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Review

Comprehensive Review of Global Perspectives on Per- and Polyfluoroalkyl Compounds: Occurrence, Fate, and Remediation in Groundwater Systems

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Abstract: Groundwater contamination with per- and polyfluoroalkyl compounds (PFASs) has become a growing worldwide environmental issue. The current review comprehensively evaluates the global perspective of PFAS pollution in groundwater. Data from 224 recent research articles covering various land use and source types were reviewed, including industrial facilities, landfills, biosolids applications, and firefighting training sites. The bibliographic analysis shows an exponential increase in publications on PFAS pollution in groundwater in the last five years, with more than 50% coming from the USA, followed by Australia, Canada, China, and Sweden. The recent groundwater PFAS pollution research provides insight into the analytical techniques, absorbing materials, treatment strategies, field tests, and enhanced natural attenuation. Nevertheless, the current review identified significant research gaps in the areas of precursor characterization, subsurface behavior, model validation with field data, and long-term and sustainable solutions. Moreover, a global cross-disciplinary approach is required to reduce and regulate PFASs' risks to humans and the ecological system. This review presents a case study of PFASs in Saudi Arabian groundwater, revealing elevated levels of PFOA and PFOS and highlighting the need for region-specific studies and remediation strategies. The review results will guide global efforts to protect drinking water supplies from life-threatening contaminants.

Keywords: PFAS; emerging contaminant; groundwater quality; sustainability

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1. Introduction

Per- and polyfluoroalkyl substances (PFASs) have emerged as persistent organic pollutants of global concern [1]. PFASs comprise a large group of anthropogenic chemicals containing fluorinated alkyl chains and have been widely used in industrial processes and consumer products since the 1950s [2]. The carbon–fluorine bonds impart oil and water repellency, temperature and chemical resistance, and surfactant properties, making PFASs useful across many applications [3]. However, the same properties lead to the environmental persistence of PFASs, bioaccumulation in food products, and potential human health effects [4].

PFASs enter the environment through manufacturing discharges, the use and disposal of PFAS-containing products, land applications of biosolids, and aqueous film-forming foam (AFFF) [5]. Consequently, PFASs have been detected universally in surface water, groundwater, soil, sediment, biota, and humans [6]. Many PFASs are not readily biodegradable and are highly mobile in groundwater [7]. Among them, Perfluorooctanoic Acid (PFOA), Perfluorooctanesulfonic Acid (PFOS), Perfluorohexane Sulfonic Acid (PFHxS), Perfluorononanoic Acid (PFNA), and Hexafluoropropylene Oxide Dimer Acid (HFPO-DA) are widely detected in the subsurface environment. Even trace levels in groundwater used for drinking water can accumulate to harmful concentrations for human consumption [8]. According to the recently released national primary drinking water regulation by the United States Environmental Protection Agency (USEPA), the maximum contamination levels of PFOS, PFOA, PFHxS, PFNA, and HFPO-DA are 4 ng/L, 4 ng/L, 10 ng/L, 10 ng/L, and 10 ng/L, respectively [9].

The current review aims to provide a timely and comprehensive analysis that synthesizes the current global state of scientific knowledge regarding the occurrence, fate, transport, source, global distribution, and remediation of PFASs in groundwater systems. It focuses on PFAS contamination in groundwater systems that serve as the primary source of drinking water supply. This review emphasizes global coverage and perspectives, focusing on findings from regions with extensive and recent PFAS research, including North America, China, Europe, Australia, and Asia. Additionally, this review incorporates data on PFASs in Saudi Arabia, underscoring the need to understand PFAS behavior in groundwater of arid regions and guiding future research in unstudied areas.

Recently, extensive research has been conducted on PFAS groundwater contamination in many countries worldwide. However, no recent comprehensive review has integrated the research findings to understand the global research trend. This knowledge gap motivates us to undertake the current review. The main goal of the review is to fill the knowledge gap by evaluating the current global state of the scientific knowledge on the PFAS contamination of groundwater, with a focus on the specific objectives as follows: (1) to evaluate the current knowledge of PFAS leaching processes, natural attenuation, subsurface transformations of the precursor compounds, fate and transport, advancements in remediation technology, and implementation at the field scale, and (2) to identify gaps and limitations in the scientific understanding and management of PFAS groundwater pollution and propose recommendations for future research directions.

2. Literature Screening and Bibliography

A literature review was performed on the Scopus database using specified search keywords such as “PFAS and groundwater”, “PFAS and contamination and groundwater”, “PFAS and aquifer”, “PFOS and groundwater”, “PFOA and groundwater”, and “PFAS and groundwater and remediation” for the period from 2016 to 2023. A total of 224 documents were obtained through the searching process, and the literature search findings were analyzed and visualized using the VOSviewer program (version 1.6.19), as shown in Figure 1. The review reveals that the major subjects were identified via the key keywords’ co-occurrence, including PFAS sources such as landfill, aqueous film-forming foam (AFFF), and wastewater. The literature analysis showed two types of PFASs that appeared with high frequency, including Perfluorooctanoic acid (PFOA) and Perfluorooctanesulfonic acid (PFOS). The third subject was highlighted by the co-occurrence of keywords such as adsorption, adsorbent, activated carbon, ion exchange, advanced oxidation, and phytoremediation, highlighting the research topic of PFAS remediation and treatment. The fourth subject includes keywords such as contaminant transport, fate, transport, retardation, vadose zone, leaching, and heterogeneity, indicating the topic of the fate and transport of PFASs in aquifer materials (Figure 1). The current literature analysis shows that the United States of America has the most publications on the PFAS contamination of groundwater, with more than 50% of the total global publications. The other countries with significant publications are Australia, Canada, China, Sweden, and Italy (Figure 2a). The literature analysis also reveals that there has been exponential growth in the number of publications in the field of PFASs and groundwater resources in the last three years, with the maximum publications reaching seventy articles in 2023, indicating the significant impact of PFASs on groundwater availability and suitability for human use (Figure 2b).

