs that represent the highest risk of occurrence in treated drinking water. We conclude with an illustrative case study to demonstrate methods that can be used to address knowledge gaps regarding PFASs in drinking water to more effectively evaluate and mitigate risk.

Methods and Approach

The current evaluation utilizes a combination of research translation approaches, review of information in traditional publication outlets, review of publicly available data, and assimilation of select data sources into a limited case study of the risk of PFAS drinking water aquifer impacts in the State of Rhode Island.

Research Translation

To address stakeholder needs in the Northeast, a series of research translation activities were implemented beginning in 2016 to connect stakeholders (primarily from Connecticut, Massachusetts, Maine, New Hampshire, New Jersey, New York, and Vermont) with PFAS experts in various focus areas to communicate relevant aspects of research and current state-of-science (Table S1). Following these efforts, PFAS experts who participated in these events collaborated to document and review the challenges and knowledge gaps that are summarized in the current commentary. A literature review was used to evaluate the knowledge gaps in the framework of the current state-of-science. This review included web searches in the Web of Science database for all English language peer-reviewed articles (e.g., primary data, reviews, editorials) published 1995–present using title and topic search string PFOA OR PFOS OR PFAS in titles and topics from 1995–present ($n = 4{,}249$).

Illustrative Case Study

This commentary presents a geospatial case study to predict risks of PFAS impacts in drinking water aquifers. The approach required an inventory of potential PFAS release sites, which are defined for the case study as facilities that may be associated with the synthesis, use, or disposal of PFASs. We first reviewed peer-reviewed literature and regulatory data to understand potential PFAS source types and associated characteristics (Table 1). Next, we reviewed publicly available regional geospatial coverages and manufacturing directories related to these source types to build a database of facilities in the state of Rhode Island (Table S2). Hard-copy archives of historical manufacturing directories were converted into a digital database using tools described in Berenbaum et al. 2016. Briefly, an open-source data processing tool named GEOREG was created to process the scanned images of Rhode Island manufacturing directories and convert the text into geocoded historical industrial and manufacturing locations from 1950s–present. GEOREG also extracts additional information regarding a facility's name, address, standard industrial classification (SIC) code (i.e., manufacturing type), and number of employees. Resulting data include more than 11,000 unique historical and contemporary manufacturing sites. From this database, we selected only sites that matched the SIC codes (Table S2) and time frame (1960s–present) relevant to PFASs. The application of potential PFAS sources in a geospatial risk evaluation of potential PFAS impacts in Rhode Island groundwater is further discussed in the case study.

We used the characteristics listed in Table 1 to rank PFAS source types according to associated risk for causing groundwater PFAS impacts. First, PFAS source data coverages identified or

Table 1. Groundwater concentrations, compounds, relevant groundwater pathways, and affected receptors resulting from groundwater PFAS source types summarized from peer-reviewed literature and regulatory reports.

Source type	Magnitude of [PFAS] (μg/L)	Max PFAS	PFASs detected	Ground water pathways	Receptors impacted	Ref. cited
PFAS/FP manufacturing	10^{-2}–10^{3}	PFOA	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFOS	VZ to GW Atm SW to GW	DW, GW, SW, B	MDOH 2012; Davis et al. 2007; Bach et al. 2017; Dauchy et al. 2012; Weston Solutions 2009
AFFF use (DoD)[a]	10^{-3}–10^{4}	6:2 FtS	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFEtS, PFPrS, PFBS, PFPeS, PFHxS, PFHpS, PFOS, PFDS, 4:2 FtS, 6:2 FtS, 8:2 FtS, FHxSA, FOSA, 4:2 FtTAoS, 6:2 FtTAoS, PFBSaAm, PFPeSaAm, PFHxSaAm, PFHxSaAmA	VZ to GW	DW, GW, SW, B	Houtz et al. 2013; McGuire et al. 2014; Schultz et al. 2004; Moody et al. 2003; MDHHS 2016; Hull et al 2017; Moody and Field 1999; Barzen-Hanson and Field 2015; Backe et al. 2013
AFFF use (airport)	10^{-3}–10^{2}	PFOA	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFOS	VZ to GW	DW, GW, SW, B	Ahrens et al. 2015; Awad et al. 2011; Yingling 2016; Antea Group 2011; Delta Consultants 2010; Horsley Witten Group, Inc., 2016
AFFF use (fire training area)[b]	10^{-3}–10^{2}	PFOS	PFBA, PFPeA, PFHxA, PFOA, PFDoA, PFTriA, PFTreA, PFBS, PFHxS, PFOS, EtFASE, MeFASE	VZ to GW	DW, GW, SW	Antea Group 2011; Cape Cod Commission 2016
AFFF use (petroleum)	10^{-3}–10^{1}	PFOS	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFBS, PFHxS, PFOS, FOSA	VZ to GW	DW, GW	Antea Group 2011
FP coating (e.g. plastics, textiles, metals)	10^{-3}–10^{1}	PFOA	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFHpS, PFOS, FOSA, 6:2 FtS, 8:2 FtS	Not specified	DW, GW	U.S. EPA 2016c; NHDES 2017a
Electronics	10^{-3}–10^{1}	PFOA	PFHpA, PFOA, PFOS	Not specified	DW, GW	Unicorn Mgmt. Consultants 2016
Waste streams (landfills)	10^{-3}–10^{3}	PFBA	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, 6:2 FtS	VA to GW Atm	DW, GW	NHDES 2017a; Weston Solutions 2016; VTDEC 2016; Oliaei et al. 2006; Oliaei et al. 2013
Waste streams (biosolids)	10^{-2}–10^{0}	PFOA	PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFOS	VZ to GW	DW, GW, SW, B	Lindstrom et al. 2011
Waste streams (septic systems)	10^{-3}–10^{-2}	PFHxS	PFHxA, PFHpA, PFOA, PFBS, PFHxS, PFOS	VZ to GW	DW, GW	Schaider et al. 2016

