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Environmental Fate, Toxicity, and Mitigation of Per- and Polyfluoroalkyl Substances (PFAS): Advances in Source Reduction and Sustainable Alternatives

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ABSTRACT: Per- and polyfluoroalkyl substances (PFAS) were widely recognized as emerging environmental pollutants due to their extensive distribution across ecosystems. PFAS were synthetic chemicals composed of alkyl chains bonded to multiple fluorine atoms, and they had been detected in various environmental compartments, including rivers, soil, oceans, and the atmosphere. These compounds originated from a wide range of industrial and consumer products such as textiles, non-stick cookware, aqueous film-forming foams (AFFFs), and cosmetics. Once released into the environment, PFAS persisted for long periods and exhibited toxic effects on both ecosystems and human health. The exceptional stability of PFAS was attributed to the strong carbon–fluorine (C–F) bonds, which were among the strongest in organic chemistry due to fluorine's high electronegativity. As a result, PFAS were commonly referred to as "forever chemicals" because of their extreme resistance to degradation. Their persistence and bioaccumulative properties had made PFAS contamination a global environmental and public health concern. In response, various source reduction strategies were implemented, including the substitution of PFAS with alternative chemicals, regulatory policies, and increased consumer awareness. In parallel, green chemistry had emerged as a promising approach for developing safer and more sustainable alternatives, such as biopolymers, fluorine-free materials, and short-chain PFAS substitutes. However, further research was still required to improve the performance, safety, and scalability of these alternatives. This study aimed to discuss the sources and environmental impacts of PFAS, evaluate source reduction strategies, and examine green chemistry-based alternatives. It also identified key challenges and outlined future research directions needed to enhance the development and implementation of effective PFAS replacements.

KEYWORDS: Per- and polyfluoroalkyl substances (PFAS), emerging pollutants, environmental persistence, green chemistry alternatives, source reduction strategies

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) were a large class of synthetic chemicals characterized by alkyl chains bonded to multiple fluorine atoms, commonly represented by the general formula C_nF_{2n+1}–R, where R referred to a functional group [1]. These compounds included perfluoroalkyl substances, which were fully fluorinated carbon chains typically containing two to thirteen carbon atoms except at the terminal functional group, and polyfluoroalkyl substances, which had similar carbon backbones but were only partially fluorinated with at least one non-fluorinated bonding site [2]. Representative PFAS such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) had been widely used in industrial and consumer applications. Because of their extreme persistence, toxicity, and ability to bioaccumulate, these compounds were classified as persistent organic pollutants (POPs) and were often referred to as "forever chemicals" [3]. Their unique physicochemical properties, including hydrophobicity and oleophobicity derived from strong carbon–fluorine bonds, made them highly useful in products such as firefighting foams, non-stick cookware, food packaging, and textile coatings [4].

However, despite their industrial usefulness, PFAS had become a major environmental and public health concern. Their strong C–F bonds made them highly resistant to natural degradation processes, leading to long-term persistence in soil, water, and biota. Conventional remediation technologies such as activated carbon adsorption, ion exchange, and advanced oxidation processes had been applied to remove PFAS from contaminated environments, but these approaches were often expensive, energy-intensive, and not fully effective for a wide range of PFAS compounds [5]. As a result, the management of PFAS contamination had increasingly shifted toward prevention and source reduction strategies.

The widespread occurrence of PFAS in the environment had created a persistent contamination problem that could not be easily resolved through conventional treatment technologies alone. Although PFAS had been restricted or phased out in some applications, they continued to be released from existing stocks, industrial processes, and consumer products. The main problem was that effective, scalable, and environmentally sustainable solutions for PFAS elimination were still limited. Furthermore, many PFAS alternatives currently in use or under development lacked comprehensive evaluation regarding their long-term environmental and health impacts.

