UNIVERSIDADE DE SÃO PAULO

ESCOLA POLITÉCNICA

GABRIEL NEGRELLI GARCIA

PFAS Remediation in Soil and Groundwater: A Systematic Literature Review of Innovative Technologies from a Sustainability Perspective

São Paulo

2022

GABRIEL NEGRELLI GARCIA

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Versão Corrigida

Monografia apresentada à Escola Politécnica da Universidade de São Paulo como parte dos requisitos para a obtenção do título de Especialista em Gestão de Áreas Contaminadas, Desenvolvimento Urbano Sustentável e Revitalização de Brownfields.

Orientador(a): Profa. Dra. Marilda Mendonça Guazzelli Ramos Vianna

São Paulo

2022

Catalogação-na-publicação

Garcia, Gabriel
PFAS remediation in soil and groundwater: a systematic literature review of innovative technologies from a sustainability perspective / G. Garcia -- São Paulo, 2022.
72 p.

Monografia (MBA em MBA em Gestão de Áreas Contaminadas, Desenvolvimento Urbano Sustentável e Revitalização de Brownfields) - Escola Politécnica da Universidade de São Paulo. Departamento de Engenharia Química.

1.Remediação do solo 2.Contaminação do solo 3.Águas subterrâneas 4.Sustentabilidade 5.Perfluoroalquil I.Universidade de São Paulo. Escola Politécnica. Departamento de Engenharia Química II.t.

ACKNOWLEDGEMENTS

I would like to thank my mother Maria Silvia and my father Homero for wishing me into existence. *Minha mãe sempre foi uma fortaleza em minha vida e me deu todo o amor e apoio do mundo. Te amo muito, mãe. Obrigado por tudo.* In this journey, I have also had the most amazing sister, Adriana, as a role-model and inspiration, as well as someone I could trust with my doubts and insecurities. For my family, I am forever grateful.

Along the way, I have also made incredible friends, all of whom have helped me to become who I am today, and to accept myself and cherish my identity. Thank you for all the love, support and laughs – in particular, my high school family, Pudim.

I am also endlessly fortunate to be married to the most amazing man in the world – Juan – who has shown me nothing but love and support and has been a source of strength and inspiration for the past four years. Not in a million years, could I have dreamt of someone that could make me happier. *Je t'aime profondément.*

I am incredibly grateful to my therapist, Vivian, without whom I would not be as comfortable in my skin as I am today and able to pursue my dreams as I have been doing since we first met in 2016.

I am also grateful to Ramboll Brazil for providing me with financial support to complete this MBA.

I want to also acknowledge that I have had the honor of learning from great professors at the University of São Paulo from 2013 to 2018, and now again from 2020 to 2022 throughout this MBA. For their guidance and patience in this project, I would like to thank Professor Marilda Mendonça Guazzelli Ramos Vianna and Teaching Assistant Gabriela Paupitz – it has been a pleasure learning from you and working with you.

In closing, as Freud once said: "From error to error, one discovers the entire truth". For the freedom to err and learn, I am also thankful.

RESUMO

Negrelli Garcia, Gabriel. **PFAS Remediation in Soil and Groundwater: A Systematic Literature Review of Innovative Technologies from a Sustainability Perspective**. 2022. 72 f. Monografia (MBA em Gestão de Áreas Contaminadas, Desenvolvimento Urbano Sustentável e Revitalização de Brownfields) – Escola Politécnica, Universidade de São Paulo, São Paulo, 2022.

Nesta monografia, apresenta-se a revisão sistemática de literatura realizada para identificar tecnologias de remediação inovadoras para o tratamento de contaminações de PFAS em água e solo. Dentre 482 artigos únicos identificados nas bases de dados SCOPUS e Web of Science, 105 foram selecionados para análise estatística. Dentre estes, seis apresentaram aplicações em escala piloto e foram analisados em detalhes, quanto ao desempenho da tecnologia empregada, e também quanto às práticas e objetivos de sustentabilidade propostos pelo *Sustainable Remediation Forum* (SURF). Verificou-se que a técnica de tratamento de água por plasma apresentou os melhores resultados no tratamento de resíduos de investigação e também de água subterrânea, além de ter desempenho positivo quanto aos critérios de sustentabilidade avaliados. Cabe salientar que, embora este seja um campo que vem avançando rapidamente nos últimos cinco anos, mais pesquisas científicas são necessárias para que novas técnicas sejam desenvolvidas e alcancem a remediação em larga escala, uma vez que a grande maioria dos estudos ainda descrevem técnicas em escala de bancada.

Palavras-chave: Remediação do solo, Contaminação do solo, Águas subterrâneas, Sustentabilidade, Perfluoroalquil

ABSTRACT

Negrelli Garcia, Gabriel. **PFAS Remediation in Soil and Groundwater: A Systematic Literature Review of Innovative Technologies from a Sustainability Perspective**. 2022. 72 f. Monografia (MBA em Gestão de Áreas Contaminadas, Desenvolvimento Urbano Sustentável e Revitalização de Brownfields) – Escola Politécnica, Universidade de São Paulo, São Paulo, 2022.

This monograph presents a systematic literature review carried out to identify innovative remediation technologies for the treatment of PFAS contamination in water and soil. Among 482 unique articles identified in the SCOPUS and Web of Science databases, 105 were selected for statistical analysis. Among these, six presented applications on a pilot-scale and were analyzed in detail, regarding the performance of the technology used, and regarding the sustainability practices and objectives proposed by the Sustainable Remediation Forum (SURF). It was observed that plasma-based water treatment showed the best results in the treatment of investigation-derived waste and groundwater, in addition to having a positive performance in terms of the sustainability criteria evaluated. It should be noted that although this is a field that has been advancing rapidly in the last five years, more scientific research is needed so that new techniques are developed and reach full-scale remediation, since the vast majority of studies retrieved described bench-scale applications.

Palavras-chave: *Remediação do solo, Contaminação do Solo, Águas Subterrâneas, Sustentabilidade, Perfluoroalquil*

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1. INTRODUCTION

A group of substances called contaminants of emerging concern (CECs) are currently of interest to environmental science and engineering due to the potential harmful effects they may exert on the environment, its ecosystems, and, eventually, human health (LEE et al., 2021). Regulation for CECs is still in its infancy, in comparison to other pollutants such as organochlorines and heavy metals (LEE et al., 2021). The prevalence of these contaminants in multiple media, such as surface and groundwater, soil, air, sediment, and wastewater, is only magnified by their resistance to biological processes that may occur naturally, thus requiring human intervention for their removal (LEE et al., 2021).

In 2014, the United States Environmental Protection Agency (USEPA) listed poly- and perfluoroalkyl substances (PFAS) as emerging contaminants (XU et al., 2021). To date, over 4,000 different PFAS have been manufactured and used worldwide for their unique characteristics, which include resistance to heat, water, and oil (USEPA, 2019). These properties are what make PFAS useful for both consumer products, such as in food packaging, water repellent and stain-resistant fabrics, as well as for specialized applications such as in firefighting foams (USEPA, 2019). Such properties are also responsible for the persistence of PFAS in the environment and for their hard-to-predict behavior once environmental release occurs (USEPA, 2019). In addition, some PFAS are bio accumulative and toxic and there have been associations between PFAS and impacts to human health (USEPA, 2019).

Common PFAS sources include (ITRC, 2021a):

- Facilities where PFAS are or have been produced or processed, or facilities that use or have used PFAS chemicals to manufacture products or in other activities;
- Areas where fluorine-containing Class B firefighting foams are stored, used or released;
    - Class B firefighting foams are surfactant solutions used for fire suppression, fire training and flammable vapor suppression at military installations, airports, petroleum refineries, chemical manufacturing plants and storage facilities (NHDES, [s.d.]) . Class B foams can be either fluorinated or fluorine-free. Only fluorinated foams contain PFAS (NHDES, [s.d.]).
- Waste management facilities, such as landfills;
- Wastewater treatment residuals and areas of biosolids production and application.

The PFAS composition of each release will vary according to the products being manufactured or used at a given source (ITRC, 2021a). For example, wood fiber insulation has been shown to contain PFHpA (7 carbons) and other 5- to 8-carbon chain PFCAs (BEČANOVÁ et al., 2016). In the semiconductor industry, PFOS has been used in the fabrication of digital cameras, cell phones, printers and scanners (POULSEN; JENSEN; WALLSTRÖM, 2005). Semiconductor waste streams have been shown to contain PFBS, PFHxS, PFOS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA and PFDoA (LIN; PANCHANGAM; LO, 2009). For an explanation of PFAS nomenclature, please refer to Section 4.2.1.

Carbon chain length is a common way to group PFAS and the criteria to apply are described below (ITRC, 2021c):

- Long-chain refers to:
    - Perfluoroalkyl carboxylic acids (PFCAs) with 8 or more carbons (7 or more carbons are completely fluorinated);
    - Perfluoroalkane sulfonates (PFSAs) with 6 or more carbons (6 or more carbons are completely fluorinated);
- Short-chain refers to:
    - Perfluoroalkyl carboxylic acids (PFCAs) with 7 or fewer carbons (6 or fewer carbons are completely fluorinated);
    - Perfluoroalkane sulfonates (PFSAs) with 5 or fewer carbons (5 or fewer carbons are completely fluorinated).

While many of the long-chain PFAS have been phased out by their manufacturers due to their potential impacts to the environment and human health, short-chain PFAS are still being manufactured, some of which may suffer chemical transformations to produce long-chain PFAS (USEPA, 2019).

For such a ubiquitous problem, it is key that remediation techniques be developed or adapted to remove PFAS from the environment. In the last five years, a variety of technologies for PFAS remediation have been studied, including chemical oxidation, photocatalytic degradation, and sorption by nanomaterials (XU et al., 2021). The focus of this work will be on technologies that involve the destruction of PFAS rather than sorption or phase separation (such as foam fractionation, membrane separation, soil washing), as the latter generates further challenges associated with disposing of PFAS-laden sorption material and/or PFAS-rich wastewater. In addition, priority will be given to papers that present pilot-scale or full-scale applications. When selecting a remediation alternative, consideration must be given to sustainability to ensure that there is a net positive result once remediation targets are reached. For this reason, the remediation technologies will be evaluated according to a sustainability framework.

2. OBJECTIVE

The objective of this study was to identify innovative and sustainable remediation technologies for the destruction of PFAS contamination in soil and groundwater as described in the scientific literature.

2.1. Specific Objectives

- To carry out a systematic review of the scientific literature to identify references regarding the remediation of PFAS contaminated soil and groundwater focusing on the contaminants' elimination;
- To critically evaluate the identified technologies according to their application in pilot-scale and/or full-scale studies;
- To compare the technologies by applying the sustainability practices and objectives proposed by the Sustainable Remediation Forum (SURF).

3. JUSTIFICATION

Per- and polyfluoroalkyl substances (PFAS) are, currently, a topic of attention to the scientific community due to their unique properties and potential negative impacts to the environment and to human health. As a result, much research is currently being conducted worldwide to identify adequate technologies for PFAS remediation, the majority of which is in bench-scale. A preliminary literature search, however, did not identify a review of PFAS remediation technologies for soil and groundwater applied in pilot-scale or full-scale which included a critical analysis of sustainability. The existence of this project is then justified to fill this gap.

4. CONTEXTUALIZATION

4.1. What Are Emerging Pollutants?

According to the United States Environmental Protection Agency (USEPA), contaminants of emerging concern (CECs) including pharmaceuticals and personal care products (PPCPs) are increasingly being detected at low levels in surface water (USEPA, [s.d.]). While these products have been used for decades, much research is still necessary to determine the potential effects of CECs in the environment (USEPA, [s.d.]).

Many CECs and PPCPs act as endocrine disruptors (EDCs), meaning they may alter hormonal function in organisms and cause myriad health effects (USEPA, [s.d.]). For aquatic organisms, for example, EDCs may not be highly toxic, but even low levels of exposure may lead to reproductive effects (USEPA, [s.d.]). Furthermore, the effects of exposure at early stages of life may not be observed until adulthood (USEPA, [s.d.]).

CECs may be categorized according to source, chemical characteristics, fate in the environment and action mechanisms (SHAHID et al., 2021). Table 4.1 provides a classification proposed by Shahid et al. (2021).

Table 4.1 - Classification of CECs and their main sources

Category of CEC	Sub-category	Major Contaminants	Sources
Personal care products	Insect repellants, synthetic musk, sunblock agents and UV filters	Diethyltoluamide, 4-benzophenone, Galaxolide, Tonalide	Wastewater treatment plant effluent, landfill leachate and surface water
Pharmaceutically active complexes	Nonsteroidal anti-inflammatory medicines, antidepressant, antibiotics, anticonvulsants, lipid regulators, β-blocker, and hormones.	Diazepam, ciprofloxacin, metoprolol, diclofenac, carbamazepine, clorfibric acid, testosterone	Effluent of medicine manufacturing facility, hospitals and health centers, livestock farms, and domestic wastewater
EDCs	Bisphenol, xenohormone, and phthalates	Bisphenol A, xenoestrogen, and dioctyl phthalate	Drinking water, surface water, sediments, soil and secondary sludge
Regulated Compounds	Pesticides and poly aromatic hydrocarbons	Chlorpyrifos and phenanthrene	Agricultural runoffs, sewage treatment plants, sediments, soil and surface water
Biocides	Herbicides, fungicides, and molluscicide	Metaldehyde, butachlor, and epoxiconazole	Surface water, aquafarming, and agricultural runoff
Industrial Chemicals	Plasticizers and fire retardants	Dimethyl adipate and Tris (1-chloro-2-propyl) phosphate	Domestic and industrial wastewater
Per- and polyfluoroalkyl substances (PFAS)	Perfluorocarboxylic acids (PFCAs) and perfluorosulfonic Acids (PFSAs)	Perfluorooctanoic acid (PFOA) and Perfluorooctanesulfonate (PFOS)	Sediments, groundwater, surface water, and wastewater
Surfactants	Ionic and nonionic surfactants	Tweens (Polysorbates) and sodium lauryl sulfate	Domestic and industrial wastewater

Source: reproduced from Shahid et al. (2021)

4.2. Per- and polyfluoroalkyl substances (PFAS)

According to Panieri et al. (2022), a universally accepted definition of PFAS does not yet exist. The first classification was proposed by Buck et al. (2011), which defined PFAS as “the highly fluorinated aliphatic substances that contain 1 or more C atoms on which all the H substituents have been replaced by F atoms, in such a manner that they contain the perfluoroalkyl moiety CnF2n+1–”. In 2021, the Organisation for Economic Co-Operation and Development (OECD) revisited the definition provided by Buck et at. (2021) to include compounds that did not fit into the previous definition. The new definition, proposed by the OECD (2021), reads

> PFASs are defined as fluorinated substances that contain at least one fully fluorinated methyl or methylene carbon atom (without any H/Cl/Br/I atom attached to it), i.e. with a few noted exceptions, any chemical with at least a perfluorinated methyl group (–CF_3) or a perfluorinated methylene group (–CF_2–) is a PFAS.

In practical terms, the USEPA defines per- and polyfluoroalkyl substances (PFAS) as a group of man-made chemicals that includes perfluorooctanoic acid (PFOA), perfluorooctanesulfonate (PFOS), GenX and several other substances (USEPA, [s.d.]). Estimated in over 4000 compounds, PFAS have been manufactured and used for several decades worldwide, starting in the 1940s in the United States (USEPA, [s.d.]). PFAS have several uses and are present in many products in daily life, such as in food-wrappers, household products (including stain- and water-repellent fabrics, non-stick products) and fire-fighting foams (which is a very important source of contamination where firefighting training occurs) (USEPA, [s.d.]). Their widespread use is frequently associated with their desirable physical-chemical properties, which include high stability (both chemical and thermal), as well as hydrophobic and lipophobic characteristics, which are associated with the perfluoroalkyl moiety (BUCK et al., 2011). As will be discussed in Section 4.2.3, these same properties pose serious challenges for PFAS remediation.

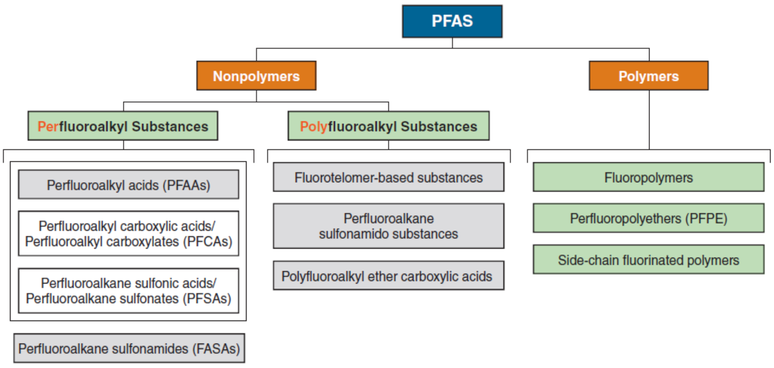
PFAS have already been detected in the environment, wildlife, and humans. In 2001, Giesy and Kannan first reported detections of perfluorooctane sulfonic acid, $C_8F_{17}SO_3H$ (PFOS), in wildlife on a global scale. Examples included detections in blood samples from ringed and grey seals from the Canadian and Norwegian Arctic in the range of 3 to 50 ng/L, and concentrations 2 to 10 times greater in seals closer to urban areas, such as in the Baltic Sea (GIESY; KANNAN, 2001). Also, Hansen et al. (2001) detected PFAS in human blood samples from biological supply companies. The potential human health effects of these detections will be discussed in Section 4.2.3.