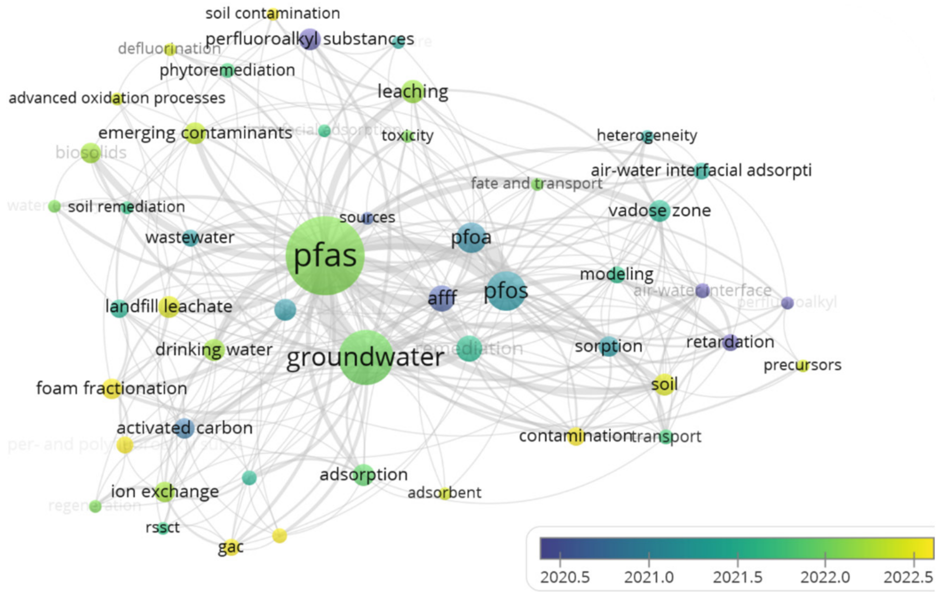

Figure 1. Map of the co-occurrence keywords displays the main topics in the PFAS contamination of groundwater systems.

Recent research has made major advancements in PFAS characterization, treatment, occurrence investigations, fate and transport mechanisms, and remediation, all of which aim to enhance PFAS-impacted groundwater management. For example, McDermett et al. (2022) [10] designed a high-resolution passive sampler to capture PFASs from aqueous solutions with high accuracy and efficiency. Their innovative sampler gave better concentration data across time and space than typical grab sampling. Li et al. (2022) [11] demonstrated a machine learning approach for preliminary PFAS exposure screening based on long-term water quality monitoring data. The method showed promising accuracy exceeding 96% for predicting contamination risks. Alam et al. (2022) [12] applied citizen science sampling to examine the potential impacts of biosolid applications on PFAS levels in private wells, elucidating potential links between PFASs in biosolids and groundwater contamination. Regarding treatment methods for PFAS-contaminated groundwater, Newell et al. (2023) [3] reviewed practical recharge estimation approaches for calculating PFAS mass discharge, a key metric, proposing a tiered framework to reduce uncertainty. Pannu et al. (2023) [7] evaluated granular activated carbon performance for removing low-level PFAS mixtures across different source waters, informing design considerations. Singh et al. (2023) [4] synthesized and demonstrated novel adsorbent materials, showing an exceptional removal capability through synergistic adsorbent modifications. Kassar and Boyer (2023) [13] offered insights on optimizing anion exchange resin selection and regeneration to improve PFAS treatment efficiency. At the field scale, Kulkarni et al. (2022) [14] successfully demonstrated sonochemical treatment for the in situ degradation of PFASs in groundwater. Moreover, Liang et al. (2022) [15] integrated ion exchange and electrochemical oxidation, presenting a promising combined solution for cost-effective PFAS removal and destruction. For occurrence studies, He (2023) [8] reviewed contamination patterns in Chinese groundwater, highlighting priority PFASs for regulation to manage pollution. Rehnstam et al. (2023) [6] applied high-resolution mass spectrometry to characterize PFAS sources better, like landfill leachate. Fate and transport studies of PFASs in the subsurface environment have received attention in recent years. For example, Chen et al. (2023) [16] modeled the transport of PFASs at the scale of pores in unsaturated porous media. Liu et al. (2021) [17] modeled long-term PFOS persistence in groundwater after source treatment due to plume migration. Rovero et al. (2021) [18] evaluate the challenges and uncertainty of modeling PFAS sorption to soils based on literature data. Schroeder et al. (2021) [19] demonstrated atmospheric dispersion as a pathway for PFASs to impact remote groundwater areas far from industrial facilities. Niarchos et al. (2023) [20] illustrate the factors controlling PFAS plumes with in situ colloidal activated carbon treatment due to complex subsurface hydrogeology.