Note: PFBA, Perfluorobutanoate; PFPeA, perfluoropentanoate; PFHxA, perfluorohexanoate; PFHpA, perfluoroheptanoate; PFOA, perfluorooctanoate; PFNA, perfluorononanoate; PFDA, perfluorodecanoate; PFUnA, perfluoroundecanoate; PFDoA, perfluorododecanoate; PFTriA, perfluorotridecanoate; PFTreA, perfluorotetradecanoate; PFEtS, perfluoroethane sulfonate; PFPrS, perfluoropropane sulfonate; PFBS, perfluorobutane sulfonate; PFPeS, perfluoropentane sulfonate; PFHxS, perfluorohexane sulfonate; PFHpS, perfluoroheptane sulfonate; PFOS, perfluorooctane sulfonate; PFDS, perfluorodecane sulfonate; 4:2 FtS, 4:2 fluorotelomer sulfonate; 6:2 FtS, 6:2 fluorotelomer sulfonate; 8:2 FtS, 8:2 fluorotelomer sulfonate; FHxSA, perfluorohexane sulfonamide; FOSA, perfluorooctane sulfonamide; 4:2 FtTAoS, 4:2 fluorotelomer thioether amido sulfonate; 6:2 FtTAoS, 6:2 fluorotelomer thioether amido sulfonate; 8:2 fluorotelomerthioether amido sulfonate (8:2 FtTAoS); PFBSaAM, perfluorobutane sulfonamido amine; PFBSaAM, perfluoropentane sulfonamido amine; PFHxSaAm, perfluoropentane sulfonamido amine; PFHxSaAmA, perfluorohexane sulfonamide amino carboxylate; EtFASE, *N*-ethyl perfluoroalkane sulfonamidoethanol; MeFASE, *N*-methyl perfluoroalkane sulfonamidoethanol; VZ, vadose zone; GW, groundwater; SW, surface water; (Atm.) atmospheric deposition and migration through the vadose zone; DW, drinking water, B, biota; DoD, Department of Defense; FP, fluoropolymer.

[a] Recent studies have identified 11 new classes of PFASs comprising 50 individual compounds in AFFF-impacted groundwater from DoD facilities (Barzen-Hanson et al. 2017b); these compounds are not listed here because quantification of their concentrations is not yet available.

[b] Represents fire training areas at municipal or private fire training institutions.

developed for Rhode Island were matched to a source type (i.e., Table 1 Source Type in Table 2). Next, Table 1 was used to determine the number of PFAS compounds known to be present and upper magnitude of PFAS concentrations measured in affected groundwater of each source type. Then, each source type was assigned a risk score of 25, 50, 75, or 100, with the maximum value assigned to those sites yielding highest PFAS groundwater concentrations and number of PFASs (Table 2). The risk score values themselves are arbitrary values used as multipliers along with the duration of operation for each identified facility in calculating a hazard index (HI). A raster of one by one km cells was overlaid on the sources in Rhode Island, and the HI values within a cell were summed (Figure 1). Next, each cell was assigned a groundwater vulnerability index (VI) based on Rhode Island aquifer classifications GAA, GA, GB, and GC (RIDEM 2012). We further divided GAA into groundwater recharge zones and wellhead protection areas. These classifications were given VI values of 100 (GAA, recharge zones), 80 (GAA wellhead protection areas), 60 (GA), 40 (GB) and 20 (GC). Summed HI values were multiplied by VI values to assign a risk index (RI) to each raster cell. Finally, we used a simple universal kriging procedure on the raster cell centroids to smooth RI values across the raster surface and provide some conservative interpolation of the RI values. We used the results to generate risk maps of the Rhode Island region. The risk scores, HI, VI, RI, and groundwater classifications are conceptually described in the discussion.

Results

Key Information Gaps

Sample collection. Investigation of human health and environmental impacts of any compound requires reliable sampling

Table 2. Risk scores utilized for calculation of the PFAS source hazard index (HI).

PFAS source	Upper magnitude (μg/L)	No. PFASs	Risk score	Table 1 source type
DoD facilities	10,000	28	100	AFFF use (DoD)
Chemical manufacturing	1,000	13	100	PFAS/FP manufacturing
Landfills	1,000	11	100	Waste streams (landfills)
Airports	100	28	75	AFFF use (Airports)[a]
Fire training areas	100	28	75	AFFF use (fire training areas)[a]
Petroleum refineries	10	28	75	AFFF use (petroleum refineries)[a]
Textiles	10	13	50	FP coating (plastics, textiles, metals)
Furniture	10	13	50	FP coating (plastics, textiles, metals)
Paper	10	13	50	FP coating (plastics, textiles, metals)
Rubber/plastics	10	13	50	FP coating (plastics, textiles, metals)
Fire Stations	N/A	28	25	N/A[a,b]
Fabricated metal	N/A	11	25	N/A[c]

[a] The number of PFASs reported for this source type was lower in the literature or no data were available (Table 1). A value of 28 was applied because this is the number of PFASs quantified at DoD AFFF-impacted facilities, and it is assumed that an equal number of PFASs may be present at all AFFF-impacted facilities.

[b] There were no data available on groundwater impacts due to fire stations, but fire stations were indicated as a probable source of groundwater impacts during stakeholder engagement. The overall risk score was presumed to be low because many fire stations do not store or use AFFF, and those that do have AFFF do not typically discharge the foams onsite. In personal communications with industry, municipal, and volunteer firefighters, some report that equipment cleaning may occur on site following AFFF use (oral communications, July 2014–July 2017).