4.2.1. Naming Convention

Due to their vast numbers and high level of complexity, PFAS merit their own naming convention to avoid confusion with non-specific acronyms, such as perfluorinated compounds (PFCs) (ITRC, 2021c).

As shown in Figure 4.1, PFAS may be organized in two large groups: polymers and nonpolymers.

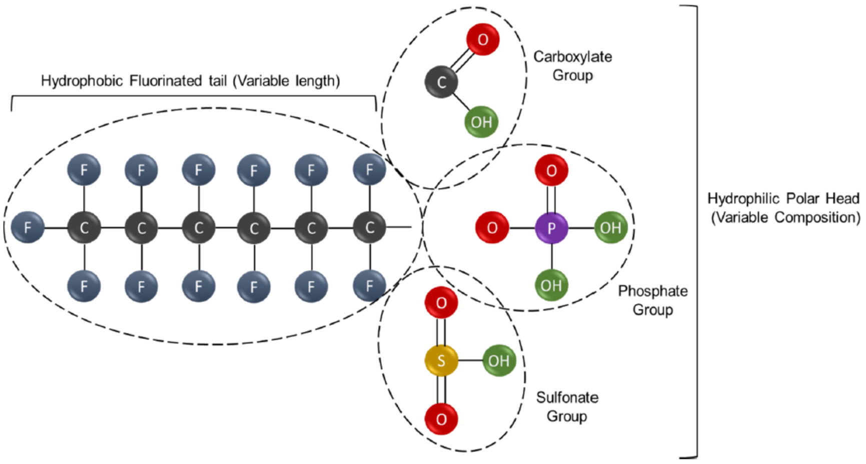
Figure 4.1 - PFAS groups



Source: (ITRC, 2021c)

Nonpolymeric PFAS appear to be more prevalent in the environment, including humans and biota, and are included in most laboratory PFAS analyte lists (ITRC, 2021). Figure 4.2 shows the general structure of nonpolymeric PFAS, which consists of a hydrophobic tail of varying length of carbon atoms at different degrees of fluorination, and a hydrophilic head, which contains a functional polar group – the most common of which are in Figure 4.2 (PANIERI et al., 2022).

Figure 4.2 - General structure of nonpolymeric PFAS



Source: prepared by Panieri et al. (2022)

Within the group of nonpolymeric PFAS, perfluoroalkyl acids (PFAAs) are the main analytes in commercial laboratory target lists and are also the focus of policymaking. They are generally referred to as PFXY (ITRC, 2021), where:

- PF stands for perfluoroalkyl;
- X denotes the carbon chain length, according to the naming convention used for hydrocarbons based on the number of carbon atoms;
- Y denotes the functional group.

Table 4.2 presents the naming structure for the most common PFAAs. In it, the protonated form of each PFAA has been omitted for brevity. In the protonated form (acid), the "-oate" suffix is replaced by "-oic acid", e.g. "Perfluorobutanoate" becomes "Perfluorobutanoic acid".

Table 4.2 - Naming structure for PFAAs

X	Acronym	Name	Formula
B = buta (4 carbon)	PFBA	Perfluorobutanoate	$C_3F_7CO_2^-$
	PFBS	Perfluorobutane sulfonate	$C_4F_9SO_3^-$
Pe = penta (5 carbon)	PFPeA	Perfluoropentanoate	$C_4F_9CO_2^-$
	PFPeS	Perfluoropentane sulfonate	$C_5F_{11}SO_3^-$
Hx = hexa (6 carbon)	PFHxA	Perfluorohexanoate	$C_5F_{11}CO_2^-$
	PFHxS	Perfluorohexane sulfonate	$C_6F_{13}SO_3^-$
Hp = hepta (7 carbon)	PFHpA	Perfluoroheptanoate	$C_6F_{13}CO_2^-$
	PFHpS	Perfluoroheptane sulfonate	$C_7F_{15}SO_3^-$
O = octa (8 carbon)	PFOA	Perfluorooctanoate	$C_7F_{15}CO_2^-$
	PFOS	Perfluorooctane sulfonate	$C_8F_{17}SO_3^-$
N = nona (9 carbon)	PFNA	Perfluorononanoate	$C_8F_{17}CO_2^-$
	PFNS	Perfluorononane sulfonate	$C_9F_{19}SO_3^-$
D = deca (10 carbon)	PFDA	Perfluorodecanoate	$C_9F_{19}CO_2^-$
	PFDS	Perfluorodecane sulfonate	$C_{10}F_{21}SO_3^-$
Un = undeca (11 carbon)	PFUnA or PFUnDA	Perfluoroundecanoate	$C_{10}F_{21}CO_2^-$
	PFUnS or PFUnDS	Perfluoroundecane sulfonate	$C_{11}F_{23}SO_3^-$
DoD = dodeca (12 carbon)	PFDoDA	Perfluorododecanoate	$C_{11}F_{23}CO_2^-$
	PFDoDS	Perfluorododecane sulfonate	$C_{12}F_{25}SO_3^-$
TrD = trideca (13 carbon)	PFTrDA	Perfluorotridecanoate	$C_{12}F_{25}CO_2^-$
	PFTrDS	Perfluorotridecane sulfonate	$C_{13}F_{27}SO_3^-$
TeD = tetradeca (14 carbon)	PFTeDA	Perfluorotetradecanoate	$C_{13}F_{27}CO_2^-$
	PFTeDS	Perfluorotetradecane sulfonate	$C_{14}F_{29}SO_3^-$

Source: adapted from ITRC (2021)

4.2.2. Physical and Chemical Properties of PFAS

PFAS is a group of compounds with largely different physical and chemical properties (ITRC, 2021c). They may occur in nature in different ionic states, including protonated (acids), negatively charged (anions), positively charged (cations) and both positively and negatively charged dipolar molecules (zwitterions) (ITRC, 2021c). These ionic states determine their charges and, in turn, their physicochemical properties and fates in the environment (ITRC, 2021c).

The availability of physical and chemical properties of PFAS varies greatly within the group (ITRC, 2020). Reliable physical and chemical properties of PFAS are scarce and some of the properties are estimated mathematically based on the chemical structure of the compound using Quantitative Structure-Activity Relationship (QSAR) models (ITRC, 2020).

4.2.3. Environmental Fate and Toxicology of PFAS

The occurrence of PFAS in the environment is directly associated with industrial use or production and incidental environmental release, as well as from use and disposal of products that may contain them in residual amounts (BUCK et al., 2011). Additionally, certain functional derivatives and polymers that contain a perfluoroalkyl moiety may be degraded biotically or abiotically in the environment to form PFOS, PFOA and other PFAS (BUCK et al., 2011).

As stated in Section 4.2.1, the most common PFAS in the environment – i.e. nonpolymeric PFAS – have a C-F tail and a nonfluorinated head containing a polar functional group (ITRC, [s.d.]). The tail is hydrophobic and the head, given its polar nature, is hydrophilic (ITRC, [s.d.]). The different behaviors of the head and tail affect the distribution of nonpolymeric PFAS in the environment depending also on the characteristics of the receiving soil (ITRC, [s.d.]). For instance, the hydrophobic properties of the tail favor association with the organic carbon in soil (ITRC, [s.d.]). The polar head, on the other hand, interacts electrostatically with natural charges in the soil and aquifer material (ITRC, [s.d.]). The net natural charges tend to be negative and can repel negatively charged heads of PFAAs which are present as anions in the environment (ITRC, [s.d.]).

PFAS have been extensively documented in aquatic biota and wildlife in the last 20 years, as evidenced by a literature review prepared by Burkhard (2021), which consulted reviews prepared by Giesy and Kannan (2001), Houde et al. (2006), Houde et al. (2011), as well as publications by Labadie and Chevreuil (2011), Wang et al. (2011), Hloušková et al. (2013), Lorenzo et al. (2016), Babut et al. (2017), Casal et al. (2017), Essumang et al. (2017) and Munoz et al. (2017).

With regard to the potential to bioaccumulate, Burkhard (2021) establishes a link between bioaccumulation potential increase and increasing chain length of PFCAs and references studies by McCarthy, Kappleman and DiGuiseppi (2017), Ahrens and Bundschuh (2014), and Giesy et al. (2010). In addition, Burkhard (2021) highlights that perfluorooctane sulfonic acid (PFOS), perfluorononaoic acid (PFNA), perfluorodecanoic acid (PFDA) and longer-chained PFCAs biomagnify in aquatic food webs by referencing studies by Martin et al. (2004), Houde et al. (2008) and Loi et al. (2011). According to McCarthy, Kappleman and DiGuiseppi (2017), PFOS tends to accumulate in protein-rich tissues and organs, such as plasma, blood and the liver, unlike non-ionic organic compounds, that preferably partition to lipids.

According to Burkhard (2021), the bioaccumulation models for legacy chemicals (e.g. PCBs, DDTs, PCDD/Fs) are not applicable to PFAS as the underlying accumulation processes are different – they estimate the need for a decade or two of scientific research for adequate models to be developed. In a review published in 2021, Burkhard identified that most bioaccumulation factors (BAFs) available in literature are for carbonyl (PFCA) and sulfonyl (PFSA) compounds, whereas data for alcohols, ethers, esters and phosphorus PFAS are limited. In addition, while Burkhard did not examine mixtures of PFAS in the environment, the author stated that precursors present in such mixtures may bias measured BAFs, so that “better knowledge on precursors and their biotransformation is a research need” (Burkhard, 2021).

For human beings, Panieri et al. (2022) lists inhalation of air and dust particulate, ingestion of contaminated food and drinking water, as well as dermal adsorption as some of the most relevant routes of exposure to PFAS. Biomonitoring of PFAS in humans is generally performed through sampling and analysis of blood serum and blood plasma (SCHRENK et al., 2020). According to Schrenk et al. (2020), PFOA, PFNA, PFHxS and PFOS contribute most to the PFAS levels observed in human serum. Human epidemiology data associate PFOA exposure with high cholesterol, increases to liver enzymes, weaker vaccination response, testicular and kidney cancer, as well as pregnancy-induced hypertension and preeclampsia (EPA, 2020b).

4.2.4. PFAS Sampling and Analysis

PFAS generally occur at very low concentrations in the environment, which can be technologically challenging to detect (MUELLER; YINGLING, 2020). To further complicate matters, many materials used in environmental investigation and sampling may potentially contain PFAS, which can act as sources of cross-contamination (MUELLER; YINGLING, 2020). For this reason, while sampling for PFAS is similar to other compounds, several considerations and protocols must be followed (MUELLER; YINGLING, 2020).

According to Mueller and Yingling (2020), with regard to sampling materials, those listed below could potentially introduce sampling biases and must be avoided (MUELLER; YINGLING, 2020):

- Polytetrafluoroethylene (PTFE);
- Waterproof coatings containing PFAS;
- Fluorinated ethylene-propylene (FEP);
- Ethylene tetrafluoroethylene (ETFE);
- Low-density polyethylene (LDPE);
- Polyvinylidene fluoride (PVDF);
- Pipe thread compounds and tape.

In addition, since PFAS have been historically used in consumer and industrial products, materials such as fast-food wrappers and waterproof fabrics must not be kept near PFAS sampling materials (MUELLER; YINGLING, 2020).

To be able to assess the quality of collected field samples, a good strategy is to collect quality control samples. In the field, these include field reagent blanks, source water blanks, equipment rinse blanks, as well as field duplicate samples (ITRC, 2021b). In addition to the field blanks, laboratory blanks may also be utilized, and these include method blank, laboratory reagent blank and instrument blank (ITRC, 2021b).

By preparing and analyzing a set of field and laboratory blanks, one may evaluate if and where sample contamination has occurred (ITRC, 2021b). According to the United States Department of Defense (DoD) and Department of Energy (DoE) (2019), if a sample contains a contaminant within 5 to 10 times the concentration in the associated blank, then the results might be biased.

Regarding sample analysis, the detection method most widely used is liquid chromatography-mass spectrometry-mass spectrometry (LC/MS/MS), which is especially suited for analysis of ionic compounds, such as the PFSAs and PFCAs (MUELLER; YINGLING, 2020). For the neutral and non-ionic analytes, gas chromatography-mass spectrometry (GC/MS) can also be used (MUELLER; YINGLING, 2020). Currently, LC/MS/MS analysis of PFAS is widely available, whereas GC/MS analysis has limited commercial availability (MUELLER; YINGLING, 2020).

4.3. Sustainable Remediation Practices

The remediation industry was created in the second half of the 20th century in response to public pressure motivated by discoveries of toxic chemicals in landfills, drinking water and other media. Since then, a global effort ensued in the identification of contaminated sites, the development of remediation techniques, as well as in the creation of legislation to guide and define goals for decontamination efforts (ELLIS; HADLEY, 2009).

In 2006, a group of remediation professionals united forces to form the Sustainable Remediation Forum (SURF) with the mission to:

> *Establish a framework that incorporates sustainable concepts throughout the remedial action process while continuing to provide long-term protection of human health and the environment and achieving public and regulatory acceptance* (ELLIS; HADLEY, 2009).

SURF defines sustainable remediation as the combination of remedies that lead to a net benefit on human health and the environment, all the while using limited resources. Their approach includes (ELLIS; HADLEY, 2009):

- Minimizing or eliminating energy consumption or the consumption of other natural resources;
- Reducing or eliminating releases to the environment, especially to the air;
- Harnessing or mimicking natural processes;
- Reusing or recycling land or otherwise undesirable materials; and/or
- Encouraging the use of remedial technologies that permanently destroy contaminants.

In planning, SURF proposes a “triple bottom line” approach, considering social, environmental and economic drivers. The principles behind each component are presented below (ELLIS; HADLEY, 2009):

- Social
    - Industry desire to improve corporate image and enhance social responsibility, thus improving shareholder value, reducing risk and improving communities;
    - Pressure from nongovernmental agencies;
    - Public awareness of sustainability issues and requests for more sustainable practices;
- Environment
    - Existing and recommended climate-change legislation at different jurisdictional levels (federal, state, municipal);
    - Environmental regulations that establish minimum environmental protection requirements;
    - Environmental enforcement agencies;
    - Net environmental benefit focus;
- Economic
    - Brownfield development incentives and growing real estate values;
    - Long-term environmental liability management.

5. METHODOLOGY

The present work was developed using a systematic literature review (SLR) approach with the aid of the StArt (State of the Art through Systematic Review) software, created by the *Laboratório de Pesquisa em Engenharia de Software* (LaPES) from the Federal University of São Carlos (UFSCar).

The information used in this study was gathered in a two-stage process. The first stage involved retrieving scientific material on the properties of PFAS, their environmental fate and transport, as well as impacts to the environment and human health from online documents published by the United States Environmental Protection Agency (USEPA) and the Interstate Technology and Regulatory Council (ITRC). The second stage, which was the retrieval of published studies, involved queries in SCOPUS and Web of Science databases with the strings provided in Table 5.1

Table 5.1 - Search strings queried in the databases

Database	Search String
Web of Science	("groundwater" OR "água subterrânea" OR "ground water" OR "soil" OR "solo") AND ("remediation" OR "remediação" OR "treatment" OR "tratamento") AND ("pfas" OR "Perfluoroalkyl" OR "Polyfluoroalkyl") (All fields)
	("groundwater" OR "água subterrânea" OR "ground water" OR "soil" OR "solo") AND ("remediation" OR "remediação" OR "treatment" OR "tratamento") AND ("pfas" OR "Perfluoroalkyl" OR "Polyfluoroalkyl" OR "Perfluoro" OR "Polyfluoro")
Scopus	TITLE-ABS-KEY (("groundwater" OR "água subterrânea" OR "ground water" OR "soil" OR "solo") AND ("remediation" OR "remediação" OR "treatment" OR "tratamento") AND ("pfas" OR "Perfluoroalkyl" OR "Polyfluoroalkyl"))
	TITLE-ABS-KEY (("groundwater" OR "água subterrânea" OR "ground water" OR "soil" OR "solo") AND ("remediation" OR "remediação" OR "treatment" OR "tratamento") AND ("pfas" OR "Perfluoroalkyl" OR "Polyfluoroalkyl" OR "Perfluoro" OR "Polyfluoro"))

Source: prepared by the author

From the four searches presented in Table 5.1, 661 papers were found. Of these, 179 were duplicates and the remaining 482 were selected according to the following inclusion/exclusion criteria:

- Inclusion:
    - The article provided information on in-situ or ex-situ treatment of soil, groundwater, or surface water contaminated by PFAS;
    - The article described a case study (bench-scale, pilot-scale or full-scale) and provided results of the application of the remediation technology.

- Exclusion:
    - The article did not provide information on in-situ or ex-situ treatment of soil, groundwater, or surface water contaminated by PFAS;
    - The article did not describe the application of the remediation technology;
    - The article was a review of literature – only individual articles were considered in this SLR;
    - The article was not available in *Sistema Integrado de Bibliotecas – Universidade de São Paulo* (SIBi USP) and could not be accessed by the author.