Previous review studies had extensively documented the occurrence, toxicity, and environmental fate of PFAS. However, several research gaps still existed. First, most reviews had focused on PFAS contamination and remediation technologies, while fewer had systematically integrated source reduction strategies with green chemistry principles. Second, although alternative materials such as short-chain PFAS, fluorine-free compounds, and biopolymers had been proposed, comparative evaluations of their performance, safety, and scalability remained fragmented. Third, limited attention had been given to bridging regulatory approaches with chemical innovation frameworks such as green chemistry. Finally, there was a lack of comprehensive synthesis that connected PFAS sources, environmental impacts, remediation limitations, and future sustainable design strategies within a single framework.

This review was important because it provided an integrated understanding of PFAS from multiple perspectives, including their sources, environmental impacts, remediation limitations, and emerging sustainable alternatives. By synthesizing existing knowledge, this study helped identify critical gaps in current research and highlighted the need for safer chemical design

strategies. In addition, the review emphasized the role of green chemistry as a proactive approach that shifted the focus from end-of-pipe treatment to pollution prevention. Therefore, the aim of this review was to critically examine the sources and environmental impacts of PFAS, evaluate current source reduction strategies, assess green chemistry-based alternatives, and identify key challenges and future research directions for the development of sustainable PFAS replacements. The classification of PFAS, functional groups, representative compounds, and structural descriptions is summarized in Table 1.

Table 1. Classification of PFAS, Functional Groups, Representative Compounds, and Structural Descriptions.

PFAS Class	Subgroup	Structural / Chemical Description	Reference
Perfluoroalkyl substances (PFAAs)	Perfluoroalkyl carboxylic acids (PFCAs)	Fully fluorinated carbon chain terminated with a carboxylic acid group; highly persistent and water soluble	[6]
	Perfluoroalkyl sulfonic acids (PFSAs)	Strong acid functional group with sulfonate head; high bioaccumulation potential	[6]
	Perfluoroalkyl sulfonyl fluorides (PASFs)	Reactive precursor compounds used in industrial synthesis of PFSAs	[7]
	Perfluoroalkane sulfonamides (FASAs)	Amide-functional PFAS that can transform into PFSAs in the environment	[7]
	Perfluoroalkyl phosphonic acids (PFPAs)	Phosphonic acid–functional PFAS with strong surface activity and persistence	[7]
	Perfluoroalkyl phosphinic acids (PFPIAs)	Bifunctional phosphorus-containing PFAS with high chemical stability	[7]
	Perfluoroalkyl ether sulfonic acids (PFESAs)	Ether-linked structure that increases mobility and environmental persistence	[7]
	Perfluoroalkyl ether carboxylic acids (PFECAs)	Ether-based replacement PFAS designed as alternatives to legacy compounds	[8]
Polyfluoroalkyl substances	Polyfluoroalkyl phosphate esters (PAPs)	Partially fluorinated phosphate esters used in surface treatment applications	[8]
	Fluorotelomer alcohols (n:2 FTOHs)	Precursor compounds that degrade into persistent PFAS in the environment	[8]
	Fluorotelomer sulfonic acids (n:2 FTSAs)	Semi-fluorinated sulfonic acids with transformation potential into PFSAs	[7]
	Fluorotelomer carboxylic acids (n:2 FTCAs)	Intermediate degradation products of fluorotelomer-based PFAS	[7]

3. Major Environmental Sources of PFAS Emissions

PFAS are released into the environment through multiple intentional-use pathways across industrial, commercial, and consumer sectors. These sources contribute significantly to their widespread distribution in water, soil, air, and biota, resulting in persistent environmental contamination.