[c] There were no data available on groundwater impacts due to electroplating, but data were available on PFASs in waste streams in the chrome plating process (U.S. EPA 2009). These data were used to determine the number of PFASs, and the upper concentration magnitude was the average of the magnitudes from other manufacturing sources.

techniques suitable for concentration levels that may represent health concerns. For PFASs, the challenge arises because of the low regulatory limits (U.S. EPA 2016b, 2016a), their ubiquitous nature (Prevedouros et al. 2006), and their use in the manufacturing process for some types of polytetrafluoroethylene (PTFE), which is a common laboratory material (Kissa 2001). Stakeholders are adopting special precautions (e.g., use of high- or low-density polyethylene containers and silicon tubing) when collecting samples for PFAS analysis (e.g., MassDEP 2017; NHDES 2017b). However, some stakeholders are adopting sampling and analysis protocols that include lists of unallowable items for which the need for prohibition is uncertain or not supported by scientific studies. These protocols include avoiding use of waterproof field notebooks, waterproof clothing, clothing laundered fewer than six times or laundered with fabric softener, cosmetics, and certain sunscreens (MassDEP 2017; NHDES 2017b). There is only limited evidence documenting the presence of PFASs in some of these products (Fujii et al. 2013; Keawmanee et al. 2015), and no cited published data measuring the potential for transfer of PFASs from these materials into samples during collection are available. Therefore, we conclude that the precautions represent an extremely conservative approach to avoid products and materials that include even trace amounts of PFASs. Data are needed to support prioritization of these precautions to avoid unnecessary inconvenience to field sampling personnel. When regulating in the low ppt level, we propose that understanding potential sources of background in samples is also key to differentiating between PFAS-affected drinking water and cross-contamination of samples.

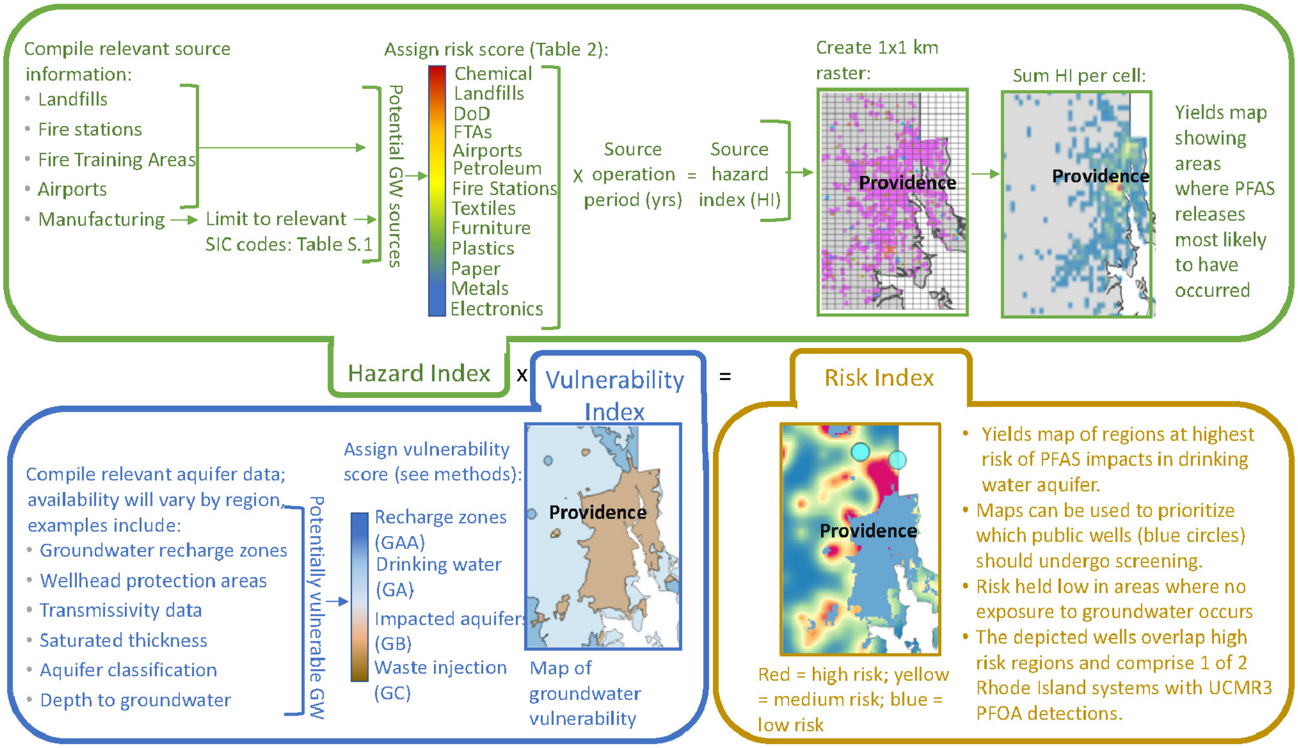


Figure 1. Overview of Rhode Island case study that utilizes a systematic approach to conduct a geospatial risk assessment of potential PFAS impacts in drinking water aquifers. Wells are shown with 1-mile buffers.