Considering the above criteria, 105 papers were included in the SLR. They were then categorized according to Table 5.2:

Table 5.2 - Categories and sub-categories utilized in the SLR

Category	Sub-category
Target medium	Soil, Wastewater, Groundwater, Surface water
In-situ or Ex-situ	-
Type of technology	Sorption (Biochar, Granular Activated Carbon, Immobilization, Ion-Exchange, Metal-Organic Framework, Adsorption (other technologies)) Phase Separation (Membrane Separation, Foam Fractionation, Soil Washing) Electrochemical (Electrooxidation, Electrodialysis, Electrocoagulation, Solvated Electrons) Photodegradation, Chemical Oxidation, Plasma Treatment, Thermal Treatment, Enzymatic Degradation, Chemical Reduction, Combination of Technologies, Ozonation, Phytoremediation, Fungal Bioreactor, Sonochemical Oxidation, Monitored Natural Attenuation, Mechanochemical Treatment
Year of publication	-
Scale of study	Bench-scale, Pilot-scale, Full-scale

Source: prepared by the author

As previously defined, the technology groups “Sorption” and “Phase Separation” were excluded from the SLR, as they do not promote contaminant destruction. After this exclusion, 53 papers were considered in a descriptive statistics evaluation of the available literature, as presented in Section 6.1.2.

Of the 53 papers, six presented pilot-scale applications, which were then discussed in Sections 6.1.2.1 (ex-situ applications) and 6.1.2.2 (in-situ applications). They were also evaluated according to the sustainability practices and objectives proposed by SURF in Section 6.1.2.3.

6. RESULTS AND DISCUSSION

6.1. Systematic Literature Review

6.1.1. Summary of Existing PFAS Remediation Techniques

As shown in Table 5.2, a variety of technologies have been proposed and tested for PFAS treatment. This section will provide brief descriptions of those technologies.

6.1.1.1. Sorption

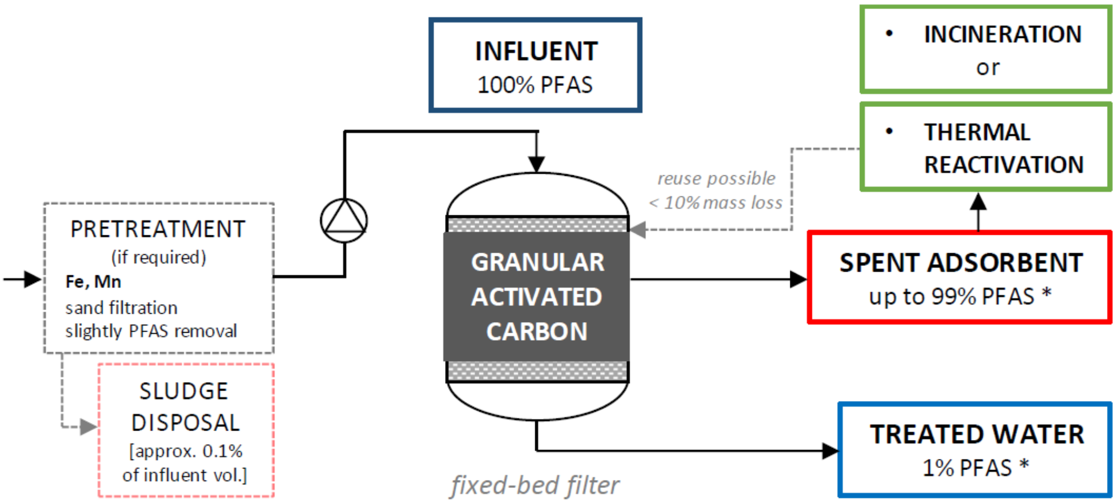
Sorption technologies utilize adsorption or absorption processes to separate PFAS from other media. They are not destructive technologies, such that an additional step (such as incineration) must be included to degrade the contaminants. Currently, these are the most used technologies worldwide (LIU; STRATHMANN; BELLONA, 2021).

As reported by Riegel, Egner and Sacher (2020), the most common application of sorbents is in flow-through fixed-bed filtration column where the influent stream is contaminated and the PFAS adsorb to the sorbent material, resulting in a less contaminated effluent. It is common for two or more columns to be installed in series, such that when the first column becomes saturated, the second column may be switched to be the lead column while the saturated column is loaded with fresh adsorbent material (RIEGEL; EGNER; SACHER, 2020).

In addition to the main treatment step, a pre-treatment step may be required to remove constituents that may compete for the adsorptive material, such as suspended solids, organic matter, iron and manganese (RIEGEL; EGNER; SACHER, 2020).

A common sorption material for field applications is granular activated carbon (GAC). Figure 6.1 shows the flow diagram proposed by Riegel, Egner and Sacher (2020), which is typical for PFAS treatment with activated carbon.

Figure 6.1 - Typical flow diagram for a PFAS treatment system using activated carbon



* 1 % of PFAS in the filter effluent seems to present a typical value for a PFAS mixture of different substances with differing absorbance properties.

Source: prepared by Riegel, Egner and Sacher (2020)

Though removal efficiencies vary strongly depending on the type of activated carbon used, the USEPA’s Health Advisory Level (HAL) of 70 ng/L for PFOA and PFOS (combined and individually) may be achieved by activated carbon treatment, although several beds in series may be required (RIEGEL; EGNER; SACHER, 2020).

While GAC is the most common material, Riegel, Egner and Sacher (2020) have identified commercial alternatives, which are listed below:

- MatCare, which consists of an organoclay mineral called palygorskite which is chemically altered with aliphatic amines, granting it a hydrophobic surface character. It is available in granular medium.
- RemBind, which is composed of amorphous aluminum hydroxide, activated carbon, organic matter and kaolinite. It is available as a fine powder, comparable to powdered activated carbon.

Another example of sorption technology is Ion Exchangers. They exploit the fact that dissolved PFAS exist as negatively charged molecules at common environmental pH values, such that they may be adsorbed by anion exchangers (RIEGEL; EGNER; SACHER, 2020). Ion exchangers can be “single-use” or may be regenerated – in the case of regeneration, the liquid regenerate is highly concentrated in PFAS and must be treated for the destruction of the contaminants (RIEGEL; EGNER; SACHER, 2020).

Anion exchangers have been shown to work at both high (hundreds of mg/L) and low (ng/L and ug/L range) concentrations of PFAS (RIEGEL; EGNER; SACHER, 2020).

For both activated carbon and ion exchangers, the affinity of per- and polyfluoroalkyl sulfonates (PFSA) is higher than that of per- and polyfluoroalkyl carboxylates (PFCA) (RIEGEL; EGNER; SACHER, 2020). Additionally, long-chain PFAS tend to adsorb better than short-chain PFAS (RIEGEL; EGNER; SACHER, 2020).

An additional option is immobilization using activated carbon, biocharcoal, modified clay or a sorbent mixture. In this technology, the sorbent is applied in-situ, with the goal of immobilizing the PFAS and preventing leaching (HØISÆTER et al., 2021) – although it is possible that leaching may occur over extended periods of time.

6.1.1.2. Phase Separation

In this category, the author included membrane separation, soil washing and foam fractionation.

Membrane separation works by steric (based on size) exclusion and/or electrostatic interactions (BOO et al., 2018). Most membrane studies have focused on a small set of PFAAs, most notably PFOA and PFOS. While certainly important, these PFAAs do not represent the complete set of PFAS present in important sources such as AFFF. High-pressure membranes may be appropriate for shorter-chain PFAS, where sorption (such as with GAC) generally performs poorly (LIU; STRATHMANN; BELLONA, 2021).

Soil washing involves the application of water with or without other solvents or surfactants to wash PFAS from the soil (HØISÆTER et al., 2021). While it is commonly done ex-situ, in-situ soil washing is possible and has the advantage of preventing excavation and transportation of PFAS contaminated soil, although it is paramount that all the leachate be collected to prevent further spreading of the PFAS contamination (HØISÆTER et al., 2021).

Foam fractionation explores the amphiphilic nature of PFAS, such as PFOS and PFOA. It involves the bubbling of a gas through an aqueous solution containing amphiphiles (i.e. molecules with hydrophilic and lipophilic regions), which adsorb onto the surface of the gas bubbles and form a layer of foam above the liquid that can be removed. This layer contains high concentrations of the amphiphile, and the residual liquid is clean (BURNS; STEVENSON; MURPHY, 2021).

6.1.1.3. Plasma-based water treatment (PWT)

Plasma-based water treatment (PWT) has drawn the attention of researchers for water treatment, in particular non-thermal plasmas, which does not consume much energy and requires simple equipment for implementation. This technology can produce highly reactive species such as H•, O and HO• radicals, free electrons and ozone, as well as oxidants, in addition to ultraviolet light (due to the emission of light by the plasma discharge), shockwaves which also carry energy, and high-density electric fields (PALMA et al., 2021).

6.1.1.4. Electron beam technology (eBeam)

Electron Beam Technology (eBeam) utilizes electron accelerators to produce highly energetic electrons in large quantities, which can cause direct damage to chemical bonds and lead to the degradation of contaminants. At high doses, eBeam can also cause thermal decomposition. It can be categorized as an Advanced Oxidation-Reduction Process (AORP) (LASSALLE et al., 2021).

6.1.1.5. Enzymatic degradation

Enzymatic Degradation involves the use of enzymes to degrade PFAS. It has been shown that PFOA can be degraded in water by enzyme catalyzed oxidative humification reactions (ECOHRs), which are reactions naturally occurring in humification processes to convert biopolymers into humic substances. These reactions are promoted by enzymes such as laccases and peroxidases, which are naturally produced by bacteria and fungi (LUO; LIANG; HUANG, 2018). Laccase is suitable for in-situ remediation as it can maintain its activity for long periods of time (LUO et al., 2015).

6.1.1.6. Photodegradation

Photodegradation processes apply a radiation source (such as a UV lamp) to destroy contaminants via different mechanisms.

Conventional advanced oxidation processes which combine UV radiation and ozone (UV-O_3) or UV radiation and hydrogen peroxide (UV-H_2O_2) have been shown ineffective for treating PFAAs (TENORIO et al., 2020).

Alternatively, heterogeneous photocatalysis using composite titanium dioxide (TiO_2) catalysts (such as TiO_2 and reduced graphene oxide or Pb-modified TiO_2) or bismuth phosphate photocatalysts under UV irradiation have been shown to degrade PFOA (TENORIO et al., 2020).

For PFSAs and PFCAs, UV photochemical processes that generate hydrated electrons (e_{aq}^-) have proven to be effective. These hydrated electrons are strong reducers that can be generated via UV excitation of sensitizers, such as iodide, 3-indole-acetic acid or sulfite (TENORIO et al., 2020).

6.1.1.7. Chemical Oxidation

Chemical Oxidation involves the application of an oxidant, such as persulfate, to directly oxidize target compounds, or to produce reactive species that in turn attack the target compounds (SHOJAEI et al., 2021). Acidic ozonation has also been shown to create highly oxidative conditions and has been studied as a pre-treatment step in alkaline ozonation (THOMAS et al., 2020).

6.1.1.8. Chemical Reduction

Chemical Reduction involves the utilization of a chemical reductant such as zero-valent metals (ZVMs) to degrade PFAS. Different ZVMs (Al, Cu, Zn and Fe) have been shown to degrade PFOS (ZENOBIO et al., 2020)

6.1.1.9. Electrochemical Processes

Electrocoagulation (EC) involves utilizing a sacrificial anode that provides charged cations to solution (such as Zn^{2+}, Al^{3+}, Fe^{3+}), which aggregate to form hydroxyl complex species. These hydroxyl complexes can strongly sorb certain pollutants and remove from the contaminated water (SHI et al., 2021). It has been shown by Lin et al. (2015) that PFAS can be removed by adsorbing on zinc hydroxide flocs generated in-situ during EC with a zinc anode.

Electrooxidation (EO) is a form of advanced oxidation process (AOP) that leads to contaminant degradation via direct electron transfer (DET) at the anode as well as oxidation by hydroxyl radicals (•OH) produced by oxidation of water molecules.

Electrodialysis or electrokinetic extraction involves applying an electric field to a portion of soil or water. This leads to a separation of charges, with cations migrating toward the cathode, and anions, to the anode. This technique has been used to remove pollutants such as heavy metals, as well as neutral, cationic and anionic organic contaminants. Since PFAS exist in charged states, they may be removed by applying this technique (SÖRENGÅRD et al., 2019).

6.1.1.10. Fungal Bioreactor

Fungal bioreactors utilize culturable fungi to degrade contaminants in the controlled environment of a bioreactor. Fungi play an important environmental role as they compose up to 75% of soil microbial biomass and, as such, may be useful in degrading contaminants such as PFAS (MERINO et al., 2018).

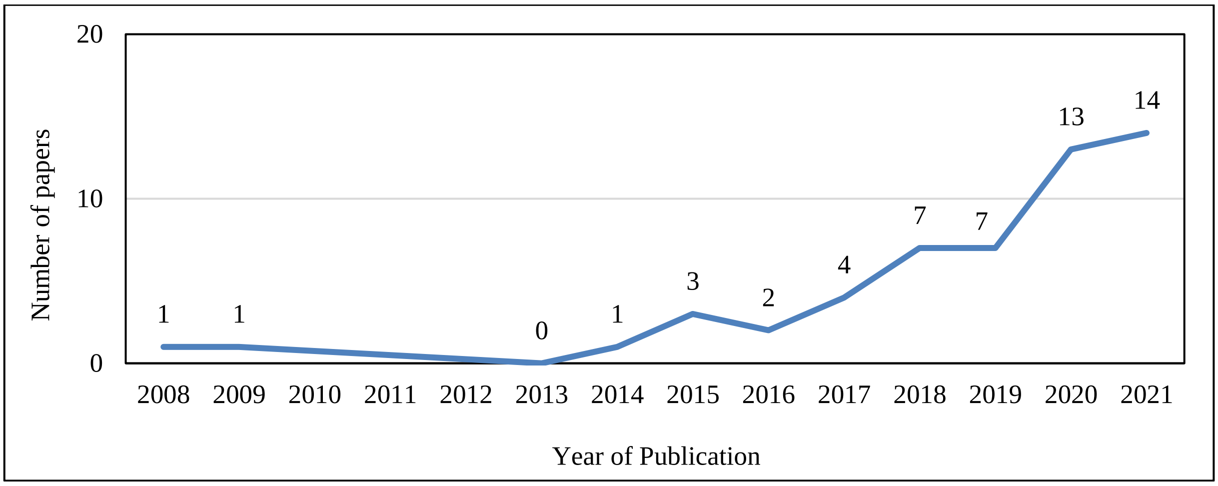
6.1.1.11. Phytoremediation

Phytoremediation involves the utilization of plants to remediate shallow groundwater and/or soil contamination. In particular, phytoaccumulation is the process through which a plant removes contaminants from the environment and accumulates them in both root and above-ground tissue. In the case of tissue (such as leaves), removal and subsequent disposal or treatment is possible (HUFF et al., 2020).

6.1.2. Critical Evaluation of PFAS Remediation Techniques

As evidenced by Figure 6.2, the number of papers on PFAS remediation has increased rapidly starting in 2014.

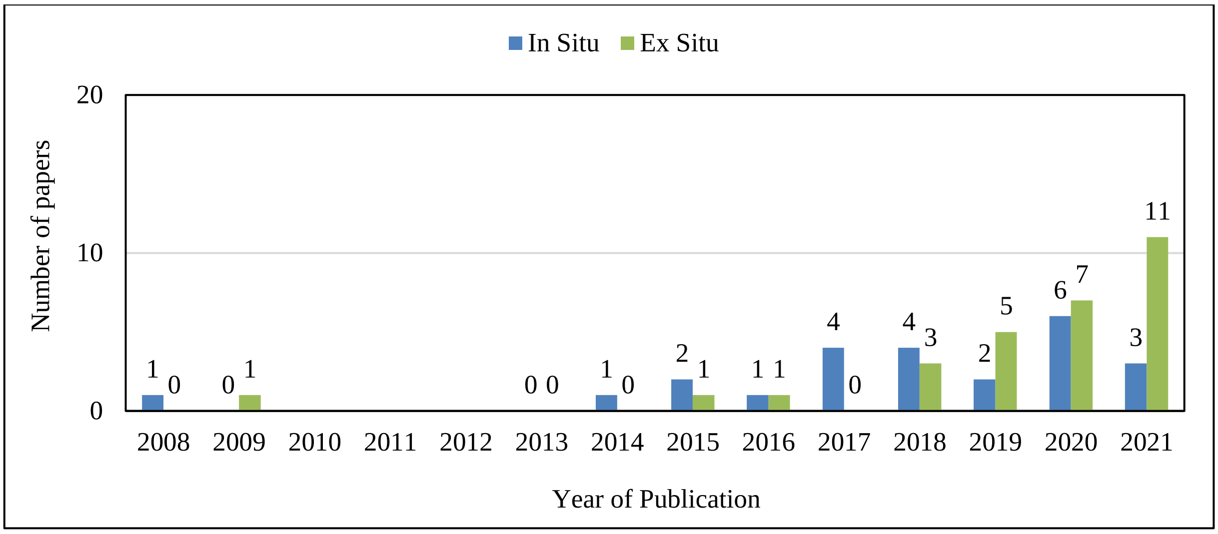
Figure 6.2 - Number of papers per year of publication



Source: prepared by the author

Furthermore, Figure 6.3 shows that most papers published in the last three years favored ex-situ remediation technologies, which may be due to this type of technology being more easily engineered and controlled, as it avoids challenges that result from subsurface heterogeneities (REDDY, 2008).