3.1. Firefighting Foams in Industrial snd Military Applications.

Aqueous film-forming foams (AFFFs) are widely used as firefighting agents for extinguishing hydrocarbon-based fuel fires in high-risk environments such as airports, oil refineries, and military installations [9]. When applied, AFFFs spread rapidly over fuel surfaces to form a thin aqueous film that reduces surface tension and creates a barrier between the fuel and oxygen, thereby suppressing ignition and preventing flame re-ignition [2]. Despite their effectiveness, AFFFs have been identified as a major source of PFAS contamination due to the inclusion of

fluorinated surfactants. Electrochemical fluorination (ECF)-based formulations contain high levels of perfluorooctane sulfonate (PFOS), with reported median concentrations of approximately 4900 mg/L and a detection frequency of 100%. Fluorotelomer (FT)-based AFFFs contain alternative PFAS compounds such as 6:2 fluorotelomer thioether amido sulfonates (6:2 FtTAoS) and 6:2 fluorotelomer sulfonamidoalkyl betaine (6:2 FTAB), with median concentrations ranging from 3188 mg/L to 7100 mg/L and detection frequencies between 45.8% and 68.2% [9]. The long-term use of AFFFs has resulted in extensive contamination of soil and groundwater, particularly in and around military training sites and fire suppression facilities. The United States Department of Defense (DoD) has reported PFAS contamination concerns across 263 of 391 installations, representing approximately 67% of its total sites, with an estimated expenditure of USD 200 million allocated for investigation and remediation activities [10].

3.2. Food Packaging and Food-Contact Applications.

Food contact materials (FCMs) constitute another significant pathway for PFAS entry into the environment and human exposure. These materials include fast-food wrappers, grease-resistant paper packaging, takeout containers, disposable plates, and certain types of cookware. PFAS are incorporated into these products to impart grease resistance, water repellency, and non-stick properties. A critical concern associated with FCMs is the migration of PFAS from packaging materials into food. This migration is influenced by several factors, including temperature, contact duration, PFAS concentration, molecular structure, and chain length [11]. Elevated temperatures, particularly in hot or fatty foods, significantly enhance migration rates, increasing potential dietary exposure. Long-chain PFAS such as PFOS and PFOA exhibit greater persistence and bioaccumulation potential compared to short-chain analogues, making them more environmentally and biologically concerning [12, 13]. In addition to packaging, PFAS are also widely used in non-stick cookware coatings, most notably polytetrafluoroethylene (PTFE), commonly known as Teflon. While PTFE provides excellent non-stick performance, mechanical abrasion and high-temperature exposure can degrade the coating, leading to particle release [14]. Scratching or overheating non-stick surfaces may therefore contribute to contamination of food with PTFE-derived particles, raising potential health concerns upon ingestion [15].

3.3. Textile Manufacturing and Finishing Processes.

The textile industry is another major contributor to PFAS emissions due to their extensive use in fabric production and finishing processes. PFAS compounds such as fluorotelomer alcohols (FTOHs), perfluorocarboxylic acids (PFCAs), fluorotelomer carboxylic acids (FTCAs), and fluorotelomer sulfonic acids (FTSAs) are commonly applied as surfactants, emulsifiers, dyeing auxiliaries, and surface treatment agents [16]. These compounds are widely used in performance textiles, including sportswear, outdoor apparel, waterproof jackets, tents, and backpacks, where they provide water, oil, and stain resistance [17]. PFAS are introduced at multiple stages of textile production. During yarn and fabric processing, they are used as lubricants to reduce friction and facilitate weaving operations, while also improving dye absorption efficiency [18]. Certain PFAS compounds, such as perfluorododecanoic acid (PFDoDA), have also been directly used as dye components in textile formulations [19]. In dyeing processes, PFAS may also function as foam suppressants in systems involving sulfur-

based dyes, improving process stability. Following dyeing, additional finishing treatments often reapply PFAS-based coatings to enhance durable water-repellent properties and improve resistance to oil, stains, and moisture [20, 21]. These repeated applications across production stages contribute to PFAS release into wastewater streams, making textile manufacturing a continuous source of environmental emissions.