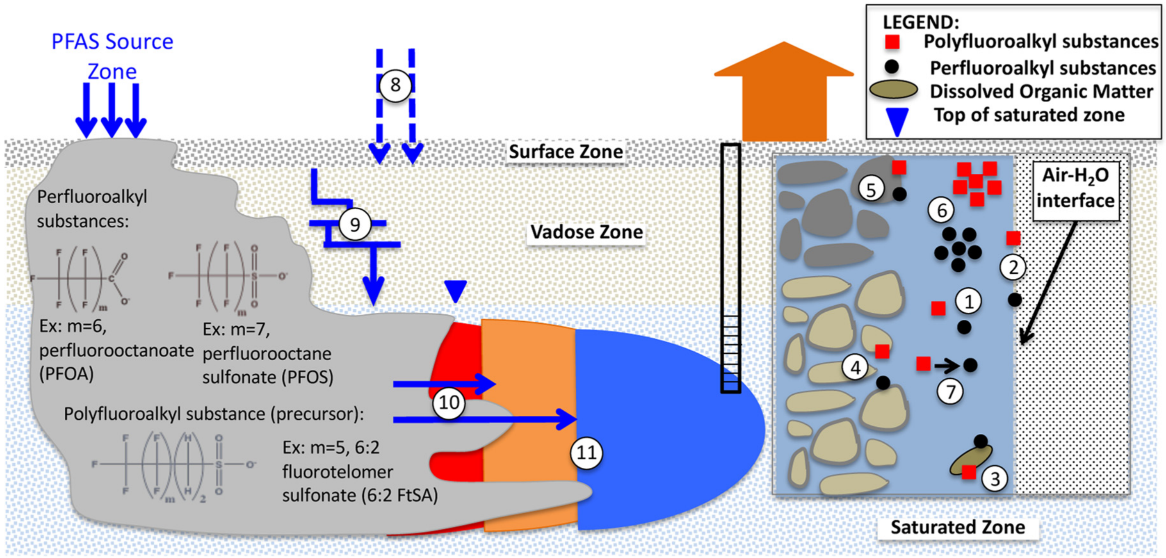
Targeted PFAS analysis. Despite notable progress over the past 20 y in utilizing liquid chromatography tandem mass spectrometry (LC-MS/MS) techniques (and gas chromatographic techniques for volatile PFASs such as fluorotelomer alcohols) for PFAS analysis (e.g., Mahmoud et al. 2009; Moody et al. 2001; Schultz et al. 2004), significant challenges remain. Many obstacles stem from the fact that the complete list of PFASs relevant to environmental and human health exposure scenarios is still unknown and ever increasing as more studies are completed identifying novel PFASs and precursor transformation products (e.g., Barzen-Hanson et al. 2017b). Therefore, though the majority of PFASs are suitable for LC-MS/MS analysis, standards needed to quantify them are not currently available, and it is difficult to keep pace with the increasing number of relevant compounds. It should also be noted that even implementation of targeted LC-MS/MS analysis may represent a challenge in terms of instrument expense, effort, and elimination of PFAS background issues (i.e., from aforementioned laboratory materials such as PTFE) for laboratories that are being required to address the issue. PFAS standards are commercially available (not including special order synthesis) for approximately 70 PFASs, and ~55% of those also have available isotopically labeled versions for use in isotope dilution approaches. It should be noted that in cases where a compound standard is available but a matching labeled standard is not, the labeled version of another PFAS may be used as an internal standard. For example, $^{1,8}0_2$- PFHxS has been used for analysis of PFBS (e.g., Guelfo and Higgins 2013; McGuire et al. 2014). In our view, these challenges make it virtually impossible for any regulatory authority to comprehensively specify which PFASs need to be investigated at a potentially affected site.

Additional analytical tools. Tools are available that can help to characterize the PFAS fraction not quantified during targeted LC-MS/MS analysis. These tools include the total oxidizable precursor (TOP) assay, high-resolution mass spectrometry (HRMS) analysis, particle induced gamma ray emission (PIGE) and adsorbable organofluorine (AOF) analysis. However, these methods are still limited in their availability and ability to quantify concentrations of individual PFASs present at a site. Detailed descriptions of TOP (Houtz and Sedlak 2012), PIGE (Ritter et al. 2017), and AOF (Wagner et al. 2013) are available elsewhere, but briefly, they enable measurement of total precursors, total fluorine, and total organic fluorine, respectively. Coupling TOP, PIGE, or AOF with targeted LC-MS/MS analysis can help researchers understand the total PFAS load present in a sample but does not result in identification of all individual PFASs present. High-resolution mass spectrometry (HRMS) using technology such as quadrupole time of flight generates high mass accuracy data that can be used in identification of unknown compounds (Barzen-Hanson et al. 2017b; Strynar et al. 2015), but quantification of PFASs without standards remains a challenge. TOP has begun to emerge as a commercially used technique, but availability of PIGE, AOF, and HRMS is often limited to noncommercial research laboratories, leaving limited access for regulators and other practitioners who want to implement these tools. During research translation events, stakeholders reported that these challenges prevent them from developing conceptual models of affected sites that include a complete list of PFASs to which environmental and human receptors may be exposed.

Source zone identification. Another key knowledge gap is PFAS source zone identification, which can be illustrated through comparison with the legacy groundwater contaminant, methyl tert-butyl ether (MTBE). MTBE was historically used as a gasoline fuel additive with peak use occurring from 1992–2005 (U.S. EPA 2016d). MTBE has high aqueous solubility (~40 g/L), has low soil sorption, and is slow to degrade, leading to the potential for more extensive groundwater plumes relative to other legacy contaminants, such as benzene (Squillace et al. 1996). MTBE had a single primary use (U.S. EPA 2016d), so point sources were commonly facilities or infrastructure associated with retail gasoline supply and distribution for which current and historical information is typically available. Though indirect sources of MTBE groundwater contamination such as atmospheric this deposition are possible, this would primarily contribute to disperse, low-level background concentrations (Squillace et al. 1997) not likely to pose a threat to human health or the environment.

When considering the sources of PFASs and comparing with the example of MTBE, we conclude the following. PFASs also may be highly water soluble with weak soil sorption and exhibit recalcitrance to natural degradation, leading to the potential for large groundwater plumes. However, unlike MTBE, there are many relevant PFASs and diverse products and applications with which they are associated (Table 1). Additionally, although PFASs may be associated with a particular process or product, such as textiles manufacturing, it cannot be concluded that all products and manufacturers in a relevant industrial category utilized PFASs. Finally, due to regulatory limits in the low ppt range, indirect sources of PFASs (e.g., groundwater impacts due to leaching of atmospheric deposition or land application of composts and wastewater biosolids) have led to environmentally relevant groundwater impacts (Lindstrom et al. 2011; Shin et al. 2011) and cannot be discounted as important PFAS sources. Assembling information on all potential PFAS sources in a particular region is further complicated because information on current and historical locations of both direct and indirect sources is often missing or unconsolidated. For stakeholders who need to identify sources of known PFAS impacts or to design targeted screening programs to assess if releases have occurred, unconsolidated source data may lead to inefficient or untargeted sampling plans, a failure to identify all sources relevant to a particular release, or the inability to determine a source, thereby increasing time required to reduce risks to public health and the environment.