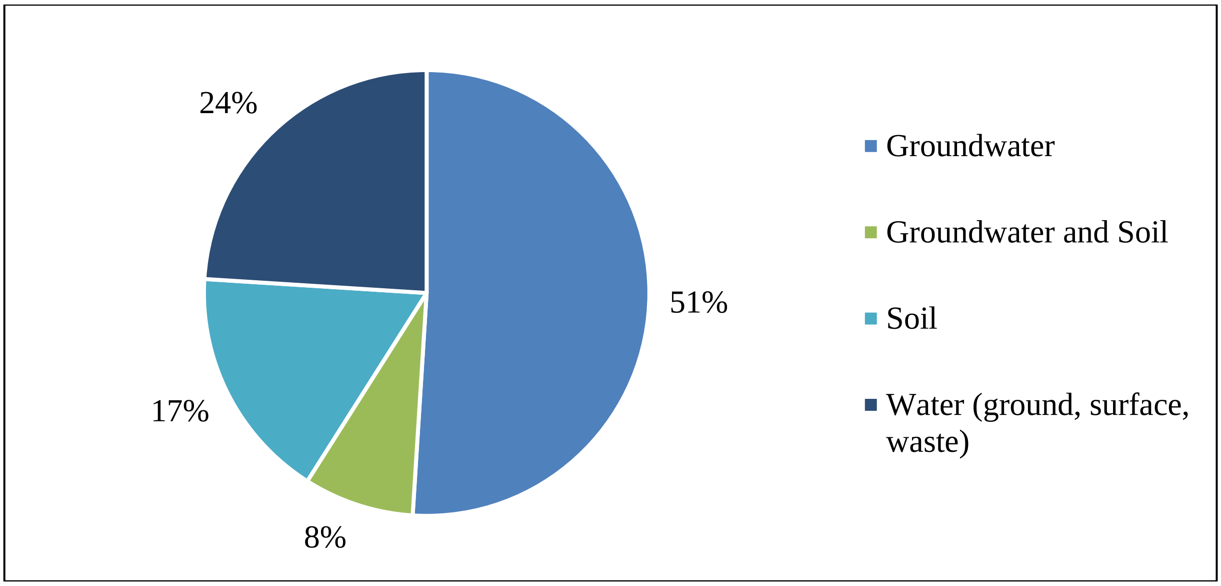
Figure 6.3 - Number of papers per type (in-situ or ex-situ) per year of publication



Source: prepared by the author

Figure 6.4 shows that most papers focus on the remediation of groundwater (51%) and water (ground, surface or wastewater) (24%).

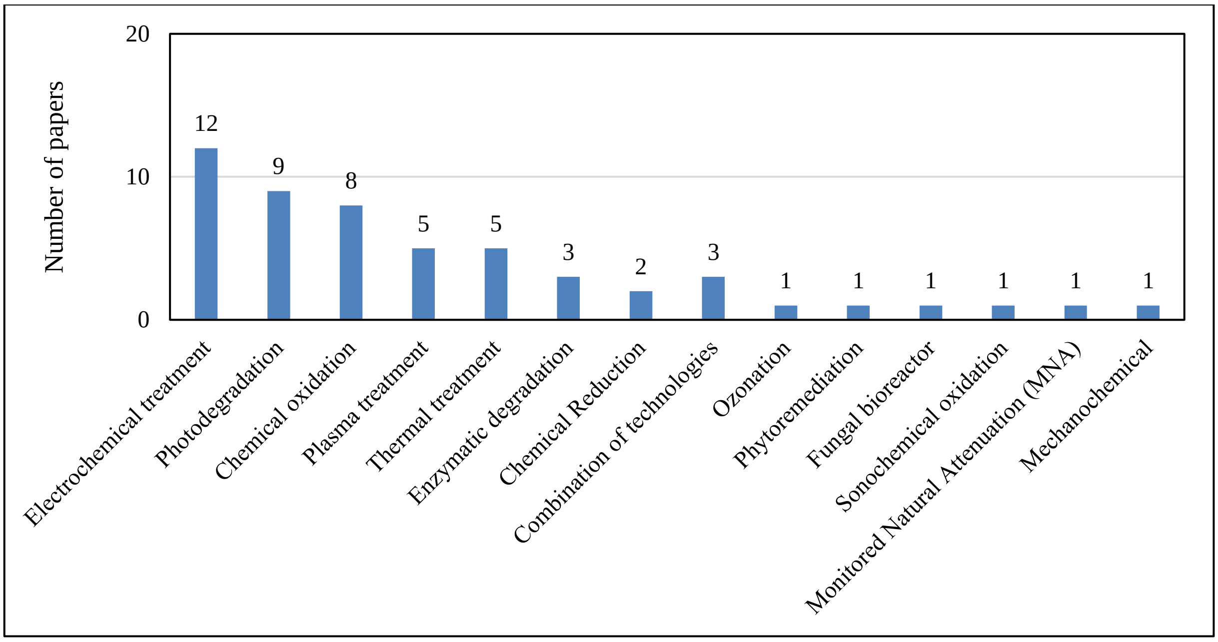
Figure 6.4 - Percent of articles per medium



Source: prepared by the author

Figure 6.5 shows the number of papers selected per remediation type.

Figure 6.5 - Number of papers per remediation type



Source: prepared by the author

Regarding the scale of the study, 47 papers present bench-scale results and six present pilot-scale results.

From the six pilot-scale studies:

- 2 utilized a combination of technologies;
- 2 utilized plasma treatment;
- 1 utilized chemical oxidation;
- 1 utilized phytoremediation.

The relatively small number of pilot-scale studies and restricted scope of technologies in comparison to bench-scale studies shows that PFAS remediation will still require much scientific attention in the coming years for larger scale projects to be implemented. The six pilot-scale studies that were selected in the SLR will now be further analyzed.

6.1.2.1. Ex-Situ Applications

The main results from the ex-situ pilot-scale remediation papers selected in the SLR will be highlighted in the following subsections.

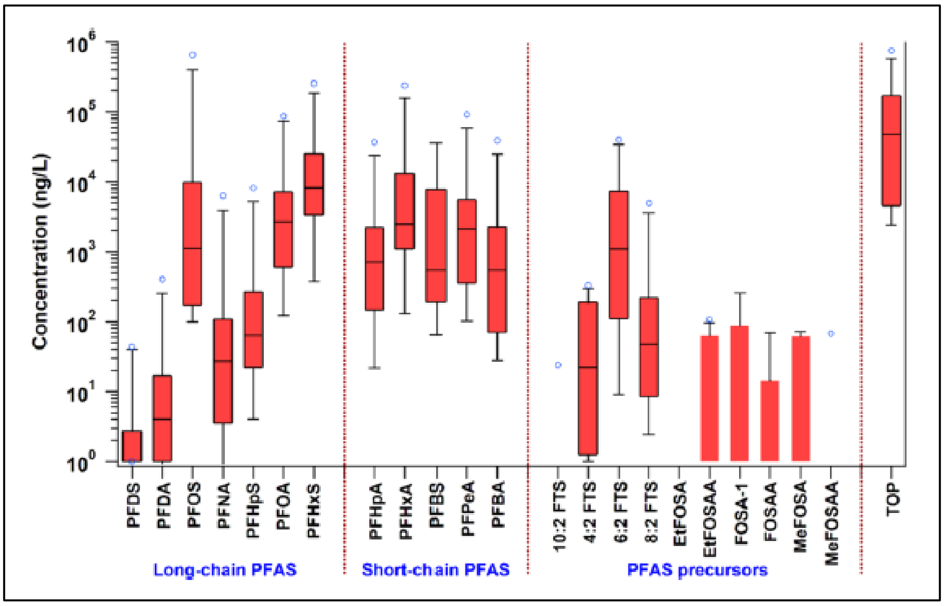
6.1.2.1.1. Paper 1: Rapid removal of poly- and perfluorinated compounds for investigation-derived waste (IDW) in a pilot-scale plasma reactor

Singh et al. (2019) proposed plasma-based water treatment as an alternative to conventional methods (such as disposal at permitted facilities or in loco pump-and-treat using granulated activated carbon or ion exchange resin, for instance) to address the large quantities of liquid investigation-derived waste (IDW) resulting from development and purge water from the installation and sampling of monitoring wells, which were part of an expansive investigation effort by the US Defense Department to identify PFAS contaminated sites, in particular AFFF-impacted sites.

The team collected 13 IDW samples from monitoring wells in fire training areas, burn pits and other sources as part of the ongoing field investigations at US Air Force installations. Each sample of 12 L was stored in a separate 18.9 L high-density polyethylene (HDPE) container at 4 °C.

To characterize the IDW samples, the researchers analyzed total organic carbon (TOC), pH, conductivity, total alkalinity, total hardness, turbidity, concentrations of PFAAs and their precursors, total oxidizable precursors (TOP) and total fluorine (F) by combustion ion chromatography (CIC). The initial PFAS (including long-chain PFAS, short-chain PFAS and precursors) and TOP concentrations are presented in Figure 6.6.

Figure 6.6 - Concentrations of PFAS and TOP

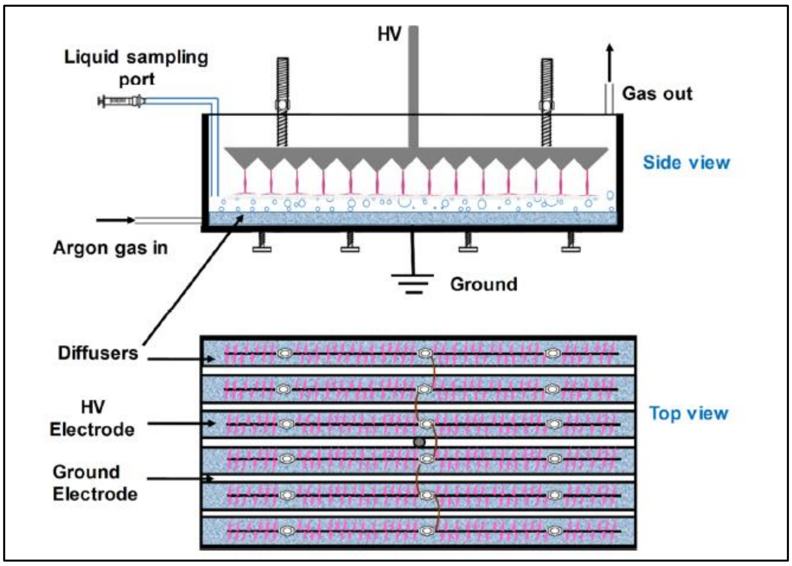


Source: prepared by Singh et al. (2019)

Overall, short-chain PFAS concentrations were the highest, with PFHxA and PFPeA presenting the highest mean values. In the long-chain PFAS group, PFOS, PFOA and PFHxS predominated.

The treatment system Singh et al. (2019) developed was a 4 L pilot-scale plasma reactor to reduce PFAA concentrations to below the USEPA's Health Advisory Concentration Level (HAL) – 70 ng/L for PFOA, PFOS and for the sum of the two – in the IDW samples. The schematic of system is shown in Figure 6.7.

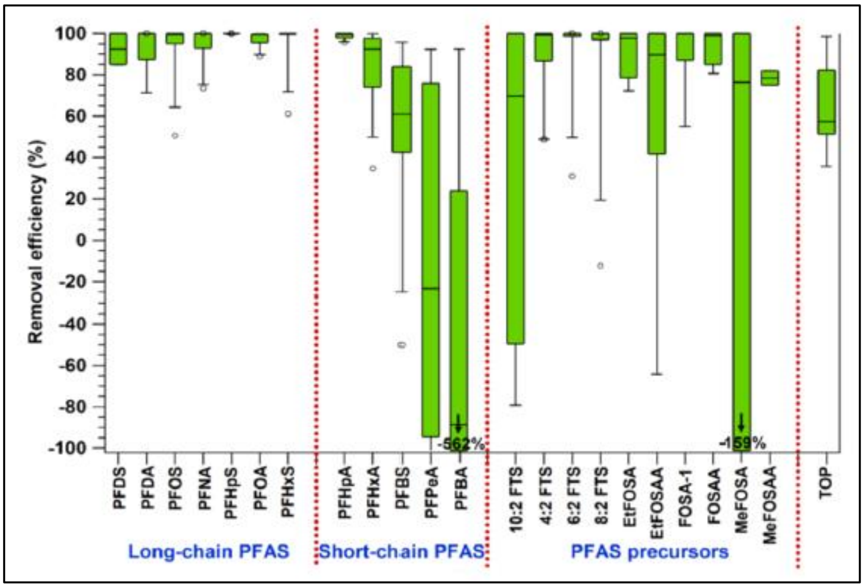
Figure 6.7 - Schematic of the 4 L plasma reactor proposed by Singh et al. (2019)



Source: prepared by Singh et al. (2019)

In the system, plasma was generated above the surface of the liquid and argon gas was bubbled through diffusers installed at the bottom of the reactor at a flow rate of 16 to 18 L/min, to form a layer of foam containing the PFAS adsorbed to the surface of the bubbles at the plasma-liquid interface. While the system was designed to operate continuously at a flow rate of 2 gallons per minute (7.57 L/min), the limited quantities of IDW used in the experiment only allowed batch operation. Figure 6.8 shows the removal efficiency of the plasma treatment for the IDW samples.

Figure 6.8 - Removal efficiencies of PFAS from the IDW samples treated with plasma



Source: prepared by Singh et al. (2019)

It is interesting that the results shown in Figure 6.8 contain negative mean removals (i.e. increasing concentration) of short-chained PFPeA (~ -20%) and PFBA (~ -90%), as well as the lower quartile of MeFOSA at -159% removal. The authors attribute this generation to the degradation of other PFAAs (in the case of PFPeA and PFBA) and other precursors (in the case of MeFOSA).

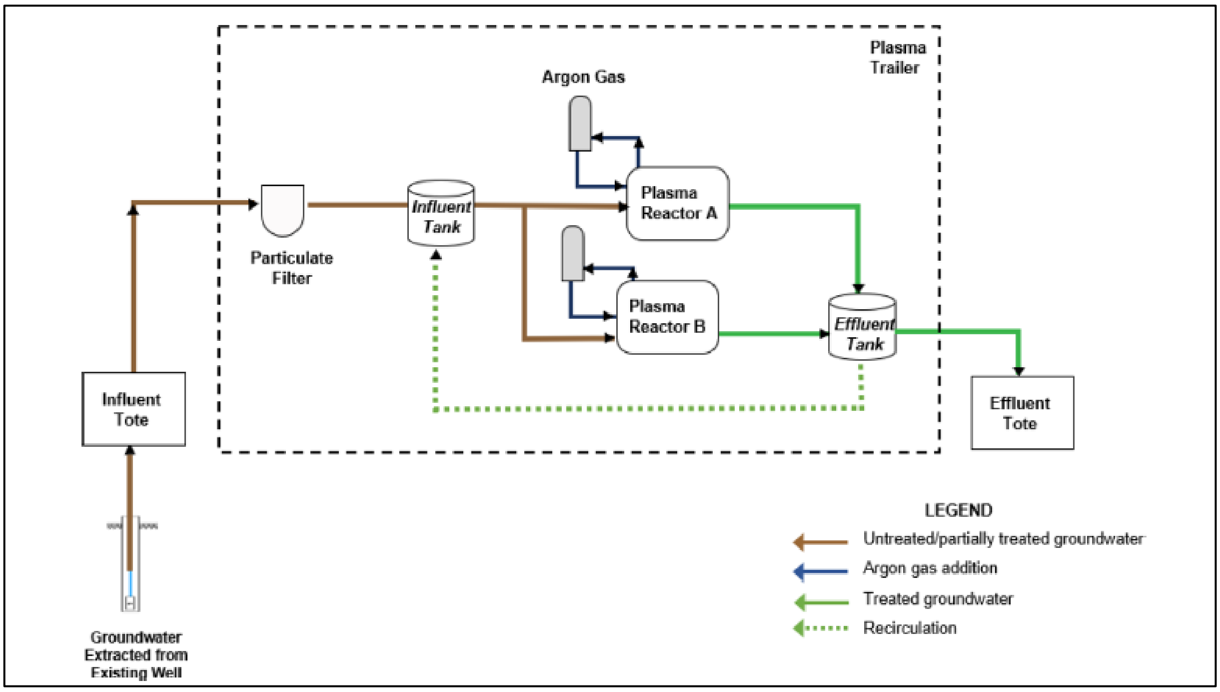
As previously stated, the remediation target adopted by the group was the USEPA’s Health Advisory Concentration Level (HAL) of 70 ng/L for PFOA, PFOS and for the sum of the two. The authors reported that for samples IDW 1 through 9, the concentrations of those compounds were below the HAL after 1 min, for samples IDW 10 and 11, after 10 minutes, for sample IDW 12, after 15 minutes, and, lastly, for IDW 13, after 50 minutes.