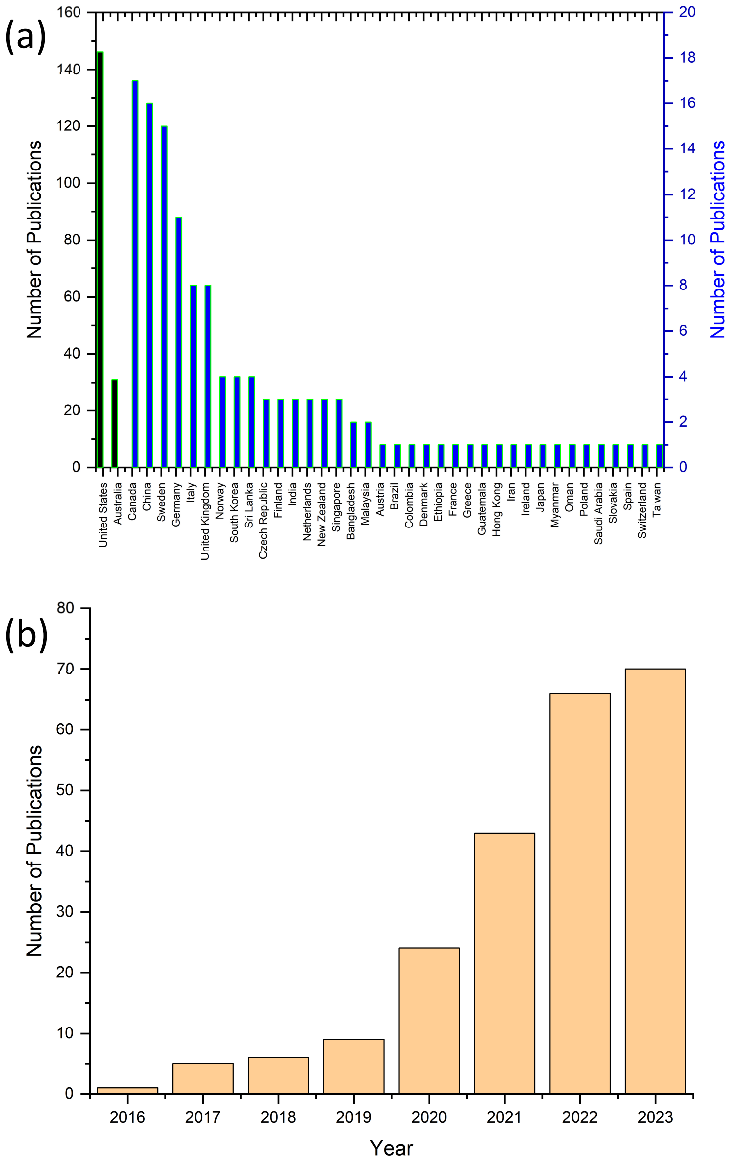

Figure 2. The total number of publications on groundwater contamination by PFASs (**a**) grouped by country and (**b**) grouped by year, covering the period from 2016 to 2023.

3. Source of PFASs in the Groundwater System

3.1. Industry

PFAS production factories have been one of the main sources of groundwater contamination in many regions worldwide (Figure 3) [21]. Groundwater near existing and historical PFOS, PFOA, PFHxS, and other PFAS production plants in China, India, and other nations has consistently been shown to have remarkably elevated levels of PFAS compounds. The concentration of PFASs in sample wells and monitoring stations downstream of production facilities displays a higher value than health recommendation limits [22–24]. For example, Xu et al. (2021) [25] investigated the PFAS contamination of groundwater near a fluorochemical industrial park in China, and they found high levels of PFOA, PFBA, PFOS, PFHxS, and PFBS with mean of 1144.6 ng/L, 44.4 ng/L, 96.2 ng/L, 355.2 ng/L, and 11,016.2 ng/L, respectively. The values of these compounds exceeded the health advisory limit and maximum contamination level as set by the USEPA [9,26–28]. Another study by Li et al. (2020) [24] found PFAS values up to 185.8 ng/L in groundwater wells in the downstream region of Maozhou River, China, where industrial factories of paper textiles and firefighting foam occurred. The extreme pollution is directly attributable to decades of PFAS-laden waste stream discharges and runoff from industrial and production sites. In addition, using aqueous film-forming foams (AFFFs), including PFASs, at military sites, commercial airports, and firefighting training facilities has also resulted in severe groundwater pollution [23,29]. PFASs penetrate underground soils and groundwater at these locations via equipment testing releases, fire-suppression system leaks, and active firefighting actions. PFAS chemicals accumulate in groundwater underneath and downstream of areas where AFFF has been widely employed for years. Recent investigations have shown elevated concentrations of PFASs in groundwater samples from AFFF-impacted sites, thoroughly surpassing drinking water limits. For example, Ruyle et al. (2023) [30] reported high levels of PFAS concentration, especially PFOS and PFOA, in groundwater at a former firefighting site in Cape Cod, Massachusetts, with a maximum PFOS concentration of 22,000 ng/L. Similarly, AFFF sites in North America, Europe, and Asia have PFAS levels several orders of magnitude beyond the EPA's health advisory limits and maximum contamination level [17,31].

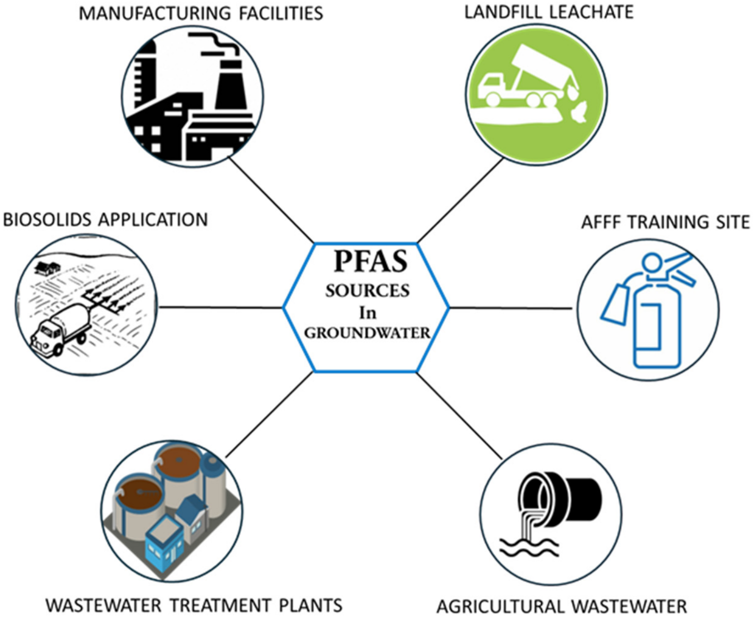

Figure 3. Diagram illustrating the potential source of PFASs in groundwater.