Subsurface fate and transport. Although studies have investigated PFAS fate and transport, significant knowledge gaps remain (Figure 2). There are knowledge gaps in the general areas of PFAS composition [Figure 2 (1)] partitioning [Figure 2 (2–6)] transformation [Figure 2 (7)], and the influence of site hydrogeology and geochemistry [Figure 2 (8–11)]. An understanding of fate and transport requires knowledge of both compound-specific (e.g., sorption, transformation) and site-specific (e.g., geology, geochemistry) factors (Fetter 1999). PFAAs are recalcitrant (Prevedouros et al. 2006), so the primary compound-specific factors that need to be understood are sorption and potential for generation from precursors. Polyfluoroalkyl substances (i.e., precursors) can transform (Harding-Marjanovic et al. 2015; Mejia Avendaño and Liu 2015; Weiner et al. 2013), so in addition to sorption, knowledge of transformation rates and pathways (e.g., intermediate products) is required. These processes rely on compound-specific properties, such as sorbate structure, but are also influenced by site-specific properties, such as sorbent type, solution chemistry, and cocontaminants (Guelfo and Higgins 2013; Higgins and Luthy 2006; Weber et al. 2017). Further, PFAS distribution at the field scale also depends on subsurface hydrogeologic conditions, including groundwater flow direction, velocity, and influence of heterogeneous geology (Fetter 1999). Finally, user-friendly groundwater modeling tools for legacy contaminants have been developed to assist stakeholders with decision points, such as site prioritization, monitoring plans and duration, and design of aquifer remediation alternatives (Aziz and Newell 2000; Newell et al. 1996), but comparable tools



Description	Key Knowledge Gaps
(1) Dissolved PFAS (see Table 1 for current state of science)	Total PFAS composition at key points (surface, vadose, saturated zones) at PFAS-impacted facilities
(2) PFAS air-water partitioning[1,2]	Impact of PFAS water-air surface activity on partitioning in unsaturated media
(3) PFAS sorption to dissolved organic matter	Potential for PFAS to partition to DOM and exhibit facilitated transport, similar to other contaminants
(4) PFAS sorption to solids[3-10]	Potential for PFAS sorption hysteresis, polyfluoroalkyl sorption, impacts of soil/solution chemistry on polyfluoroalkyl sorption
(5) PFAS sorption to solids altered by co-contaminants or remedial activity[10--13]	Impact of co-contaminants, remediation on sorption/desorption of polyfluoroalkyl substances
(6) PFAS micelle/hemimicelle formation	Potential formation of PFAS micelles or hemimicelles as concentrations increase and saturation decreases
(7) Precursor biotransformation to perfluoroalkyl substances[14-16]	Transformation pathways, applicability of laboratory precursor transformation rates at the site level
(8) Infiltration	Impacts of infiltration and resulting changes in saturation on PFAS transport through the surface and vadose zones
(9) Vadose zone hydrogeology	Influence of fractures, root zone, low permeability zones, or capillary zone on PFAS migration to the saturated zone
(10) Saturated zone hydrogeology	Impact of variable hydraulic conductivity or other heterogeneous subsurface conditions on saturated PFAS transport
(11) Transition from reducing to oxic conditions[7,10]	Potential for redox conditions caused by co-contaminants or their degradation products (e.g. CH_4) to influence zones of precursor transformation

Figure 2. Conceptual model of micro and macroscale PFAS fate/transport processes and associated knowledge gaps. Superscripted numbers refer to the following references: [1]Kissa 2001; [2]Banks et al. 1994; [3]Higgins and Luthy 2006; [4]Liu and Lee 2007; [5]Liu and Lee 2005; [6]Ferrey et al. 2012; [7]Ololade et al. 2016; [8]Tang et al. 2010; [9]Barzen-Hanson et al. 2017a; [10]Weber et al. 2017; [11]Guelfo and Higgins 2013; [12]McKenzie et al. 2015; [13]McKenzie et al. 2016; [14]Harding-Marjanovic et al. 2015; [15]Mejia Avendaño and Liu 2015; [16]Weiner et al. 2013.

are currently not available for PFASs. In our opinion, these challenges in understanding PFAS fate and transport limit the ability to identify at-risk receptors, understand the probability for a source to affect those receptors, and to help identify potential sources of newly discovered groundwater releases.

PFAS toxicology and use in regulation. Other efforts have summarized the current state of science regarding PFAS toxicology (ASTDR 2015; DeWitt et al. 2009, 2015; Lau et al. 2007; Negri et al. 2017) and epidemiology (Bach et al. 2015, 2016, Chang et al. 2014; Negri et al. 2017; Steenland et al. 2010). In our view, results of toxicology studies highlight several challenges related to understanding health effects of PFASs. First, studies address the toxicology of only a subset of PFASs. Outside of PFOA/PFOS, toxicology studies are available for other PFAAs, such as PFBA mouse studies (Das et al. 2008; Foreman et al. 2009), PFBS rat studies (Lieder et al. 2009a, 2009b), a PFHxA rat study (Loveless et al. 2009), PFNA mouse studies (Das et al. 2015; Fang et al. 2008), a PFNA rat study (Feng et al. 2010), and a PFHxS rat study (Butenhoff et al. 2009). Studies are also generally limited for polyfluoroalkyl compounds (Buck 2015), especially recent replacement products, although some data are available for PFOA replacement products GenX (Beekman et al.