While the results are mixed, the results for long chain PFAS were promising enough for the authors to recommend further development of the technology. Moving forward, the attention should be in improving the treatment efficiency for short-chained PFAS – which are generally more difficult to treat – and in scaling up the reactor, as 4 L is relatively little for field applications.

6.1.2.1.2. *Paper 2: Field demonstration of a pilot-scale plasma reactor for the rapid removal of poly- and perfluoroalkyl substances in groundwater*

Following the results described in Paper 1, Nau-Hix et al. (2021) designed a plasma-based water treatment system to treat contaminated groundwater at the Wright-Patterson Air Force Base (WPAFB) in the State of Ohio, in the USA. Figure 6.9 shows the remediation system assembled by the Project team.

Figure 6.9 – Remediation system design proposed by Nau-Hix et al. (2021)



Source: prepared by Nau-Hix et al. (2021)

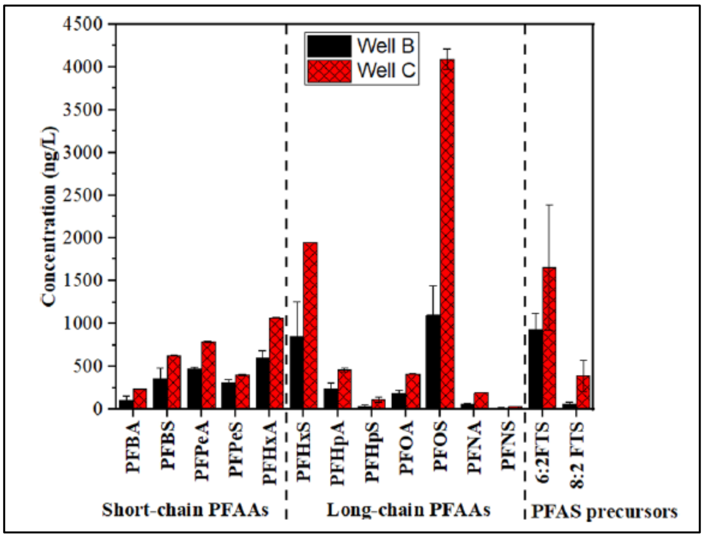
The plasma reactors used in the pilot test consisted of 4 L water reservoirs with submerged gas diffusers at the bottom and a system to generate plasma above the liquid surface. By bubbling argon gas via the submerged diffusers, the PFAS in the contaminated water are transported to the plasma-liquid interface, where the chemical degradation occurs.

For the field experiment, the team collected groundwater from two wells (denoted well B and well C). They primed both plasma reactors (denoted Plasma Reactor A and Plasma Reactor B) with municipal water and purged the air with argon gas. Contaminated water was supplied to both reactors at a maximum combined rate of 8.4 L/min. The water was cycled through each reactor 8 to 10 times at a rate between 1.1 and 4.2 L/min. For the first cycle, water was delivered directly to the reactors. For the following cycles, water was then delivered from the influent tank. It was not possible to pump all the water from either the influent or effluent tanks, so a dead zone of 11 L (each tank had 95 L capacity and were filled to between 45 L and 68.1 L) was present, such that some mixing of treated water with residual water took place.

In addition to the field reactors, a secondary batch plasma reactor was built to assess the degradation of short-chain PFAAs with the addition of a cationic surfactant (cetrimonium bromide, CTAB), to enhance PFAA transport to the plasma-liquid interface. To feed this reactor, 1.5 L of water from the field reactors was collected after 5 treatment cycles and then further treated for 2 hours, with CTAB dosed every 15 minutes to keep its concentration at 0.2 mM – frequent CTAB dosing was required due to its degradation by the plasma.

Figure 6.10 shows the initial PFAS concentrations in the groundwater from wells B and C.

Figure 6.10 - Initial PFAS concentrations

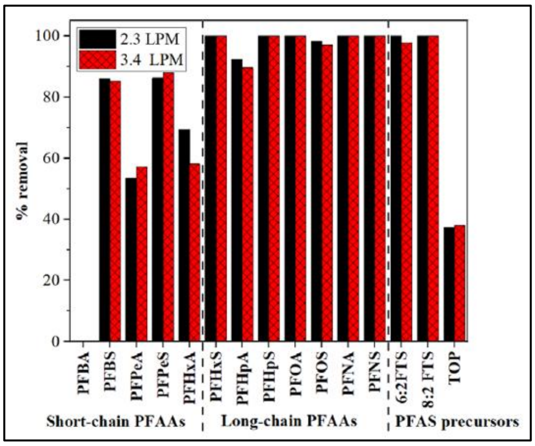


Source: prepared by Nau-Hix et al. (2021)

The total PFAS concentration in groundwater was higher at well C (27,500 ± 2,300 ng/L) than at well B (15,100 ± 3,800 ng/L). In both wells predominated long-chain PFAAs, PFHxS and PFOS, and PFAS precursor 6:2 FTS.

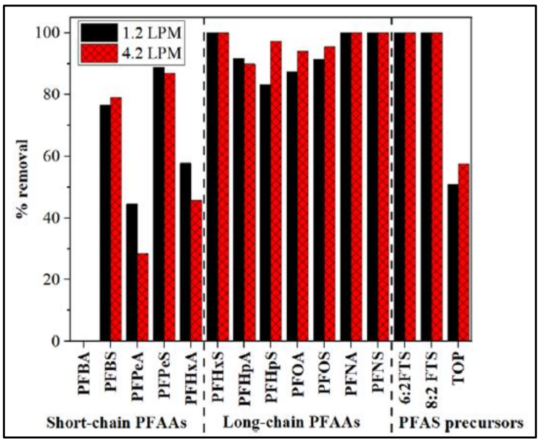
Figure 6.11 and Figure 6.12 show the percent removal of PFAAs and PFAS precursors for wells B and C, respectively, after a single cycle through the plasma reactor.

Figure 6.11 - Percent removal of PFAAs and PFAS precursors for well B



Source: prepared by Nau-Hix et al. (2021)

Figure 6.12 - Percent removal of PFAAs and PFAS precursors for well C

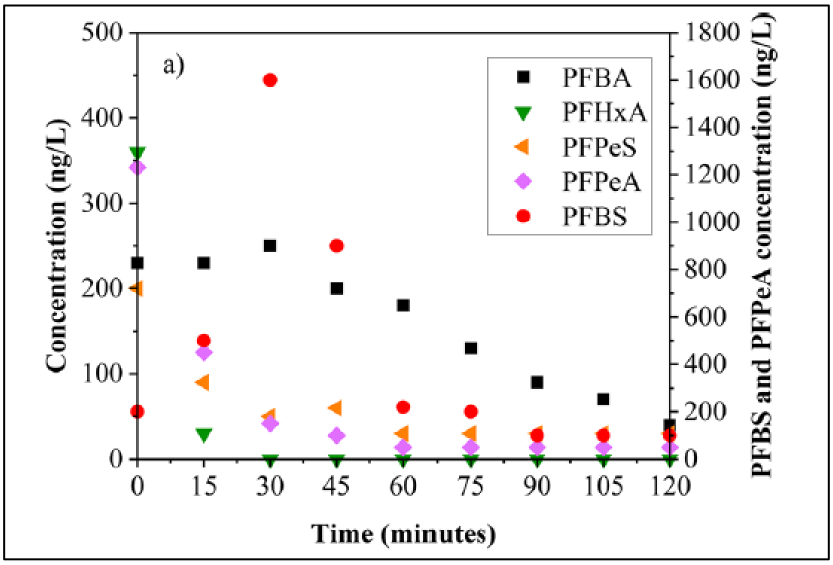


Source: prepared by Nau-Hix et al. (2021)

Overall, long-chain PFAA concentrations were reduced by over 80% in the water from both wells. Conversely, the shorter-chained compounds experienced lower degrees of degradation, likely due to their lower affinity to accumulation at the bubble-liquid interface and transport to the surface of the liquid, where exposure to the plasma would occur.

As previously mentioned, Nau-Hix et al. (2021) also ran batch experiments to test the effects of adding CTAB (a cationic surfactant) to the degradation of short-chain PFAA. The results were positive, with an overall reduction of total short-chain PFAA concentration of 88% after 120 minutes – some compounds required more time to reduce concentrations as they were concurrently formed by the degradation of longer-chained PFAA (PFBS, for instance, can be formed from the degradation of PFPeA). Figure 6.13 shows the evolution of short-chain PFAA concentrations over time treated with CTAB – as previously reported, the mixture of water from wells B and C was first treated in the field reactors, and then further treated in a 1.5 L batch reactor with the addition of CTAB.

Figure 6.13 - Evolution of short-chain PFAA concentrations over time with the addition of CTAB



Source: prepared by Nau-Hix et al. (2021)

Figure 6.13 shows that the addition of the surfactant to the plasma treatment assisted in the destruction of short-chain PFAA, which was a challenge identified in Paper 1, when the same technology was applied to treat investigation-derived waste (IDW). It should be noted, however, that after 30 minutes of treatment, the concentration of PFBS had increased from 50 ng/L to 450 ng/L, suggesting – according to the authors – that it was being produced by the degradation of PFPeA or unidentified precursors.

In both studies, Singh et al. (2019) and Nau-Hix et al. (2021) compare the electricity requirement for different treatment technologies. They used the EE/O metric, calculated as shown in the following equation:

$$EE/O = \frac{E \cdot 1000}{V \cdot \log\left(C_i/C_f\right)}$$

Where:

- EE/O is the energy needed to degrade a contaminant by an order of magnitude, measured in kWh/m^3;
- E is the energy consumed by the system, measured in kWh;
- V is the treatment volume, measured in L;
- C_i and C_f are, respectively, the initial and final concentrations of the contaminant of concern, measured in the same unit (e.g. ng/L).

Their EE/O estimate for their own technology, considering PFOA and PFOS as contaminants of concern, was 16 ± 5.8 kWh/m^3, which is lower than that of advanced reduction of PFOA (24 kWh/m^3), electrochemical oxidation of PFOA (132 kWh/m^3) or sonochemical treatment of PFOS (>20,000 kWh/m^3) (SINGH et al., 2019) (NAU-HIX et al., 2021).

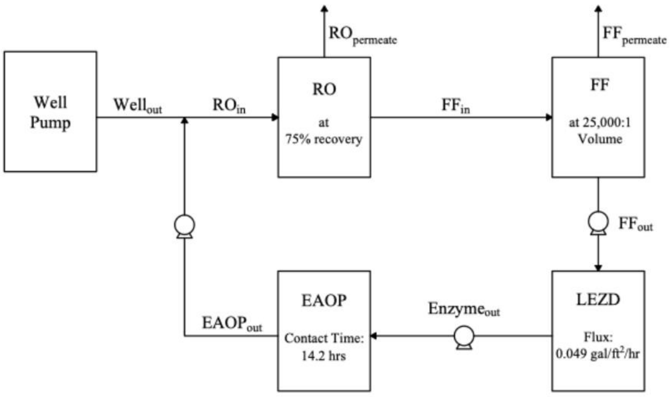
While the results are indeed encouraging, the authors did not discuss in the paper the challenges in scaling the technology to larger applications. For instance, would it be economically viable to run this system continuously with the periodic addition of the surfactant – which, in the tests, needed to be dosed every 15 minutes as it was destroyed by the plasma? Or could the poor effectiveness for short-chain PFAS be addressed in some other manner?

6.1.2.1.3. Paper 3: Destruction of per- and polyfluoroalkyl substances (PFAS) via Laccase enzymatic degradation and electrochemical advanced oxidation

Broman et al. (2021) propose a combination of techniques to extract and concentrate PFOA and PFOS from groundwater and then degrade the concentrate in a two-step treatment train.

Figure 6.14 shows the treatment train proposed by the authors.

Figure 6.14 - Two-step treatment train for PFOA and PFOS



Source: prepared by Broman et al. (2021)

The design parameters calculated by the authors are provided in Table 6.1.

Table 6.1 - Treatment train design parameters proposed by Broman et al. (2021)

Stream Name	Well_{out}	RO_{in}	$\text{RO}_{\text{permeate}}$	FF_{in}	$\text{FF}_{\text{permeate}}$	FF_{out}	$\text{Enzyme}_{\text{out}}$	EAOP_{out}
Volumetric flowrate (m^3/min)	2.65	~2.65	1.99	0.66	0.66	$1.3.10^{-5}$	$1.3.10^{-5}$	$1.3.10^{-5}$
PFOA concentration	50 ng/m^3	50 ng/m^3	Below detection limit	200 ng/m^3	Below detection limit	5 g/m^3	3.3 g/m^3	24.5 ng/m^3
PFOS concentration	50 ng/m^3	50 ng/m^3	Below detection limit	200 ng/m^3	Below detection limit	5 g/m^3	3.3 g/m^3	24.5 ng/m^3
Short Chain Concentration	1.49 ng/m^3	0	0	0	0	0	3.5 g/m^3	0.30 g/m^3
Absolute Pressure (kPa)	101.3	413	101.3	101.3	101.3	137.9	101.3	101.3
Temperature (°C)	Ambient	Ambient	Ambient	Ambient	Ambient	Ambient	Ambient	Ambient
Hydrogen Peroxide (mol/L)	0	0	0	0	0	0	0,05	0
Sodium Sulfate (mol/L)	0	0	0	0	0	0	0,10	0

Source: adapted from Broman et al. (2021)

As shown in Table 6.1, the groundwater will be pumped at a rate of 2.65 m^3/min with an aggregate concentration of PFOA and PFOS of 100 ng/m^3. It then will undergo a pre-treatment step of Reverse Osmosis (RO) which reduces the volumetric flowrate by 75%. The resulting brine will be processed by Foam Fractionation (FF), separating a permeate stream with approximately the same flowrate as the input stream with virtually no PFOS/PFOA, and an effluent stream containing an aggregate concentration of 10 g/m^3 at a much lower flowrate (25,000:1 proportionally). This effluent stream is then treated via Laccase Enzymatic Degradation, which reduces PFOA and PFOS concentration by 35% by degrading them into smaller chain compounds. The final step is the Electrochemical Advanced Oxidation Process, which has been designed to reduce concentrations of PFOA and PFOS by over 99.9%.

Broman et al. (2021) state that the design parameters for the laccase-immobilized membrane were defined according to studies on chloro-organics. The difference in chemistry may negatively impact the efficiency of the treatment train and as recommended by the authors, should be tested in a pilot-scale prior to full-scale implementation.

With regard to maintenance, the authors propose to replace the spent membrane every 25 days, disposing of the waste material in a hazardous waste landfill. While this approach is generally accepted by the USEPA, landfills in the United States are not currently required to treat leachate containing PFAS (USEPA, 2020a), such that improper management of the leachate could lead to environmental release of PFAS, defeating the purpose of the remediation technology in the first place. This may also be cause for concern if the technology is applied elsewhere, as the regulations on landfill leachate may vary by jurisdiction.

6.1.2.2. In-Situ Applications

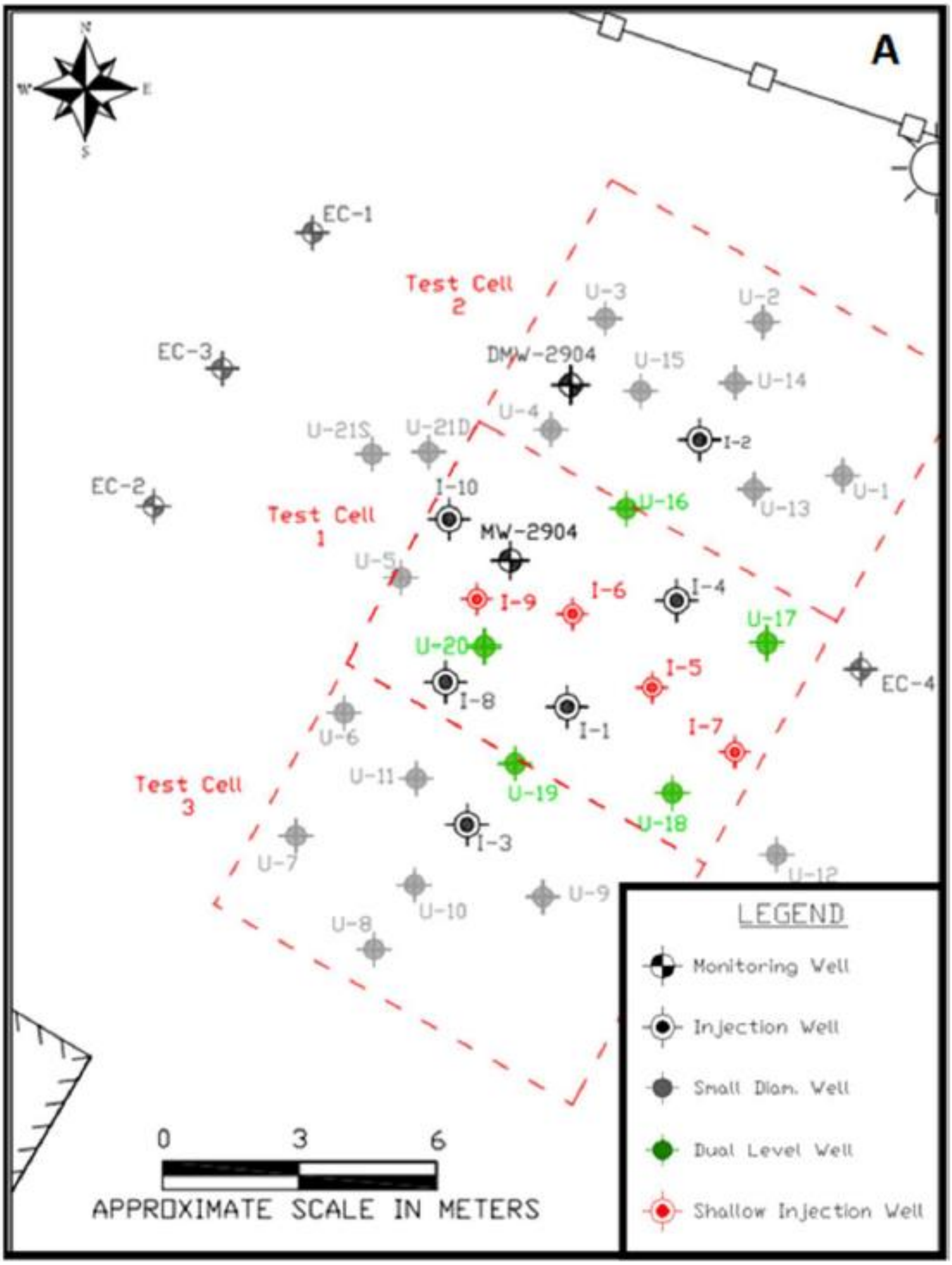
The main results from the in-situ pilot-scale remediation papers selected in the SLR will be highlighted in the following subsections.