3.2. Landfill Leachate

Leachate from municipal solid waste dumps has become a major source of PFAS release into the environment (Figure 3) [32]. PFAS chemicals are found in several discarded consumer items, commodities, and solid waste dumped in landfills. As precipitation and moisture seep through landfills, they release a complex combination of dissolved PFASs into the leachate [33,34]. Several PFAS chemicals were found in landfill leachate samples, including PFOS, PFOA, PFHxS, and PFBS, as reported by several researchers [6,34,35]. Leachate regularly leaks or overflows from landfill containment systems, contaminating soil, surface water, and groundwater [36]. Chen et al. (2023) [35] studied 26 PFASs in China's landfill leachate, groundwater, and surface water. They found a high PFAS concentration in municipal solid waste (MSW) leachate, up to 10,000 ng/L. In contrast, groundwater and surface water have lower concentrations with mean values of less than 500 ng/L. Recent studies have investigated the change in PFAS contamination in groundwater near landfill sites with respect to time, space, and depth. For example, Sadia et al. (2023) [36] investigated the presence of 57 PFAS compounds in the groundwater of the Netherlands near a landfill site. They found that sum PFAS concentrations decreased with distance from the landfill site from 80–1900 ng/L to 30–460 ng/L. Similarly, the sum PFAS concentration decreased with depth from 1900 ng/L at 65 m to 86 ng/L at 89 m.

3.3. Wastewater Treatment Facilities

Treated wastewater effluent discharged from municipal wastewater treatment facilities has been recognized as a source of PFAS contamination in groundwater systems (Figure 3) [37]. PFASs are introduced into wastewater streams from both industrial sources and residence sewage. Although current treatment systems could reduce PFAS levels, noticeable quantities of PFASs have still been detected in wastewater treatment effluent [38]. Using treated wastewater for irrigation or aquifer recharge exposes PFASs to an environment where they may accumulate and reach a health-concern level [39]. Recent studies have investigated PFAS concentration in the groundwater system and soil in the region, which used treated wastewater for aquifer recharge and irrigation. For instance, Cáñez et al. (2021) [37] studied PFAS chemicals at the Sweetwater Recharge Facility (SRF) in Tucson, Arizona, wherein treated wastewater recharges the local aquifer. They found that the reclaimed water has PFAS levels exceeding the EPA health advisory limit (70 ng/L) [40]. They also found that the combined PFOS and PFOA concentrations correlated with rising and decreasing groundwater levels, suggesting PFASs are delivered to the aquifer during recharge episodes. Another recent study by Xingchun et al. (2023) [41] investigated PFAS contamination in shallow and deep groundwater in a Beijing-region reclaimed water irrigation area. They found that 90% of samples contained PFASs, mostly PFOA, PFBA, and PFBS. The average value of the sum PFAS level was 10.55 ng/L, ranging from 1.05 to 34.28 ng/L. The PFAS concentration was noted to decrease with depth, suggesting a larger contamination risk for shallow aquifers.

3.4. Agriculture Application of Biosolids

The use of sewage sludge biosolids, a byproduct of wastewater treatment, as agricultural fertilizer has also been associated with increased levels of PFAS contamination in groundwater (Figure 3) [42]. Using biosolids results in the spread of PFASs such as PFOS, PFOA, and other similar substances onto agricultural fields, which has the potential to seep gradually into the shallow groundwater system [12,42,43]. Scientists worldwide have documented the presence of PFASs and similar substances in groundwater and soil after using PFAS-containing soil fertilizers such as biosolids [43,44]. For example, Johnson (2022) [43] investigated PFAS contamination in soil and groundwater at an agricultural field in the United States's western region where biosolids had long been applied. They found that the surface and vadose zone soil contained several PFASs, with PFOS, PFDA, and PFOA being the most prevalent. They also showed that PFOA and PFOS infiltrated into the soil depths of up to 9 m, with detectable concentrations (0.029 μg/L and 0.002 μg/L for PFOA and PFOS, respectively) in groundwater around 17 m below the surface. Alam et al. (2022) [12] investigated the levels of 20 PFASs in groundwater near biosolid application locations in Pennsylvania, USA. They found detectable PFAS concentrations in 73.5% of the analyzed samples with a maximum value of 80.4 ng/L, exceeding the USEPA 2022 health advisory limits [45] and 2024 maximum contamination level [9].

4. Global Trends of PFASs in The Groundwater System

PFASs have raised worldwide concerns due to their enduring, bioaccumulate, and possibly harmful properties. Figure 4 presents a comprehensive global distribution of the mean values of the sum PFAS concentrations across different groundwater settings. We classified the data on PFAS pollution into four categories to facilitate the mapping and interpretation of pollution occurrence, utilizing the latest guidelines issued by the USEPA [9,28,46]. These categories include 0–20 ng/L (low PFAS pollution), 20–70 ng/L (moderate pollution), 70–3200 ng/L (high pollution), and >3200 ng/L (extreme pollution). The highest mean values of sum PFASs were reported from eastern Canada and eastern China. However, low pollution of PFASs was observed in India, the Maltese Islands, and

South Korea. The other countries reported various pollution levels of PFASs in groundwater, from low to moderate in the USA, moderate to high in Australia, high levels in Brazilian groundwater, moderate in Sweden, and low to high pollution levels in other regions of China.