2016; Caverly Rae et al. 2015; Gannon et al. 2016) and ADONA (Gordon 2011). Second, the mechanism of action for PFAA-associated toxicity is not well understood, although peroxisome proliferator-activated receptor alpha (PPARα) activation is often implicated (Das et al. 2015; Lau et al. 2007). Third, although animal studies are useful in elucidating target organs, there are notable differences in how humans and animals interact with PFAAs. For example, PFAAs are documented to have half-lives on the order of years in humans (Li et al. 2018; Worley et al. 2017) but only hours to days in laboratory animals (Lau et al. 2007). Long half-lives indicate human serum concentrations will remain elevated, suggesting toxicity in humans may persist even after environmental PFAS levels decrease. Further, applying PPARα activation in animal studies to humans is complicated by several species differences that are well described elsewhere (Post et al. 2017). Fourth, the potential for synergistic toxicity is not well characterized, despite human exposure to PFAS mixtures. Two related *in vitro* studies found that mixtures of 2–4 PFASs yielded additive, not synergistic, activation of murine PPARα (Carr et al. 2013; Wolf et al. 2014). However, as noted, questions remain regarding the role of PPARα and the applicability of animal PPARα studies to humans. Additionally, *in vitro* studies are unable to capture pharmacokinetics alterations that may lead to synergistic toxicity.

Knowledge gaps in toxicology pose challenges for regulators and other stakeholders tasked with managing PFAS releases. Although data are available for PFOS and PFOA, there is still a lack of consensus regarding which toxicological end point and subpopulations should be targeted in development of drinking water standards. This lack of consensus can be illustrated through comparison of the U.S. EPA LHAs (U.S. EPA 2016b, 2016a) to standards developed for PFOA in New Jersey (NJDWQI 2016) and Vermont (VTDOH 2016). Drinking water quality standards (DWQS) are generally calculated as follows (e.g., U.S. EPA 2016b, 2016a):

$$DWQS = \frac{RfD * BW}{DWI} * RSC \qquad [1]$$

where RfD is the reference dose (i.e., the maximum daily dose for which no adverse health effects are expected to occur), BW is the body weight, DWI is the drinking water ingestion rate, and RSC is the relative source contribution, or proportion of PFAS exposure from drinking water. The LHA (U.S. EPA 2016b, 2016a) and DWQS differ between New Jersey (NJDWQI 2016) and Vermont (VTDOH 2016) in part because of key differences in the values used to calculate these standards (Table 3). In standards development, values for DWI may change, depending on the subpopulation considered (e.g., adults, children), and, in the case of PFOA agencies, differ on which rates are deemed adequately protective: lactating women (U.S. EPA), BW adjusted rate for the first year of life (Vermont), or adult water intake (New Jersey). Table 3 also illustrates that there is debate regarding the most appropriate RfD. Specifically, the NJDWQI believes that the EPA failed to consider more sensitive end points, such as the liver and immune effects for which the NJ RfD is considered protective (NJDWQI 2016). The NJDWQI also expressed concerns, which we share, regarding low-dose findings, such as lack of repetition, nonmonotonic data (delay in phalanges ossification and mammary gland development), and unknown clinical significance (mammary gland development without disruption of lactation, delay in phalanges ossification without malformations, mild reductions in immune factors without increase incident of infection) (NJDWQI 2016).

Although all approaches result in standards in the low ppt range, these variations lead to different interpretations of what would be considered an affected drinking water system. For example, the U.S. EPA UCMR3 efforts sampled ∼5,000 public drinking water systems in the U.S. for six PFASs, including PFOS, PFOA, and PFNA (U.S. EPA 2016e). The number of systems in the data set that would be considered problematic based on the U.S. EPA LHAs more than doubles if the New Jersey or Vermont standards are applied to the data set (Guelfo and Adamson 2018). Additionally, different standards may be applied to drinking water of adjacent communities separated by state lines. Such is the case in New York and Vermont, raising questions about why a community in one state may continue to drink groundwater that would be considered unsafe by an adjacent community across a state line.

Groundwater remediation. Conventional treatment techniques are ineffective for removal or destruction of the full suite of PFASs present in affected water (e.g., Rahman et al. 2014; Schultz et al. 2006). For example, processes relying on *in situ* chemical oxidation cannot fully destroy all PFASs but can enhance oxidation of precursors to end point PFAAs (Houtz and Sedlak 2012). Additionally, they may destroy perfluoroalkyl carboxylates (e.g., PFOA) under some conditions but are ineffective at degrading perfluoroalkyl sulfonates (e.g., PFOS) (Bruton and Sedlak 2017; Park et al. 2016). Filtration with granular activated carbon (GAC) and anion exchange resins (AER) have been shown to remove PFOS and PFOA but may not be as effective for treatment of short chain PFAAs and precursors (Appleman et al. 2013; Xiao et al. 2017; Yu et al. 2009; Zaggia et al. 2016). Further, filtration does not achieve compound destruction, so additional treatment or disposal of spent media is required. There has been some success at the bench and pilot scale using advanced oxidation processes that rely on electrochemical or plasma-based techniques to destroy PFASs in extracted, affected groundwater (Chaplin 2014; Stratton et al. 2017). Despite significant progress, these techniques are generally not ready for full-scale implementation, and key concerns include potential treatment-rate limitations and energy requirements. Last, design of any treatment technique may also need to account for cocontaminants (e.g., hydrocarbon constituents) that may be present in some aquifers (McGuire et al. 2014).