6.1.2.2.1. Paper 4: Impact of ISCO Treatment on PFAA Co-Contaminants at a Former Fire Training Area

Eberle, Ball and Boving (2017) investigated the effects of an in-situ chemical oxidation (ISCO) treatment aimed at remediating a comingled contamination of chlorinated volatile organic compounds (cVOCs) and perfluoroalkyl acids (PFAAs). This type of co-contamination is common in aqueous film-forming foam (AFFF) impacted sites, as AFFF is used to extinguish chemical fires.

The site was a former fire training area at Joint Base Langley-Eustis, Virginia, USA. While the site was abandoned in 1980, irregular fire training activities continued until 1990. Chlorinated VOCs were the major priority pollutants, in particular 1,1,1-trichloroethane (1,1,1-TCA), dichlorobenzenes and tetrachloroethene (PCE) in both and groundwater. Due to the known history of the site as a fire training area, the team screened for PFAAs in soil (five samples) and groundwater (11 samples). Figure 6.15 presents the pilot test area.

Figure 6.15 - ISCO pilot test area used by Eberle, Ball and Boving (2017)



Source: prepared by Eberle, Ball and Boving (2017)

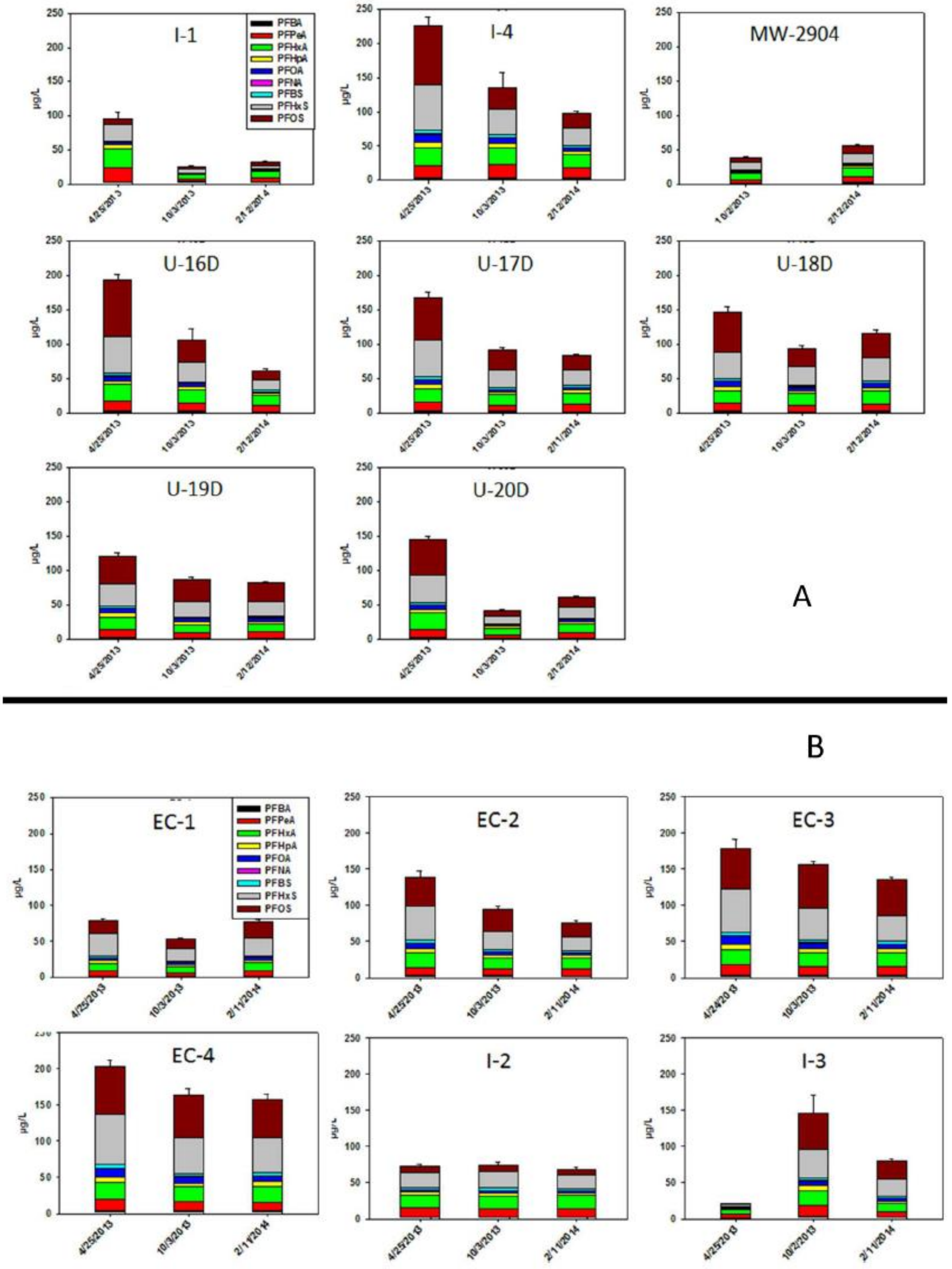
The test area was divided in three test cells. Well MW-2904 is where the contamination in the groundwater was historically highest and is located within Test Cell 1. Wells I-1, I-4, I-8 and I-10 were used for injection in the deep zone of the aquifer (defined as the region between 3.0 and 6.0 meters below ground surface – mbgs). Wells I-5, I-6, I-7 and I-9 were used to inject in the shallow zone of the aquifer (between 0.6 and 3.0 mbgs). Wells U-16 through U-20 were dual-screened and used to monitor conditions within Test Cell 1 – when discussing samples from dual-screened wells, “D” refers to deep (between 3.4 and 6.1 mbgs) and “S” refers to shallow (between 1.5 and 3.7 mbgs) samples. Wells EC-1 through EC-4, I-2 and I-3 were used to monitor migration of oxidants and contaminants outside Test Cell 1.

ISCO activities were conducted in Test Cell 1 from April through August 2013 (113 days total). 1.5 pore volumes (50.8 m^3) of peroxone (combination of ozone and hydrogen peroxide) activated persulfate (OxyZone) was injected in three separate events. The first injection event (30 m^3 over 24 days) occurred in May 2013, the second (10 m^3 over 4 days) in July 2013 (two-month interval) and the final (10 m^3 over 4 days) in August 2013 (one-month interval). An injection of 3.0 m^3 of a 3% (w/w) cyclodextrin was carried out only in the first ISCO application, to possibly enhance solubility of PFAAs due to complexation. Surfacing of injected liquids occurred in shallow zone wells I-5, I-6 and I-9, so well I-7 was the only reliable shallow injector well. In the end, the deep zone received more oxidant solution (33.4 m^3) than the shallow zone (17.4 m^3).

Baseline groundwater samples were collected in April 2013 from deep zone screened wells, and three samples from wells screened in the shallow zone. Post-remediation samples were collected 48 days after the last injection (October 2013) and 180 days after (February 2014). The post-remediation sampling scope included the wells from the baseline, and additional samples from shallow screened wells (U-16S through U-19S) and deep screened well MW-2904. Post-treatment PFAA soil samples were also collected in December 2013 and compared to pre-treatment samples collected during initial site characterization in 2012.

The groundwater results are shown in Figure 6.16. “A” denotes the wells within Test Cell 1, and “B” denotes wells outside the Cell. The result from April 2013 is from the baseline sampling event. The remaining are from post-treatment sampling.

Figure 6.16 - Groundwater sampling results from Eberle, Ball and Boving (2017)



Source: prepared by Eberle, Ball and Boving (2017)

Pre-treatment samples in Test Cell 1 had total PFAA concentrations above 100 µg/L in the deep zone wells. PFOS accounted for 26 to 50% of total aqueous PFAAs in the deep zone. In the shallow zone, pre-treatment PFAA concentrations were lower (approximately 25 µg/L).

After all three injection events, in the first post-treatment sampling, there was a decrease in total PFAA in almost all deep screened wells, primarily of PFOS and PFHxS. The lack of shallow screened wells pre-treatment samples limits the evaluation in this level of the aquifer. In the second post-treatment sampling event, PFAS concentrations remained either unchanged or decreased across the site. Small increases occurred in wells U-18D and U-20D. Outside Test Cell 1, well I-3 showed increases in total PFAS concentrations in the first post-treatment sampling event, which decreased in the second sampling event. The authors concluded that there was no evidence for lateral plume displacement or rebound during the post-treatment monitoring period. They did not conclude for the increase dissolution of PFAS as a result of the cyclodextrin injection.

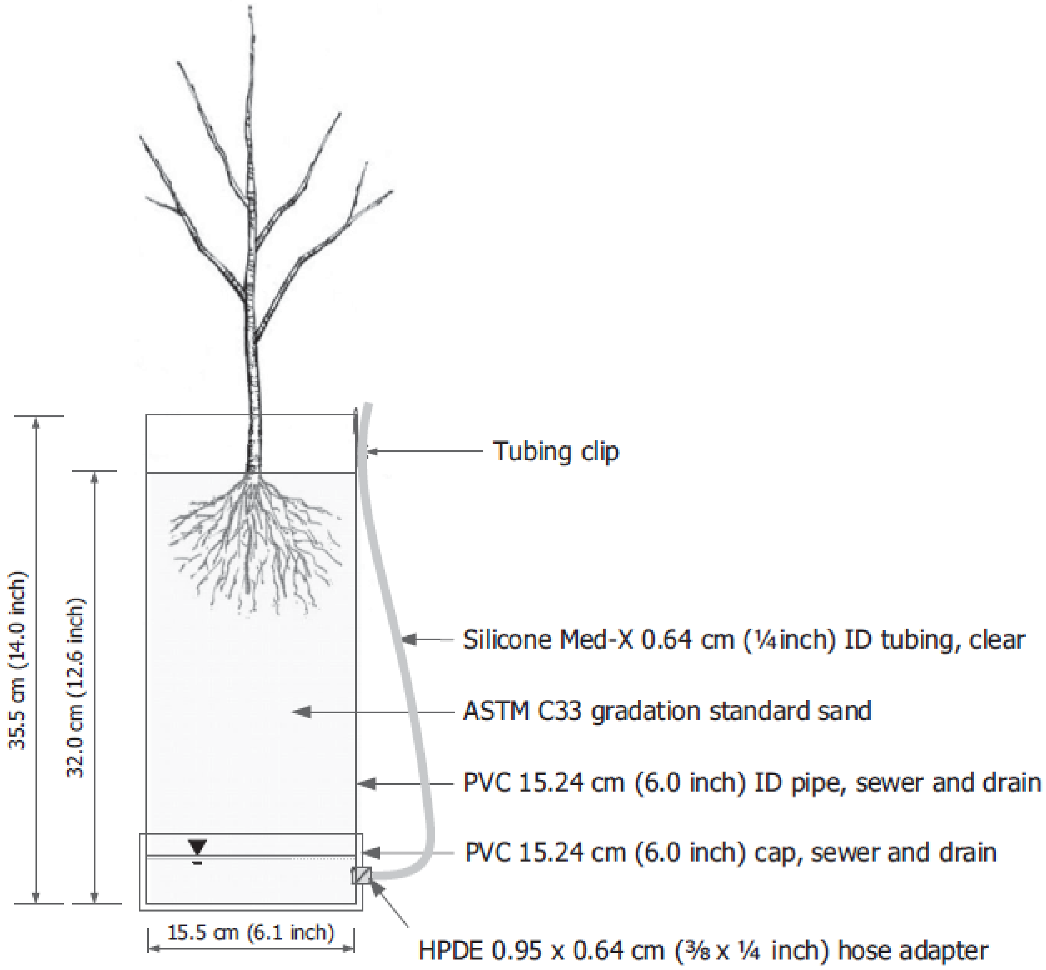
While there were reductions in PFAS concentrations in groundwater (in the range of 20 to 80%) due to the ISCO treatment that originally targeted the cVOC contamination, their reduction was not as strong as those in ex-situ treatment technologies (usually in the range of 90% depending on the PFAS).

6.1.2.2.2. *Paper 5: Accumulation of six PFAS compounds by woody and herbaceous plants: potential for phytoextraction*

Huff et al. (2020) conducted a greenhouse study to evaluate the potential for fifteen plant species to absorb PFAS.

The team selected eight herbaceous plant species and seven woody species based upon prior successful use for phytoextraction of other contaminants and their occurrence in sites known to be contaminated with PFAS. The specimens were planted in columns as schematized in Figure 6.17.

Figure 6.17 - Columns utilized by Huff et al. (2020)



Source: prepared by Huff et al. (2020)

The columns were installed inside a greenhouse that was temperature controlled at 25 ± 3 °C and with a relative humidity of 70 ± 5%. Supplemental lighting was used to extend daylight duration to 16 hours during autumn and winter. Pests were also controlled. The plants were grown for an establishment period of between 14 and 18 weeks, during which they were weekly fertilized to supply the required nutrients for adequate development. The plants were allocated in randomized blocks within three replicate blocks.

Six PFAS were chosen for testing, for the following reasons:

- PFOS and PFOA (8-chain carbon compounds) because the EPA was then evaluating the need for maximum contaminant levels;
- The EPA was developing toxicity values for PFBS (4-chain) as part of its February 2019 PFAS Action Plan;
- PFHxA, PFHxS (6-chain) and PFPeA (5-chain) were chosen to represent intermediate carbon chain lengths.

In addition, n-methyl perfluooctane sulfonamide (MeFOSA) was included in the dosing solution and analyzed as detect or non-detect (not quantified).

Contaminants were dosed in aqueous solution at a nominal concentration of 1 mg/L of each compound. The solution was applied after the establishment period, in weekly 100 mL doses to the surface of each column using a syringe to evenly distribute the solution over the soil.

Tissue samples were collected in two events. In the initial sampling event (after six contaminant doses for the herbaceous species and five doses for the woody specimens), 46 samples were collected from four herbaceous and two woody species. In the second event, a total of 128 samples were analyzed for the seven herbaceous and seven woody species. The herbaceous plants received twelve doses of contaminant, with the exception of *Brassica juncea*, *Helianthus annus* and *Trifolium incarnatum* which matured before the end of the design treatment period. The woody species received eleven doses. All six PFAS were shown to accumulate in aboveground tissue, and, with few exceptions, the PFAS plant tissue concentrations were statistically significant. Bioconcentration factors were calculated by the authors and are presented in the referred paper.

Huff et al. (2020) conclude that PFAS were shown to accumulate in above-ground portions of both herbaceous and woody plants chosen for the study. *Festuca rubra*, for instance, was shown to accumulate more than 25% of PFPeA, PFHxA and PFBS dosed in the study. Huff et al. (2020) propose that phytoremediation systems combining short-lived herbaceous plants and long-lived tree species could be used in a PFAS remediation design for sites with contaminated soil and shallow groundwater.

Although their findings were promising, the authors did not discuss the possibility that animals – such as birds – could accidently ingest the contaminated above-ground portions of the plants utilized to treat the PFAS contamination, consequently introducing the compounds in the food chain and contributing to their geographical spread. How would this affect the ecology in the area? If these birds were hunted by humans, would this result in the ingestion of PFAS-contaminated meat by residents near the treatment site? These are all important questions to consider in the case of a larger-scale application of the technology.