The compiled global data on PFASs in urban and agricultural wells often reported specific PFASs, such as PFOS and PFOA. China has detected PFOS in quantities above 50,000 ng/L [5], indicating the country's extensive use of these chemicals in industrial applications. Moreover, PFOA has been detected at levels as high as 10,000 ng/L in municipal and agricultural wells [8], revealing its extensive use and the risk of groundwater pollution. Nevertheless, these patterns are not exclusive to China; PFOS and PFOA are often found in Spain, Australia, Germany, and the USA. However, the levels vary from region to region, with high levels of sum PFASs in Canada and China (sum PFAS > 3200 ng/L) (Figure 4). Other PFASs, such as PFHxS, PFNA, and PFBS, have been identified in agricultural wells and industrial site monitoring wells in Australia [47]. The concentrations of these compounds reach the level of 5000 ng/L, as reported by McGregor (2020) [48]. The wide range of PFASs indicates an intricate influx of different products containing PFASs into the environment, most likely due to the country's significant industrial and agricultural operations. PFASs PFPeA, PFHxA, and 6:2 FTS have been identified in both municipal and industrial wells in the United States and Sweden, as reported by Newell et al. (2021) [49], Guelfo and Adamson (2017) [22], and Alam et al. (2022) [12]. The detection of these substances signifies the use of PFASs in diverse industrial sectors, ranging from fire-suppression foams to coatings with resistance to stains, ultimately resulting in the pollution of groundwater. Meanwhile, the detection of PFASs such as HFPO-DA, PFDoA, PFTrDA, PFUnDA, and PFDoDA in both municipal and industrial wells across the USA, China, Canada, and the Netherlands underlines the widespread environmental dissemination of these substances [13–15,36]. Their presence in groundwater sources can be attributed to direct (e.g., industrial discharge and landfill leachate) and indirect (e.g., atmospheric deposition and surface runoff) contamination pathways.

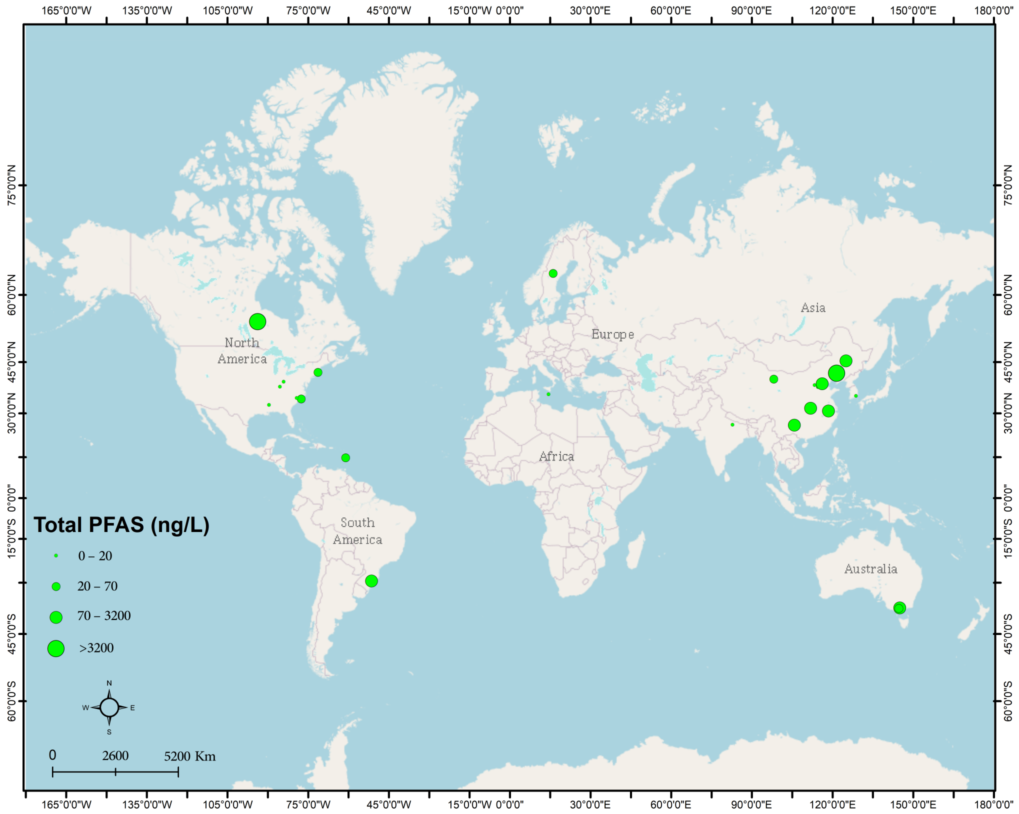

Figure 4. Map showing the global distribution of sum PFASs in groundwater systems [17,23,24,34,47,50–60].

5. Fate and Transport Mechanisms

5.1. Leaching Processes

PFASs migrate from sources like landfills and biosolids-amended fields to groundwater through vadose zone leaching processes. Several factors have been reported in the literature as key controls on the leaching process, including the infiltration rate, soil properties, and source hydraulic loading effect [3]. Moreover, the macropores and joints in the subsurface formation act as preferential pathways that can accelerate PFAS transport to the groundwater system [4]. The properties of PFASs, like chain length, also impact PFAS leaching processes in the vadose zone, with longer-chain Perfluoroalkyl Acids (PFAAs) more readily leached; however, shorter-chains are more mobile once leached [6]. Anionic PFAAs move readily through low-organic-matter soils, while precursor–neutral forms are more retarded [7]. The climate influences the leaching process of PFASs, with higher PFAS mass discharge during wet periods 13]. Additionally, irrigation and stormwater infiltration enhance PFAS leaching from biosolids-amended fields, waste sites, and urban areas [36].

5.2. Natural Attenuation

Many natural mechanisms attenuate PFAS plumes during groundwater migration, including hydrodynamic dispersion, sorption, and degradation [22]. Hydrophobic partitioning retards neutral precursor transport, while anion exclusion enhances PFAA plume spreading [49,61–63]. Newell et al. (2021) [49] proposed field evidence guidelines to evaluate attenuation potential via subsurface retention. McGregor (2020) [48] found that reversible attachment/detachment sorption interactions with aquifer solids occur in PFAAs and precursors, while the sorption affinity depends on the carbon chain length and functional groups. Using column experiments, Singh et al. (2023) [4] demonstrate that soil cation exchange can reduce PFAA leaching significantly. However, limited and variable retardation has been observed in field investigations, restricting the use of partition coefficients in models [17]. It is reported that the biodegradation of PFAAs is negligible, and the transformation of precursor compounds like fluorotelomer alcohols occurs in groundwater systems [64]. Precursors from WWTPs and landfills undergo sequential transformations to PFCAs via oxidative pathways [65]. More research is required on the importance of transformation versus transport and sorption for plume evolution. Assessing attenuation processes using multiple lines of evidence is critical to predicting PFAS plume dynamics for contaminated site management and water supply sustainability [25].