Despite knowledge gaps, filtration with GAC is a common technique used to address PFAS in affected drinking water systems (e.g., Damon 2016; NYDEC 2016; Weston & Sampson 2016). Although effective for treatment of PFASs currently targeted for regulation (i.e., PFOS, PFOA), these systems are not optimized for removal of the full suite of PFASs present in some affected groundwater. In our view, this suggests exposure to PFASs for which toxicological outcomes are not yet fully understood may be ongoing. In the event that water quality standards are developed for additional PFASs, we also point to the potential scenario that sites formerly remediated for PFOS/PFOA will need to be revisited for treatment of PFASs not previously considered. To minimize these

Table 3. Values used in development of PFOA advisories and standards and associated maximum recommended levels in drinking water.

Agency	Advisory or standard (ng/L)	RfD (mg/kg – day)	DWI/BW (L/kg – day)	RSC	Toxicological end point	Reference
USEPA	70	2.E-05	0.054	0.2	delay in phalanges ossification, mice	U.S. EPA 2016b, 2016a
NJDWQI	14	2.E-06	0.029	0.2	Hepatoxicity, mice	NJDWQI 2016
VTDOH	20	2.E-05	0.175	0.2	delay in phalanges ossification, mice	VTDOH 2016

Note: NJDWQI, New Jersey Drinking Water Quality Institute; VTDOH, Vermont Department of Health; DWI, drinking water ingestion rate; BW, body weight; RSC, relative source contribution.

potential risks, we agree with others that it may be necessary to implement combined remedies or treatment trains (Crimi et al. 2017; Kucharzyk et al. 2017) and argue that these approaches should target removal or destruction of total PFASs present.

Discussion

Often during site assessment, evaluation of levels of exposure and potential human health consequences are of paramount concern (U.S. EPA 1989). In our view, even when areas such as occurrence, fate/transport, and remediation are well understood, the health consequences of PFASs will remain uncertain. Although the volume of research in both toxicology and human health studies has increased markedly in the last decade, firm conclusions relating individual PFASs to specific health outcomes have remained elusive. As noted, a wide range of potential links between PFAS exposure and health outcomes have been reported (e.g., ASTDR 2015; Steenland et al. 2010), but the uncertainties remain substantial. As one extends that interest into other legacy PFASs and particularly into the newer generation of PFASs, the empirical evidence guiding interpretation of health effects declines substantially and is virtually absent for many of the compounds, suggesting a need for strategies to prioritize PFASs for further study.

PFAS Framework

Frameworks have been developed in previous studies to evaluate large groups of compounds and prioritize those that should be targeted for further research on factors such as analysis, occurrence, fate and transport, and treatability (de Voogt et al. 2009; Howard and Muir 2010, 2011, 2013; Kumar and Xagoraraki 2010; Strempel et al. 2012), but these approaches often rely at least in part on toxicity information, the limitations of which have already been discussed for PFASs. Nevertheless, we propose that additional criteria used in these approaches, such as occurrence, persistence, and treatability, might be coupled with evaluation of exposure into a framework to guide future research and inform best management practices (Figure 3). Despite challenges outlined herein, progress has been made in understanding drinking water occurrence, persistence, and treatability, such that it is possible to begin identifying PFASs that should be targeted for further study. That is particularly the case for evaluation of exposure (Step 1), and here we present a limited case study illustrating a risk-based evaluation of the potential for PFAS exposure in drinking water due to presence of potential source zones. Last, we note that the body of publicly available information regarding aspects such as compound persistence and toxicity continues to grow as part of legislative efforts, such as the European Union's Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) (ECHA 2017a). REACH requires industry to identify and communicate risks associated with substances they use (ECHA 2017b), creating a valuable resource that can be applied toward compound prioritization using this framework.

Geospatial Evaluation of PFAS Exposure Risk

As discussed, there are challenges in identifying sources of and exposure to PFAS groundwater impacts on a regional scale. We present a case study of potential PFAS groundwater impacts in the state of Rhode Island to illustrate methods that can be used to overcome these challenges. Previous studies have assessed regional risks of degraded groundwater quality due to other classes of contaminants by compiling information on potential sources of contamination (e.g., population-dense regions, landfills, gas stations) and comparing their location with groundwater that is vulnerable to impacts (Babiker et al. 2005; Rahman 2008). Groundwater vulnerability may be evaluated through hydrogeologic characteristics, such as depth to groundwater and transmissivity (Gemitzi et al. 2006; Wang et al. 2012). A similar approach can be adopted for PFASs by compiling data on potential PFAS sources and groundwater vulnerability and applying a risk-based system to evaluate the potential for PFAS impacts (Figure 1).

As described in the Methods section, risk scores were assigned to each PFAS source type (Table 2). This score was based on the fact that not all PFAS sources are likely to be associated with the same (or any) severity of groundwater effects. A limitation to the calculation of the risk scores is that the number of PFASs detected at a given type of site may be a product of the limited PFASs that were analyzed. For example, many precursors could be present that have never been investigated. Additionally, some source types. such as electronics facilities and metal plating. had limited or no groundwater data from affected sites, so the upper magnitude of concentrations may not be representative. Nevertheless, the data are considered to be representative in terms of understanding the different source types relative to each other. Further, the resulting source ranking that reflects Department of Defense (DoD), chemicals manufacturing, and landfills as the highest risk sites is consistent with literature and regulatory reports in terms of capturing release types that are known to have caused significant drinking water aquifer PFAS impacts (Davis et al. 2007; Moody et al. 2003; NH DHHS 2016; Oliaei et al. 2013). Risk scores were used to calculate HI values (see Methods) which, when mapped, depict areas where PFAS releases are most likely to have occurred (Figure 1).