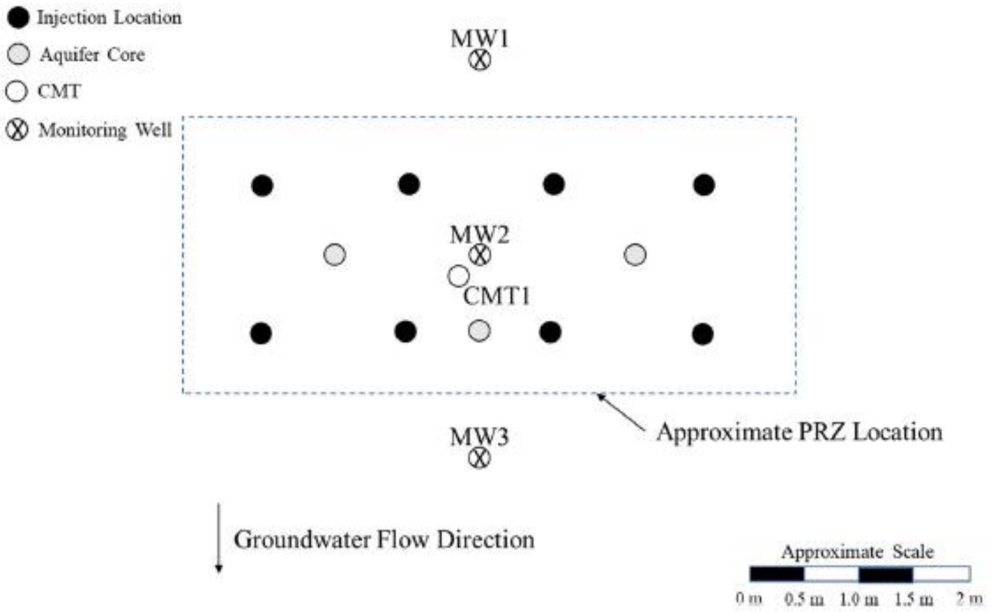
6.1.2.2.3. *<u>Paper 6: Six pilot-scale studies evaluating the in-situ treatment of PFAS in groundwater</u>*

McGregor (2020) evaluated six technologies for the in-situ treatment of groundwater contaminated with PFAS, including chemical oxidation (with persulfate and hydrogen peroxide) and sorption (powdered activated carbon - PAC, colloidal activated carbon - CAC, biochar and ion-exchange resin).

The experiment was carried out in a site with an unconfined aquifer consisting of fine-grained sand and the water table at 5.3 mbgs. The shallow aquifer at the site was impacted with PFAS and petroleum hydrocarbons in the gasoline range (concentrations up to 3,500 ug/L). Each of the six pilot-test areas span 40 m² and was instrumented with three monitoring wells (named MW1, MW2 and MW3) and one 3-channel multitubing well (CMT1).

The reagents were injected using direct-push technology (DPT) at eight points in each test cell to form the six permeable reactive zones (PRZs). Figure 6.18 shows a schematic of the PRZs.

Figure 6.18 - Schematic of the PRZs used by McGregor (2020)

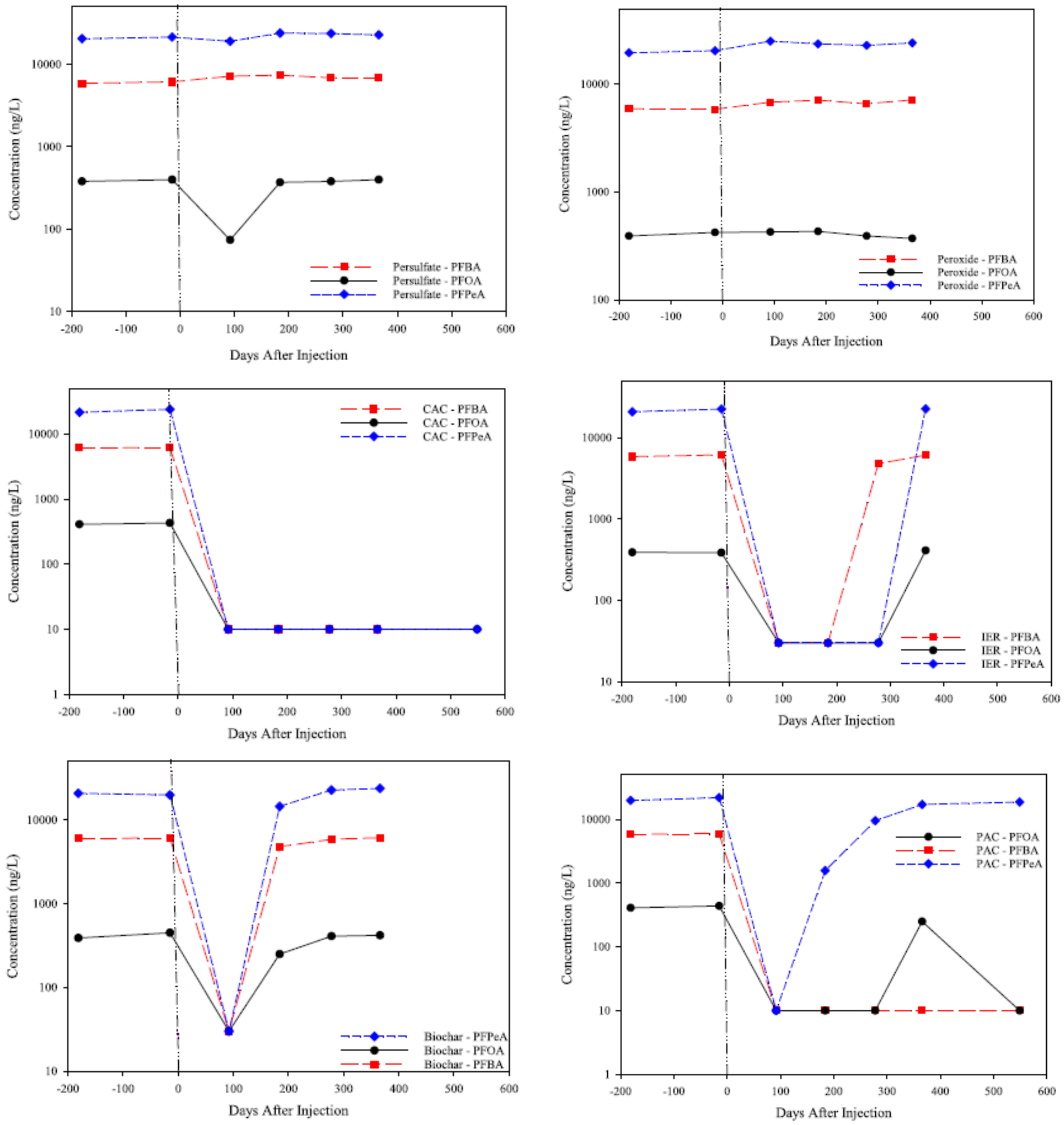


Source: prepared by McGregor (2020)

The reagents were injected in aqueous solution at 10% by weight at pressures ranging from 20 to 250 pounds per square inch (psi) (or 138 kPa to 1,724 kPa). The solution was injected in a bottom-up approach at 0,3-m vertical intervals. The solution volume targeted 40% of the effective porosity of the PRZ calculated based on an effective porosity of 20%. The effectiveness of the remediation was evaluated with the aid of groundwater samples collected on seven occasions from MW2 within each test area: twice before the injection (baseline) and on days 92, 184, 278, 366 and 549 post-injection.

Of the 23 PFAS analyzed, six were detected above their respective detection limits, with between five and nine carbon atoms: PFBA, PFPeA, PFHpA, PFOA, PFNA. The concentrations for select PFAS are shown in Figure 6.19 for all six technologies tested.

Figure 6.19 - PFAS concentrations in groundwater samples collected at MW2 in each test cell of the study by McGregor (2020)



Source: prepared by McGregor (2020)

The treatment of the six PFAS using chemical oxidants alone (no activation) showed ineffective after 366 days of treatment, with concentrations equal to the initial values or slightly higher. While the other treatments were able to reduce groundwater concentrations, they are all part of the sorption category, that do not effectively degrade the contamination and may lead to posterior release of the contamination. This suggests that chemical oxidation with sodium persulfate and hydrogen peroxide without activation are not effective at treating PFAS contamination of groundwater in-situ.

For other types of contaminants, such as organochlorine compounds (e.g. PCE and degradation products), oxidation is more effective with the addition of an activator (for sodium persulfate, for example, increasing the pH to around 11 leads to alkaline activation of the oxidant). For such hard-to-treat contaminants such as PFAS, it is possible that the activation of the oxidant could have produced more favorable results.

6.1.2.3. Evaluation of Remediation Technologies According to Sustainability Indicators

Table 6.2 applies the sustainable remediation practices and objectives proposed by Ellis and Hadley (2009) to the six applications identified in the SLR. The practices and objectives were adapted to the proposed scope, as it consists of a conceptual evaluation, such that 39 were selected.

The following nomenclature was adopted:

- Paper 1: Rapid removal of poly- and perfluorinated compounds for investigation-derived waste (IDW) in a pilot-scale plasma reactor;
- Paper 2: Field demonstration of a pilot-scale plasma reactor for the rapid removal of poly- and perfluoroalkyl substances in groundwater;
- Paper 3: Destruction of per- and polyfluoroalkyl substances (PFAS) via Lacasse enzymatic degradation and electrochemical advanced oxidation;
- Paper 4: Impact of ISCO Treatment on PFAA Co-Contaminants at a Former Fire Training Area;
- Paper 5: Accumulation of six PFAS compounds by woody and herbaceous plants: potential for phytoextraction;
- Paper 6: Six pilot-scale studies evaluating the in-situ treatment of PFAS in groundwater.
    - In line with the objectives of this study, only the pilot-scale tests of chemical oxidation will be considered in the classification - i.e., the sorption technologies tested will not be considered.

For each practice or objective that was fulfilled, “Y” was entered into the table. Conversely, “N” was entered when the practice or objective was not fulfilled. “NA” was entered when the practice or objective was not applicable to the technology. “P” was entered when there was potential that the practice or objective could be incorporated in the project design. The following scoring system was then applied:

- “Y” = 1;
- “P” = 0,5;
- “N” or “NA” = 0.

The total scores were calculated based on the above point system and normalized according to the maximum number of points per sustainability area (18 points for social, 21 for environmental and 13 for economic) on a scale from 0 (lowest) to 10 (highest).

Table 6.2 - Sustainability Evaluation Matrix

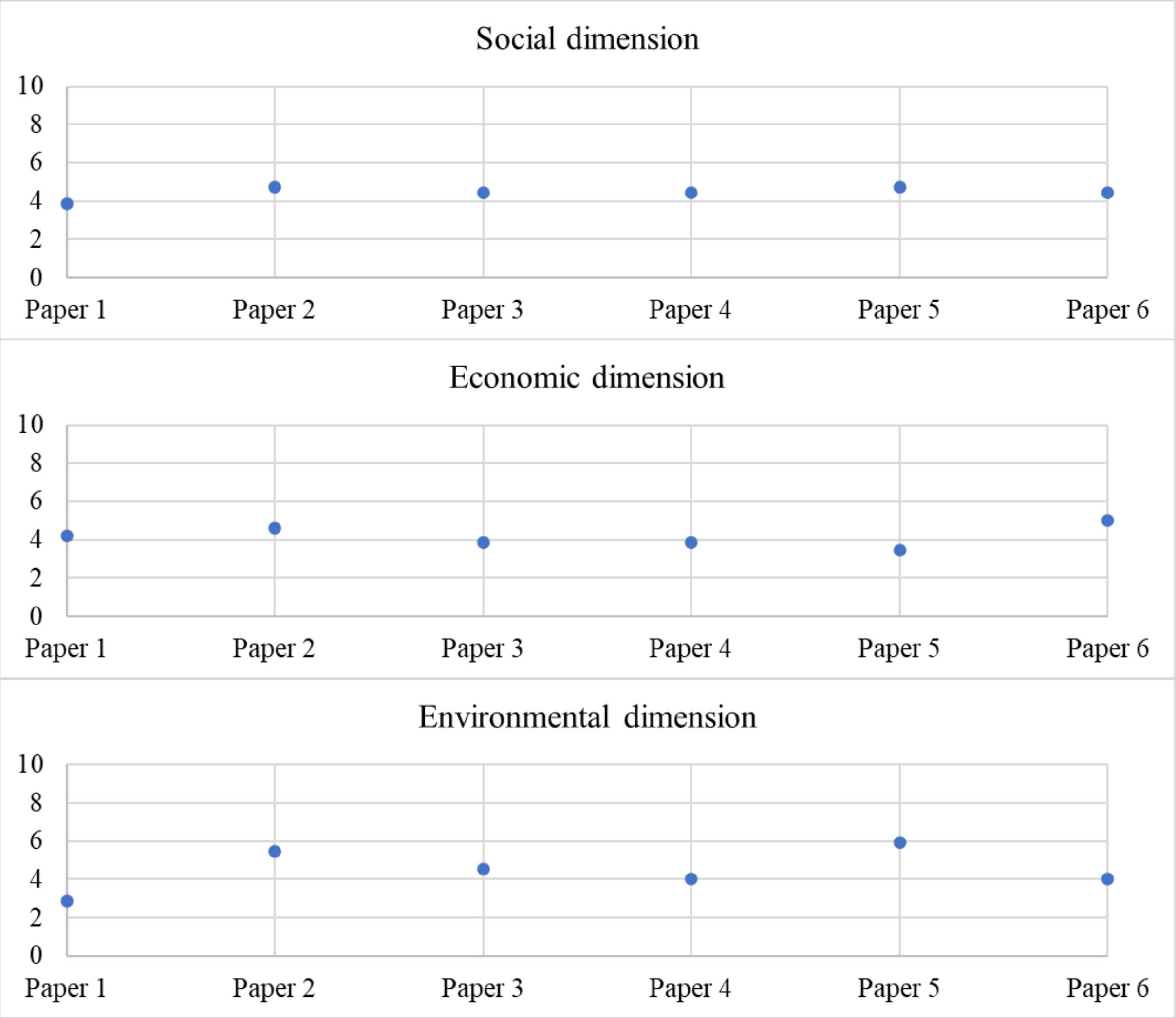
Sustainable Remediation Practices and Objectives	Triple Bottom Line Element(s)	Paper 1	Paper 2	Paper 3	Paper 4	Paper 5	Paper 6
Minimize freshwater consumption	Environmental	Y	Y	Y	N	Y	Y
Maximize water reuse	Environmental	P	Y	NA	P	N	NA
Conserve groundwater resources	Environmental	NA	Y	Y	Y	Y	N
Prevent runoff and negative impacts to surface water	Environmental	P	NA	NA	NA	P	N
Minimize bioavailability of contaminants through source and plume control	Environmental	NA	Y	Y	Y	P	N
Maximize biodiversity	Environmental	NA	NA	NA	NA	P	N
Minimize soil and habitat disturbance	Environmental	NA	NA	NA	N	Y	N
Favor minimally invasive in-situ technologies	Environmental	NA	Y	Y	N	Y	Y
Favor low-energy technologies (e.g., bioremediation, phytoremediation) where possible and effective	Environmental	P	P	N	Y	Y	P
Protect native ecosystem and avoid introduction of non-native species	Environmental	NA	NA	NA	NA	P	N
Minimize risk to ecological receptors	Environmental	NA	NA	NA	NA	P	N
Preserve natural resources	Environmental	NA	NA	NA	NA	P	P
Use telemetry or remote data collection when possible	Environmental	NA	P	P	P	P	P
Reduce emissions of greenhouse gases contributing to climate change	Environmental	P	P	P	P	P	P
Prevent offsite migration of contamination	Environmental	NA	Y	Y	P	Y	N
Minimize material extraction and use	Environmental, Economic	NA	Y	Y	Y	P	Y
Minimize waste	Environmental, Economic	Y	Y	P	P	P	Y
Maximize materials reuse	Environmental, Economic	NA	NA	NA	NA	NA	P
Recycle or reuse project waste streams	Economic	Y	NA	NA	NA	NA	P
Use operations data to continually optimize and improve the remedy	Economic	P	P	P	P	NA	P
Consider the net economic result	Economic	P	P	P	P	P	P
Improve the tax base/economic value of the property/local community	Economic, Social	NA	NA	NA	NA	NA	NA
Maximize employment and educational opportunities	Economic, Social	NA	NA	NA	NA	NA	NA
Minimize O&M cost and effort	Economic, Social	P	P	P	P	P	P
Minimize health and safety risk during remedy implementation	Economic, Social	Y	Y	P	P	Y	P
Maximize acres of a site available for reuse	Economic, Social	P	P	P	P	P	P
Maximize number of sites available for reuse	Economic, Social	NA	P	P	P	P	P
Use locally sourced materials	Environmental, Economic, Social	P	P	P	P	P	P
Minimize noise, odor, and lighting disturbance	Environmental, Social	P	P	P	P	Y	Y
Favor technologies that permanently destroy contaminants	Environmental, Social	Y	Y	Y	Y	N	P
Avoid environmental and human health impacts in already disproportionately impacted communities	Social	P	P	P	P	Y	P
Consider net positive/negative impact of the remedy on local community	Social	P	P	P	P	P	P
Assess current, potential, and perceived risks to human health, including contractors and public, over the remedy life cycle	Social	P	P	P	P	P	P

Sustainable Remediation Practices and Objectives	Triple Bottom Line Element(s)	Paper 1	Paper 2	Paper 3	Paper 4	Paper 5	Paper 6
Prevent cultural resource losses	Social	NA	NA	NA	NA	NA	NA
Integrate stakeholders into decision-making process	Social	P	P	P	P	P	P
Solicit community involvement to increase public acceptance and awareness of long-term activities and restrictions	Social	P	P	P	P	P	P
Maintain or improve public access to open space	Social	NA	P	P	P	P	P
Create goodwill in the community through public outreach and open access to project information	Social	P	P	P	P	P	P
Consider future land uses during remedy selection and choose remedy appropriately	Social	NA	P	P	P	P	P

Source: adapted from Ellis and Hadley (2009)

The normalized scores (on a scale from 0 to 10) for each of the three dimensions of the triple bottom line are shown in Figure 6.20.