4. Environmental and Health Impacts of PFAS

4.1. Aquatic Environmental Contamination and Ecotoxicological Effects.

PFAS enter aquatic environments primarily through the discharge of industrial effluents, municipal wastewater, and atmospheric deposition. Once introduced, these compounds are highly mobile in aquatic systems and can persist in surface water bodies such as rivers, lakes, and marine environments. Due to their stability and amphiphilic properties, PFAS readily interact with aquatic organisms through direct ingestion, respiratory uptake, or surface adsorption onto biological tissues. Exposure to PFAS has been associated with both non-specific systemic toxicity and organ-specific toxicological effects in aquatic organisms [22]. A key mechanism underlying non-target toxicity is oxidative stress, which occurs when the production of reactive oxygen species (ROS) exceeds the capacity of biological antioxidant defense systems [23]. ROS include highly reactive molecules such as superoxide radicals ($O_2^{\bullet -}$), hydroxyl radicals (•OH), and nitric oxide radicals (NO•) [25]. Excess ROS generation disrupts cellular homeostasis and leads to lipid peroxidation, protein oxidation, DNA damage, and apoptosis [24].

In aquatic species such as tilapia, PFAS exposure has been shown to elevate antioxidant enzyme activities, including superoxide dismutase (SOD), catalase (CAT), and lipid peroxidation levels [26]. In zebrafish embryos, PFAS exposure alters the expression of genes associated with nuclear respiratory factors (NRF-1 and NRF-2), kinase signaling pathways, apoptosis regulation, and mitogen-activated protein kinase (MAPK) transcriptional networks [22]. Furthermore, exposure to perfluorooctane sulfonate (PFOS) significantly modified insulin receptor gene expression in zebrafish, resulting in disrupted glucose metabolism and insulin resistance [27]. In addition to systemic toxicity, PFAS induce organ-specific effects, particularly endocrine and thyroid disruption. These compounds can interact with the hypothalamic–pituitary–gonadal (HPG) axis and associated receptors, thereby altering hormonal regulation [23]. For example, exposure of zebrafish to perfluorononanoic acid (PFNA) at 1 mg/L significantly altered the transcription of genes involved in follicle-stimulating hormone receptors, luteinizing hormone receptors, and steroidogenic acute regulatory proteins [28]. Similarly, PFDoA exposure has been shown to suppress thyroid hormone precursors such as thyrotropin-releasing hormone and corticotropin-releasing hormone, while increasing iodothyronine deiodinase activity, leading to elevated triiodothyronine (T3) levels and developmental abnormalities such as skeletal and fin malformations [29, 30].

Environmental monitoring studies have also reported widespread PFAS contamination in aquatic systems. In Malaysia's Klang Valley, higher serum PFAS concentrations were observed in males compared to females, with increasing levels correlated with age [31]. The Langat River, which supplies drinking water to more than 27% of the Klang Valley population, has been identified as one of the most polluted rivers, with PFOS and PFOA concentrations

reaching 43.5 ng/mL and 5.94 ng/mL, respectively [32]. In Singapore, coastal waters exhibited lower PFAS concentrations compared to surface and wastewater sources, although industrialized areas such as the Johor Strait showed elevated contamination levels [33]. In Sweden, PFAS contamination in drinking water affected approximately 300,000 people, with reported concentrations exceeding 90 ng/L in 35% of water sources, a level considered unsuitable for consumption [34].

4.2. Human Toxicological Effects Of PFAS Exposure.

4.2.1. Dysregulation of Lipid Metabolism and Hypercholesterolemia.

PFAS interact with biological systems by integrating into phospholipid bilayers and binding to various proteins, including serum albumin, lipoproteins, organic anion transporters (OATs), and fatty acid-binding proteins [35]. The binding affinity of PFAS is influenced by hydrophobic interactions and electrostatic complementarity between PFAS functional groups and amino acid residues at protein binding sites [36]. Studies have shown that perfluorooctane sulfonate (PFOS) binds to apolipoprotein A-I (ApoA-I), a major component of high-density lipoprotein (HDL), thereby impairing lipid-binding capacity and disrupting HDL function [37]. This interference reduces the activity of lecithin–cholesterol acyltransferase (LCAT), an enzyme responsible for converting free cholesterol into cholesterol esters. Consequently, cholesterol metabolism is inhibited, leading to elevated circulating cholesterol levels and dyslipidemia.