5.3. Precursor Transformations

PFASs are introduced to groundwater as precursor compounds like fluorotelomer alcohols and sulfonamides through waste streams from manufacturing, land applications, and AFFF use [18]. Precursors undergo biotic and abiotic transformations during subsurface transport to form stable PFAAs detected as groundwater contaminants [66]. Several pathways have been proposed for precursor transformation based on intermediates identified in laboratory and field studies [64]. Proposed reaction mechanisms include dehydrogenation, oxidation, hydrolysis, decarboxylation, and dealkylation, converting precursors stepwise to Perfluoroalkyl carboxylic acids (PFCAs) like PFOA or Perfluorosulfonic acids (PFSAs) like PFOS [63]. Reaction rates depend on the environmental system's redox state, pH, temperature, co-solutes, and microbes. Precursor biotransformation occurs in WWTPs, soils, and aquifers, and microcosm studies show the contributions of ammonia-oxidizing bacteria [15]. Abiotic transformations of PFASs, including oxidation, hydrolysis, and elimination reactions enhanced by radical species or catalysts, have been reported in the recent literature [7]. Transformation kinetics derived from microcosms inform reactive transport models. However, submetabolite toxicity and reaction intermediates require further research [67]. Precursor degradation occurs concurrently with sorption and dispersion during plume evolution [68]. Quantifying the relative importance of transformation versus transport and retention remains challenging. Improved analytical methods are needed to detect precursors and transformation products at environmentally relevant concentrations in groundwater. Coupled characterization and reactive transport modeling promise to elucidate precursor behavior and risks.

6. Remediation Technologies

6.1. Ex Situ Methods

Ex situ or pump-and-treat methods involve extracting contaminated groundwater for above-ground PFAS treatment and discharge or reinjection after cleanup. Granular activated carbon (GAC) adsorption is a commonly applied ex situ technology for PFAS remediation due to its high capacity across a broad range of compounds [7,69]. However, regeneration or incineration is required once saturated [70]. Ion exchange and polymeric resins also effectively adsorb PFASs for ex situ applications [71]. Chemical oxidation using persulfate or permanganate degrades PFASs but is limited by kinetics and competitive organics [22]. Nanofiltration, reverse osmosis, and electrocoagulation remove diverse PFASs from extracted groundwater through size exclusion and electrostatic interactions [72]. Thermal treatment methods like incineration have been used to degrade concentrated PFASs in streams; however, they require high energy [48]. Burns et al. (2021) [64] assessed the Surface-Active Foam Fractionation (SAFF) method in a field scale for removing PFASs from polluted groundwater, and they found that the SAFF successfully eliminated more than 99.5% of PFASs. The ex situ sequencing of separation and destructive techniques enables the efficient polishing of trace PFASs [64]. Ex situ pump-and-treat requires extensive wells networking for the plume capture and reinjection capacity for treated water, and the off-site disposal of the spent media increases costs and environmental impacts [17]. Integrating multiple technologies through a treatment train approach can achieve the desired cleanup goals for PFAS-contaminated groundwater.

6.2. In Situ Methods

In situ technologies treat PFASs within aquifers using injected reagents or induced chemical/biological processes without extracting water. In situ chemical oxidation, which uses catalyzed hydrogen peroxide or activated persulfate, has been used effectively to degrade PFASs within injection zones [73]. Hydrophobic sorption amendments like activated carbon or organoclays retain PFASs when emplaced in high-permeability zones [66]. Degradation approaches include direct photolysis using UV lamps, sonochemical ultrasonic reactors, and electrochemical techniques that mineralize PFASs, which some researchers have implemented [14,74]. Bioremediation stimulates native microbial communities to degrade precursor compounds in saturated zones through electron donor addition [64]. Thermal technologies like electrical resistance heating volatilize and recover PFASs in situ [65].

In situ methods avoid extraction costs and offer passive operation; however, they have challenges achieving uniform delivery and contact for heterogeneous aquifers [25]. Amendment performance relies on adequate hydrogeologic characterization and hydraulic control [75]. PFAS destruction is often incomplete, requiring polishing or multiple passes [14]. Field demonstrations have shown promise but require validation across different sites [15]. In situ techniques present promising innovative solutions for PFAS remediation but need further development and testing. Integrated in situ technologies like oxidation or stabilization with ex situ polishing may provide optimal treatment trains tailored to site conditions and plume characteristics [72]. Integrating source zone, plume, and hydraulic control remedies can achieve remediation goals more efficiently while minimizing costs and environmental impacts. Remediation planning requires assessing the performance, implementation ability, and sustainability of available techniques. Site hydrogeology, access, infrastructure, and regulatory requirements will dictate suitable PFAS cleanup options from the emerging technology portfolio.