Figure 6.20 - Normalized scores for the six papers selected in the SLR in each dimension of the triple bottom line



Source: prepared by the author

It was difficult to distinguish between the technologies in the social and economic dimensions, mainly due to the conceptual nature of the present study. A more thorough assessment would be required in a real-world application to fully assess these two dimensions. With regard to the environmental dimension, Papers 2 and 5 propose technologies that slightly outperform the other 4 papers.

7. CONCLUSION

The objective of this study was to identify innovative and sustainable remediation technologies for the destruction of PFAS contamination in soil and groundwater as described in the scientific literature. Through searches in the scientific databases SCOPUS and Web of Science, 482 unique articles were obtained on this subject matter. Among these, 105 papers were selected according to a set of criteria detailed in Chapter 5, which included a multitude of remediation technologies applied to water (groundwater, surface water and wastewater) and soil at different scales (bench-, pilot- and full-scale). Papers that described sorption-only applications were excluded, as this group of technologies is one of the more traditional for PFAS remediation, thus resulting in a subset of 53 articles.

A descriptive statistics evaluation of the 53 articles revealed that the number of studies increased rapidly since 2014, with 27 articles having been published in 2020 and 2021 alone. The majority, i.e. 29 of the articles, described ex-situ remediation technologies, and 51% were for the remediation of contaminated groundwater. This shows that in-situ remediation, in particular of soil, is still a subject of interest for future research.

Among the 53 articles, 47 were bench-scale studies and six were pilot-scale studies. These six articles were further analyzed and the technology that showed the most promise was plasma-based water treatment (PWT) as it was able to degrade long-chain PFAA concentrations upwards of 80% in both investigation-derived waste (IDW) and groundwater. For shorter-chained compounds, the team investigated the application of a cationic surfactant and were able to reduce total short-chain PFAA concentrations by 88% after 120 minutes of treatment. The team also compared the energy requirements of their technology against advanced reduction, electrochemical oxidation and sonochemical treatment, and showed that PWT may be utilized at a lower energetic cost.

With regard to the sustainability of the technologies, it was difficult to distinguish them in the social and economic dimensions due to the conceptual nature of the analysis proposed in this study. However, in the environmental dimension, Papers 2 (ex-situ plasma-based groundwater treatment) and 5 (in-situ phytoremediation) outperformed the other four. It should be noted that in-situ phytoremediation, though, is generally limited in its applicability to soil and shallow groundwater contaminations and must be applied in combination with other technologies.

In conclusion, considering the set of articles retrieved in this SLR, plasma-based water treatment was shown to be the most innovative technology currently approaching real-world application, also performing well sustainability-wise. However, plasma-based water treatment needs further development to be able to treat short-chain PFAS adequately – which is a weakness common to other remediation technologies. While this is a positive result, this SLR also shows that further research is needed to develop new technologies for PFAS remediation and to scale up the most promising to pilot- and full-scale applications.

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ATTACHMENT A – LIST OF ARTICLES ANALYZED IN THE SLR

Table A.1 – List of the 53 articles analyzed in the SLR

Title	Authors	Year of Publication
Sonochemical Degradation of Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoate (PFOA) in Landfill Groundwater: Environmental Matrix Effects	Cheng, Jie and Vecitis, Chad D. and Park, Hyunwoong and Mader, Brian T.and Hoffmann, Michael R.	2008
Peroxidase-mediated degradation of perfluorooctanoic acid	Colosi, Lisa M. and Pinto, Roger A. and Huang, Qingguo and Weber, Jr., Walter J.	2009
High-valent iron-based oxidants to treat perfluorooctanesulfonate and perfluorooctanoic acid in water	Yates, Brian J. and Darlington, Ramona and Zboril, Radek and Sharma, Virender K.	2014
Electrochemical treatment of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) in groundwater impacted by aqueous film forming foams (AFFFs)	Schaefer, C.E. and Andaya, C. and Urtiaga, A. and McKenzie, E.R. and Higgins, C.P.	2015
Efficient Sorption and Removal of Perfluoroalkyl Acids (PFAAs) from Aqueous Solution by Metal Hydroxides Generated in Situ by Electrocoagulation	Lin, Hui and Wang, Yujuan and Niu, Junfeng and Yue, Zhihan and Huang, Qingguo	2015
Laccase-Catalyzed Degradation of Perfluorooctanoic Acid	Luo, Qi and Lu, Junhe and Zhang, Hao and Wang, Zunyao and Feng, Mingbaoand Chiang, Sheau-Yun Dora and Woodward, David and Huang, Qingguo	2015
Heat-activated persulfate oxidation of PFOA, 6:2 fluorotelomer sulfonate, and PFOS under conditions suitable for in-situ groundwater remediation	Park, S. and Lee, L.S. and Medina, V.F. and Zull, A. and Waisner, S.	2016
Transformation of Polyfluorinated compounds in natural waters by advanced oxidation processes	Anumol, T. and Dagnino, S. and Vandervort, D.R. and Snyder, S.A.	2016
Treatment of Aqueous Film-Forming Foam by Heat-Activated Persulfate under Conditions Representative of in Situ Chemical Oxidation	Bruton, T.A. and Sedlak, D.L.	2017
Impact of ISCO Treatment on PFAA Co-Contaminants at a Former Fire Training Area	Eberle, D. and Ball, R. and Boving, T.B.	2017
Plasma-Based Water Treatment: Efficient Transformation of Perfluoroalkyl Substances in Prepared Solutions and Contaminated Groundwater	Stratton, G.R. and Dai, F. and Bellona, C.L. and Holsen, T.M. and Dickenson, E.R.V. and Mededovic Thagard, S.	2017

Alternate Reductants with VB12 to Transform C8 and C6 Perfluoroalkyl Sulfonates: Limitations and Insights into Isomer Specific Transformation Rates, Products and Pathways	Park, Saerom and de Perre, Chloe and Lee, Linda S.	2017
Laccase induced degradation of perfluorooctanoic acid in a soil slurry	Luo, Q. and Liang, S. and Huang, Q.	2018
Electrochemical Transformations of Perfluoroalkyl Acid (PFAA) Precursors and PFAAs in Groundwater Impacted with Aqueous Film Forming Foams	Schaefer, C.E. and Choyke, S. and Ferguson, P.L. and Andaya, C. and Burant, A. and Maizel, A. and Strathmann, T.J. and Higgins, C.P.	2018
Sorption, Aerobic Biodegradation, and Oxidation Potential of PFOS Alternatives Chlorinated Polyfluoroalkyl Ether Sulfonic Acids	Chen, H. and Choi, Y.J. and Lee, L.S.	2018
Treatment of perfluoroalkyl acids by heat-activated persulfate under conditions representative of in-situ chemical oxidation	Bruton, T.A. and Sedlak, D.L.	2018
Development of macroporous Magneli phase Ti4O7 ceramic materials: As an efficient anode for mineralization of poly- and perfluoroalkyl substances	Lin, Hui and Niu, Junfeng and Liang, Shangtao and Wang, Chong and Wang, Yujuan and Jin, Fangyuan and Luo, Qi and Huang, Qingguo	2018
Fungal biotransformation of 6:2 fluorotelomer alcohol	Merino, Nancy and Wang, Meng and Ambrocio, Rocio and Mak, Kimberly and O'Connor, Ellen and Gao, An and Hawley, Elisabeth L. and Deeb, Rula A.and Tseng, Linda Y. and Mahendra, Shaily	2018
Rapid Degradation and Mineralization of Perfluorooctanoic Acid by a New Petitjeanite Bi3O(OH)(PO4)(2) Microparticle Ultraviolet Photocatalyst	Sahu, Sushant P. and Qanbarzadeh, Mojtaba and Ateia, Mohamed and Torkzadeh, Hamed and Maroli, Amith S. and Cates, Ezra L.	2018
Assessing Continued Electrochemical Treatment of Groundwater Impacted by Aqueous Film-Forming Foams	Schaefer, C.E. and Andaya, C. and Maizel, A. and Higgins, C.P.	2019
Electrodialytic per- and polyfluoroalkyl substances (PFASs) removal mechanism for contaminated soil	Sörengård, M. and Niarchos, G. and Jensen, P.E. and Ahrens, L.	2019
Perfluoroalkyl and polyfluoroalkyl substances thermal desorption evaluation	Crownover, E. and Oberle, D. and Kluger, M. and Heron, G.	2019

Inhibition of Perchlorate Formation during the Electrochemical Oxidation of Perfluoroalkyl Acid in Groundwater	Yang, Shasha and Fernando, Sujan and Holsen, Thomas M. and Yang, Yang	2019
Rapid Removal of Poly- and Perfluorinated Compounds from Investigation-Derived Waste (IDW) in a Pilot-Scale Plasma Reactor	Singh, Raj Kamal and Multari, Nicholas and Nau-Hix, Chase and Anderson, Richard H. and Richardson, Stephen D. and Holsen, Thomas M. and Thagard, Selma Mededovic	2019
Application of surfactant modified montmorillonite with different conformation for photo-treatment of perfluorooctanoic acid by hydrated electrons	Chen, Zhanghao and Tian, Haoting and Li, Hui and Li, Jian Sheng and Hong, Ran and Sheng, Feng and Wang, Chao and Gu, Cheng	2019
Degradation of Perfluorooctane sulfonate by Reactive Electrochemical Membrane Composed of Magnéli Phase Titanium Suboxide	Shi, Huanhuan and Wang, Yaye and Li, Chenguang and Pierce, Randall and Gao, Shixiang and Huang, Qingguo	2019
Structural Dependence of Reductive Defluorination of Linear PFAS Compounds in a UV/Electrochemical System	Rao, U. and Su, Y. and Khor, C.M. and Jung, B. and Ma, S. and Cwiertny, D.M. and Wong, B.M. and Jassby, D.	2020
Evaluation of PFAS treatment technology: Alkaline ozonation	Thomas, R. and Jenkins, K. and Landale, B. and Trigger, G. and Holsen, T.M. and Dore, S. and Pope, D. and Wasielewski, J.	2020
Six pilot-scale studies evaluating the in-situ treatment of PFAS in groundwater	McGregor, R.	2020
Remediation of PFAS-Contaminated Soil and Granular Activated Carbon by Smoldering Combustion	Duchesne, Alexandra L. and Brown, Joshua K. and Patch, David J. and Major, David and Weber, Kela P. and Gerhard, Jason I.	2020
Accumulation of six PFAS compounds by woody and herbaceous plants: potential for phytoextraction	Huff, David K. and Morris, Lawrence A. and Sutter, Lori and Costanza, Jed and Pennell, Kurt D.	2020
Thermal desorption as a high removal remediation technique for soils contaminated with per- and polyfluoroalkyl substances (PFASs)	Sorengard, M. and Lindh, A-S. and Ahrens, L.	2020

Destruction of Per- and Polyfluoroalkyl Substances (PFASs) in Aqueous Film-Forming Foam (AFFF) with UV-Sulfite Photoreductive Treatment	Tenorio, Raul and Liu, Jinyong and Xiao, Xin and Maizel, Andrew and Higgins, Christopher P. and Schaefer, Charles E. and Strathmann, TimothyJ.	2020
Removal of perfluoroalkanesulfonic acids (PFSAs) from synthetic and natural groundwater by electrocoagulation	Bao, Jia and Yu, Wen-Jing and Liu, Yang and Wang, Xin and Liu, Zhi-Qunand Duan, Yan-Fang	2020
Photodegradation of Hexafluoropropylene Oxide `Timer Acid under UV Irradiation	Li, Chen and Mi, Na and Chen, Zhanghao and Gu, Cheng	2020
Enhanced thermal activation of peroxymono sulfate by activated carbon for efficient removal of perfluorooctanoic acid	Liu, Guanhong and Li, Chao and Stewart, Brittney Ashley and Liu, Lin andZhang, Meng and Yang, Mingyang and Lin, Kuangfei	2020
Reductive transformation of perfluorooctane sulfonate by nNiFe(0)-Activated carbon	Zenobio, Jenny E. and Modiri-Gharehveran, Mahsa and de Perre, Chloe and Vecitis, Chad D. and Lee, Linda S.	2020
Degradation of perfluorooctanoic acid by zero-valent iron nanoparticles under ultraviolet light	Xia, Chunjie and Liu, Jia	2020
Remediation of perfluorooctanoic acid (PFOA) polluted soil using pulsed corona discharge plasma	Zhan, J. and Zhang, A. and Héroux, P. and Guo, Y. and Sun, Z. and Li, Z. and Zhao, J. and Liu, Y.	2020
Degradation of PFOS and PFOA in soil and groundwater samples by high dose Electron Beam Technology	Lassalle, J. and Gao, R. and Rodi, R. and Kowald, C. and Feng, M. and Sharma, V.K. and Hoelen, T. and Bireta, P. and Houtz, E.F. and Staack, D. and Pillai, S.D.	2021
Iron-activated persulfate oxidation degrades aqueous Perfluorooctanoic acid (PFOA) at ambient temperature	Tran, T. and Abrell, L. and Brusseau, M.L. and Chorover, J.	2021
An electrocoagulation and electrooxidation treatment train to remove and degrade per- and polyfluoroalkyl substances in aqueous solution	Shi, H. and Chiang, S.-Y.D. and Wang, Y. and Wang, Y. and Liang, S. and Zhou, J. and Fontanez, R. and Gao, S. and Huang, Q.	2021

Enhanced Recovery of Per- And Polyfluoroalkyl Substances (PFASs) from Impacted Soils Using Heat Activated Persulfate	Shojaei, M. and Kumar, N. and Chaobol, S. and Wu, K. and Crimi, M. and Guelfo, J.	2021
Mechanochemical remediation of perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) amended sand and aqueous film-forming foam (AFFF) impacted soil by planetary ball milling	Turner, L.P. and Kueper, B.H. and Jaansalu, K.M. and Patch, D.J. and Battye, N. and El-Sharnouby, O. and Mumford, K.G. and Weber, K.P.	2021
Destruction of Per- And Polyfluoroalkyl Substances (PFAS) via Lacasse Enzymatic Degradation and Electrochemical Advanced Oxidation	Broman, J. and Ceja, A. and Godoy, T. and Rivera, D.R. and Dionne, P. and Schipper, J. and Henkemeyer, S. and Cegielski, S. and Wong, G. and Kaur, A.	2021
PFAS degradation in ultrapure and groundwater using non-thermal plasma	Palma, D. and Papagiannaki, D. and Lai, M. and Binetti, R. and Sleiman, M. and Minella, M. and Richard, C.	2021
Photodegradation of Perfluorooctane sulfonic Acid on Fe-Zeolites in Water	Qian, L. and Kopinke, F.-D. and Georgi, A.	2021
Monitored natural attenuation to manage PFAS impacts to groundwater: Potential guidelines	Newell, Charles J. and Adamson, David T. and Kulkarni, Poonam R. and Nzeribe, Blossom N. and Connor, John A. and Popovic, Jovan and Stroo, Hans F.	2021
Field Demonstration of a Pilot-Scale Plasma Reactor for the Rapid Removal of Poly- and Perfluoroalkyl Substances in Groundwater	Nau-Hix, Chase and Multari, Nicholas and Singh, Raj Kamal and Richardson, Stephen and Kulkarni, Poonam and Anderson, Richard Hunter and Holsen, Thomas M. and Thagard, Selma Mededovic	2021
Improved electrical driving current of electrochemical treatment of Per-and Polyfluoroalkyl Substances (PFAS) in water using Boron-Doped Diamond anode	Sukeesan, Suratsawadee and Boontanon, Narin and Boontanon, Suwanna Kitpati	2021

Hydrothermal Alkaline Treatment for Destruction of Per- and Polyfluoroalkyl Substances in Aqueous Film-Forming Foam	Hao, Shilai and Choi, Youn-Jeong and Wu, Boran and Higgins, Christopher P. and Deeb, Rula and Strathmann, Timothy J.	2021
Impacts of Reactor Configuration, Degradation Mechanisms, and Water Matrices on Perfluorocarboxylic Acid Treatment Efficiency by the UV/Bi3O(OH)(PO4)(2) Photocatalytic Process	Qanbarzadeh, Mojtaba and Wang, Dawei and Ateia, Mohamed and Sahu,Sushant P. and Cates, Ezra L.	2021
Rapid photo-reductive destruction of hexafluoropropylene oxide trimer acid (HFPO-TA) by a stable self-assembled micelle system of producing hydrated electrons	Chen, Zhanghao and Teng, Ying and Huang, Liuqing and Mi, Na and Li, Chenand Ling, Jingyi and Gu, Cheng and Jin, Xin	2021

Source: prepared by the author