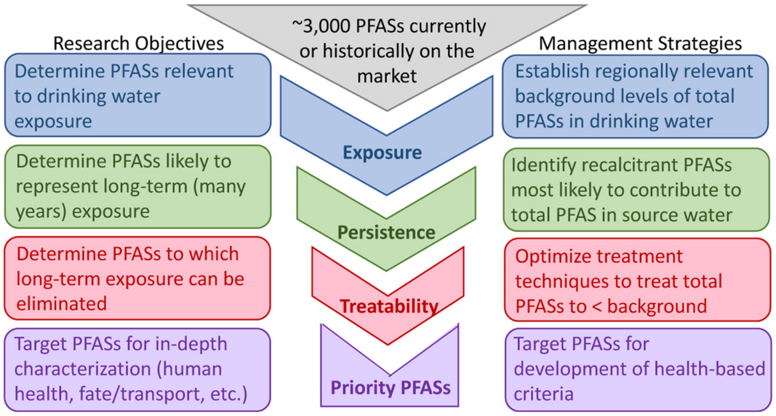


Figure 3. Framework for research and management strategies that prioritize PFASs based on highest risk of exposure in drinking water. It should be noted that here exposure refers only to drinking water; other routes of ingestion, such as food, are not considered.

The HI, or likelihood of PFAS release, should not be used as the sole indicator for potential aquifer impacts and subsequent exposure because releases that occur must also be transported through the subsurface to the saturated zone in order to affect aquifers, and certain hydrogeologic conditions make this more likely (i.e., higher groundwater vulnerability) in some regions (Fetter 1999). Rhode Island aquifer classifications already represent an evaluation of groundwater vulnerability. The State of Rhode Island classifications are based on locations of shallow zones of recharge to deeper aquifers and wellhead protection areas (GAA), drinking water aquifers evaluated based on transmissivity and saturated aquifer thickness (GA), aquifers presumed or known to be degraded (GB), and groundwater where waste injection is permitted (GC) (RIDEM 2012). VI values based on these classifications (see Methods) define the risk of environmental releases affecting usable groundwater when such events occur (Figure 1). When the HI and VI are combined to calculate RI, the highest RI values represent areas at highest risk for PFAS impacts in drinking water and subsequent exposure (Figure 1).

Results of the evaluation of groundwater PFAS impacts in Rhode Island reveal high HI values centered around regions with a high density of former manufacturing facilities that are situated in the population-dense region of Providence. Although there is a high likelihood of a PFAS release having occurred in this region, the RI values near Providence are low due to depressed VI values. Aquifers in the immediate vicinity of Providence are classified as GB, indicating that there is no use of the groundwater (RIDEM 2012), so there should be no drinking water exposure to PFAS impacts present. The highest RI values occurred in more rural regions with a lower density of sources (i.e., lower HI) but higher VI values due to proximity to groundwater recharge areas and drinking water aquifers. RI maps can be applied towards understanding potential sources of known PFAS groundwater impacts or in prioritizing drinking water wells that should be targeted in sampling programs, with the ultimate goal of understanding and mitigating risks associated with PFAS exposure. In Rhode Island, the majority of wells in high RI regions are private or small community wells, which were not screened as part of U.S. EPA UCMR3 screening efforts. Notably, two PFOA detections were discovered in Rhode Island as part of UCMR3, with concentrations of $20-81\ \mathrm{ng/L}$. The geospatial evaluation of PFASs in Rhode Island aquifers found that wells in both of these systems (some systems are sourced from multiple wells) have overlap with areas of high RI (Figure 1). Finally, it should be noted that this approach represents a limited case study for illustrative purposes, and efforts to further ground truth, refine the geospatial approach, and characterize which (if any) PFASs are present are part of ongoing research.

Conclusions

In summary, interactions with stakeholders from affected communities in the Northeast U.S. have identified a number of key knowledge gaps in several areas, including sampling and analysis, fate and transport, toxicology, regulation, and water treatment. An important result is a lack of consensus regarding management and regulation of PFASs in drinking water. Both laboratory and epidemiological studies support the potential for negative health outcomes due to PFAS exposure. In response, water quality regulations for PFASs are starting to emerge, but these regulations primarily apply to PFOS/PFOA. Regulatory levels are based solely on extrapolation from mechanistic studies in animal models and incorporate substantial uncertainty factors as a margin of safety. We conclude that these recently recommended standards and advisories for select PFASs should not be interpreted as indicating that health will be adversely affected if levels are exceeded. Rather, in explaining the health implications of elevated levels of PFASs in water sources to various stakeholders, it is important to be clear that knowledge is limited, in most cases severely so, and that declaration of safe or harmful levels of contamination is not possible.

Despite this uncertainty, we believe that it is important not to be complacent about human exposure to PFASs via drinking water, and strategies are needed to begin addressing water quality impacts even as research is ongoing. Therefore, we conclude that understanding knowledge gaps will help to guide investigation, management, and mitigation of specific releases, and that the framework developed here can be used to facilitate broader strategies for research and management focused on total PFASs in drinking water. The latter will help accelerate the process of mitigating exposure to PFASs for which detailed studies are lacking.

Results presented herein suggest that it is possible to begin implementing a comprehensive strategy towards PFAS management despite the considerable gaps in current knowledge, particularly regarding toxicity. In particular, this work compiles an already a large body of evidence related to potential PFAS sources and occurrence in groundwater (Table 1) that can be applied toward understanding exposure. We illustrated this by performing a risk-based, geospatial case study of potential PFAS source zones in Rhode Island drinking water aquifers. When compared with limited groundwater aquifer results, high-risk zones identified in the geospatial evaluation were proximal to drinking water wells with detectable PFAS concentrations (Figure 1). Further, new regulations such as REACH have led to increased sharing of industry data related to compound behavior in the environment, and this sharing helps build connections between research in academia, industry, and government. In our view, growing such partnerships facilitates effective management of chemical use and release. This commentary focuses on streamlined research strategies and best management practices for PFASs in drinking water, and this focus could be extended to evaluate other routes of exposure. Similar approaches might also be applied to other complex mixtures of aqueous contaminants with the overall effect of leveraging the current state of the science towards understanding drinking water impacts and reducing risks to human health.

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