6.3. Recent Advancement of PFAS Remediation

According to the recent literature for 2022–2023, noteworthy progress has been made in treating and remedying PFAS contamination in groundwater systems (Figure 5). It includes the development of novel adsorbent materials, the combination of new treatment processes, the emergence of in situ remediation methods, the improvement of existing technologies, the use of destructive technologies, the evaluation of sustainability, and the establishment of guidelines for passive remediation. Novel adsorbent materials, including biochar and fluoride mops, have been developed to remove PFASs from groundwater selectively. The efficacy of those materials reaches 97% [4,76]. Integrating technologies, such as using ion exchange resins with electrochemical oxidation or applying UV radiation in combination with electrochemistry, enables a more efficient approach to treating PFASs [15,77,78]. Novel in situ remediation techniques, such as colloidal activated carbon injection and electrochemical oxidation, eliminate the need for groundwater extraction [20,79]. Pilot and field studies have successfully optimized using granular activated carbon and anion exchange technologies to remove PFASs, as reported by some studies [7,72]. Sustainability is being considered via life cycle evaluations for PFAS treatment alternatives [80]. The development of guidelines for monitoring the natural attenuation of PFASs is underway, drawing upon scientific investigations conducted by [61]. In addition, performance evaluations have been carried out on developing technologies such as plasma reactors [66,81], foam fractionation, and metal–organic frameworks to remove PFASs [82]. In summary, recent research has shown advancements in novel treatment strategies, the enhancement of existing techniques, the incorporation of sustainability factors, and the implementation of passive remediation methods for groundwater polluted with PFASs.

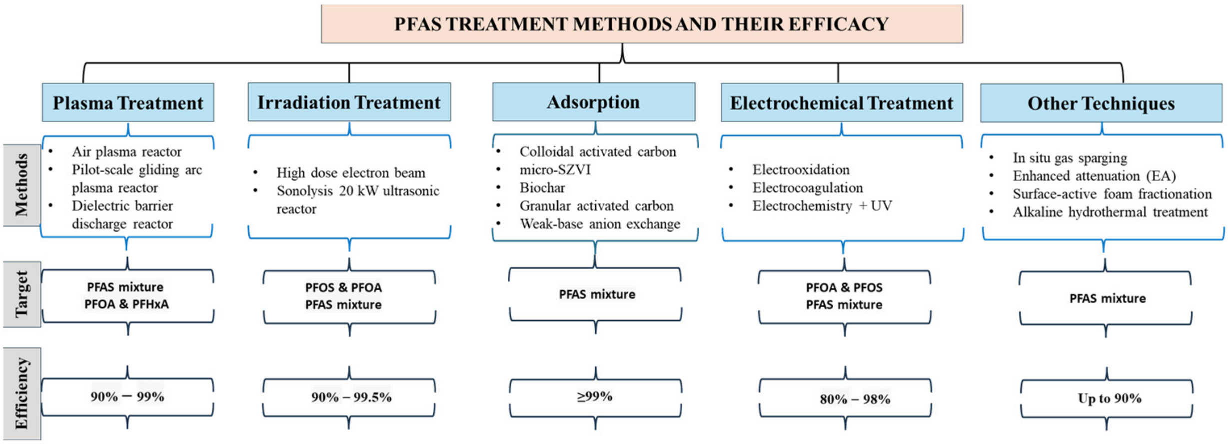

Figure 5. PFAS treatment methods and their efficiency (sourced from [7,13–15,49,63–66,73,76,78,81,83–86]).

7. PFASs in Saudi Groundwater

The case study on the occurrence of PFASs in Saudi Arabian groundwater was conducted in the Al-Qatif region of eastern Saudi Arabia. Seven groundwater samples were collected from shallow coastal aquifers with less than 1.4 m water depth. The aquifers are situated within Quaternary alluvial and Aeolian formations, which comprise layers of sand, gravel, silt, and clay overlain by tertiary carbonate rocks [87,88]. The groundwater samples were collected using 1 L amber glass vials. The samples were preserved following the guidelines set by the United States Environmental Protection Agency [40]. The analysis was conducted by ALS Arabia utilizing the USEPA method 537.1, which encompasses identifying and quantifying selected perfluorinated alkyl acids (PFAAs) and brominated compounds in drinking water [89]. The technique involves the utilization of solid phase extraction (SPE) followed by liquid chromatography–tandem mass spectrometry (LC-MS/MS) analysis, as outlined by the United States Environmental Protection Agency [90]. The PFAS analysis was subjected to quality control measures, including spikes, duplicate samples, and certified reference materials. Seventeen PFASs were analyzed, including the fluorotelomer sulfonic acids (6:2 FTS, 8:2 FTS), perfluorobutane sulfonic acid (PFBS), perfluorobutanoic acid (PFBA), perfluorodecane sulfonic acid (PFDS), perfluorodecanoic acid (PFDA), perfluorododecanoic acid (PFDoDA), perfluoroheptane sulfonic acid (PFHpS), perfluoroheptanoic acid (PFHpA), perfluorohexane sulfonic acid (PFHxS), perfluorohexanoic acid (PFHxA), perfluorononanoic acid (PFNA), perfluorooctane sulfonamide (FOSA), perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluoropentanoic acid (PFPeA), and perfluoroundecanoic acid (PFUnDA).

The concentration (ng/L) and occurrence (%) of PFASs in Saudi Arabian groundwater are illustrated in Figure 6. The analysis detected four PFASs (detection limit (DL) (10 ng/L)), including PFBS, PFBA, PFOS, and PFOA. The short-chain PFBS and PFBA display average values of 53 ng/L and 11 ng/L, respectively, with an occurrence percentage of 14%. The concentration of PFBS compounds in tested groundwater samples is below the international guidelines values [45]. PFBS is a chemical that replaced PFOS in several manufactured products such as paint, cleaning agents, food packaging, floor wax, and carpet cleaners [91]. The occurrence of PFBS below the guidelines may not pose a human risk for the coastal community in the study area. The long-chain PFOS and PFOA compounds display average concentrations of 31 ng/L and 11 ng/L, with 28% and 14% occurrence percentages, respectively. The concentration of PFOS and PFOA chemicals exceeded the maximum contamination level of 4 ng/L for each set by the USEPA [9]. Long-term exposure to PFOS and PFOA could cause concerns regarding its impact on human developmental processes, immune system functionality, liver, lipid metabolism, thyroid activity, and reproductive health [27,28]. According to a recent investigation conducted by Grandjean et al. (2022) [92], a correlation was found between exposure to perfluorooctane sulfonate (PFOS) using drinking water and a diminished immune response to vaccines in infants, even at concentrations as low as 0.01 µg/L. The PFASs detected in the study area were reported in the shallow groundwater of other countries. For example, Tang et al. (2023) [5] reported PFOS and PFOA in Changshu and Hubei Province, China, with concentrations up to 844 ng/L and 475 ng/L, respectively. In Melbourne, Australia, PFOS compounds were found in shallow wells near landfill sites with a high concentration range from 1.3 to 4800 ng/L and averaging 413.3 ng/L [34]. For the short-chain PFASs, the literature reported that the compounds most detected in groundwater are PFBA and PFBS [25,93].

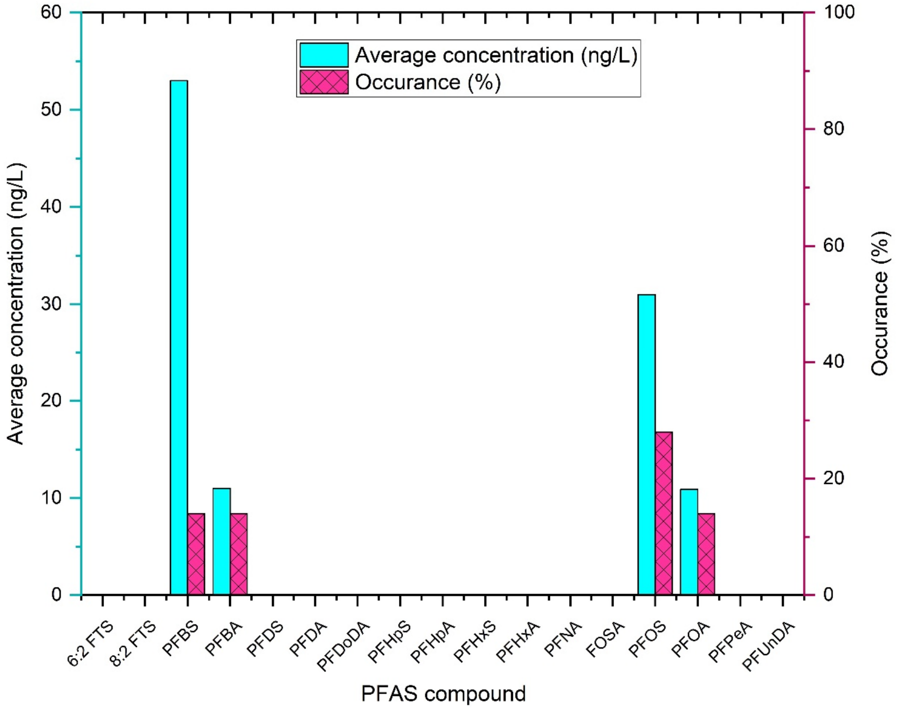

Figure 6. Concentration and occurrence of the detected PFASs in Saudi Arabian groundwater.

8. Research Needs and Future Outlook

The review analysis identified gaps for future research on PFAS occurrence, fate and transport modeling, and remediation. The critical needs include the following:

- Advanced research on PFAS contaminant leaching, attenuation, and migration processes in the subsurface environment is required to understand subsurface loading and movement better.
- Determine reliable sorption coefficients for interactions between PFASs and aquifer materials in real-world implementation.
- Enhance research to include a broader range of PFASs, especially precursor chemicals, to enhance understanding of their environmental fate and toxicity.
- To improve model accuracy, develop comprehensive datasets to validate the PFAS transport models using multi-year field data.
- Conduct performance studies of remediation technologies, especially in situ methods across multiple locations, to assess their scalability and effectiveness.
- Increase PFAS research and monitoring efforts in densely populated developing countries such as India to better assess contamination levels, exposure risks, and potential impact on public health.
- A comprehensive investigation of the PFAS contamination of the groundwater system in arid regions like Saudi Arabia and the surrounding countries is required to protect the limited freshwater resources and human health.

To effectively address these research gaps, it is essential to foster cross-disciplinary cooperation among experts in analytical chemistry, environmental engineering, hydrogeology, modeling, toxicology, and regulatory studies. Maximizing knowledge gains can be achieved through the availability and exchange of field data among investigators and various locations.

9. Conclusions

The current review provides a comprehensive analysis of recent global data on PFAS pollution in groundwater, highlighting the severe hazard these contaminants pose to drinking water, especially in developed nations. The review identified the key sources of PFASs in groundwater, including manufacturing discharges, landfill leachate, biosolids applications, and military and commercial AFFF usage, with PFOS and PFOA being the dominant compounds. Moreover, recent research showed an increasing diversity of PFASs and precursors, underscoring the need to monitor a broad range of PFASs.

This review highlighted critical research gaps that require further investigation via cross-disciplinary collaboration, particularly on topics of source delineation, subsurface processes, predictive models, and sustainable remediation technologies. Moreover, the innovative in situ detection and remediation of a wide range of PFASs need development to manage the impact of PFASs effectively. Significant advancements in PFAS remediation have been made by developing new removal adsorbents, treatment trains, and field-ready in situ technologies. However, further testing across varied locations is needed to create cost-effective solutions. Additional research needs include understanding the subsurface fate and transport of PFAS chemicals, exploring the absorption and degradation mechanisms, and the field-scale implementation of laboratory findings.

The Saudi Arabia case study presents preliminary evidence of PFAS contamination, with values above USEPA drinking water limits for PFOS and PFOA. Further monitoring and investigation are needed to better understand PFAS behavior in arid regions with limited freshwater resources. In conclusion, this review summarizes global knowledge on PFASs in groundwater, identifies significant research gaps, and recommends faster progress to protect groundwater quality and sustainability globally.

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