4.2.2. Carcinogenicity and Mechanistic Pathways of Cancer Development.

PFAS exposure has been associated with carcinogenic outcomes through multiple biological mechanisms, including oxidative stress, endocrine disruption, epigenetic modification, and immune dysregulation [38]. Excessive ROS generation overwhelms endogenous antioxidant systems such as catalase (CAT), superoxide dismutase (SOD), and glutathione peroxidase (GPx), resulting in oxidative damage to lipids, proteins, and nucleic acids [39, 40]. Endocrine disruption represents another major pathway, where PFAS interfere with hormone signaling by mimicking or blocking natural hormones, thereby altering receptor binding and disrupting cellular proliferation and differentiation processes [41, 42]. During critical developmental stages, PFAS exposure can impair testicular development by disrupting Sertoli cell function (spermatogenesis) and Leydig cell steroidogenesis, ultimately affecting testosterone synthesis [43, 44].

Epigenetic alterations also play a significant role in PFAS-induced carcinogenesis. DNA methylation, regulated by DNA methyltransferases (DNMT1, DNMT3A, DNMT3B) and ten-eleven translocation enzymes (TET1–TET3), involves methyl group addition to cytosine residues in CpG sites [45]. Exposure to PFOA has been shown to alter DNMT3A mobility at concentrations above 100 μM [46]. Genome-wide methylation studies identified PFAS-associated modifications at 39 CpG sites linked to neurodevelopmental pathways [47]. Furthermore, increased DNA methylation in peripheral blood leukocytes has been associated with elevated breast cancer risk in epidemiological studies [48]. PFAS also induce chronic inflammation through immune activation pathways. They stimulate cytokine release, activating T-helper cells and promoting further production of pro-inflammatory mediators and ROS [49]. Mechanistically, PFOA activates Toll-like receptors (TLR2 and TLR4), leading to NF-κB

activation, which regulates inflammatory gene expression [50, 51]. This cascade promotes inflammasome formation via NLRP3, sustaining chronic inflammatory states linked to carcinogenesis.

4.2.3. Thyroid Dysfunction and Hormonal Imbalance.

Thyroid hormone synthesis relies on thyroglobulin, produced in follicular thyroid cells, which serves as a precursor for thyroxine (T4) and triiodothyronine (T3) [52]. These hormones are transported in the bloodstream and regulated through thyroid-stimulating hormone (TSH) via a negative feedback mechanism [53]. PFAS with longer carbon chain lengths (≥C8) exhibit stronger binding affinity to thyroid transport proteins through hydrogen bonding interactions. Sulfonated PFAS demonstrate higher binding energies compared to carboxylated PFAS due to stronger interactions between fluorinated tails and hormonogenic residues, which disrupt T4 synthesis [52]. Epidemiological evidence indicates that PFAS exposure is positively associated with circulating T4 levels, although relationships with TSH remain inconsistent [53].

4.3. PFAS Uptake and Accumulation in Agricultural Systems.

Plants can accumulate PFAS through root uptake and foliar absorption, leading to internal translocation within plant tissues [54]. The extent of uptake is influenced by root protein content, lipid composition, and PFAS physicochemical properties. Long-chain PFAS generally exhibit higher adsorption to root membranes, resulting in reduced translocation but increased root accumulation. Experimental evidence shows that mung bean plants exhibit high PFAS uptake due to elevated root protein content, with PFOS demonstrating stronger accumulation than PFOA because of higher protein binding affinity [55]. Additionally, zwitterionic PFAS compounds may enhance adsorption through interactions with phospholipid membranes, increasing overall uptake efficiency [56]. Food chain contamination studies further highlight PFAS presence in edible crops. Piva et al. [57] reported higher PFAS concentrations in ready-to-eat vegetables (0.13 ng/g) compared to fresh (0.07 ng/g) and frozen samples (0.06 ng/g), likely due to processing and packaging exposure. Structural plant barriers such as the Casparian strip limit PFAS translocation, particularly for high-molecular-weight compounds, but do not completely prevent accumulation [54]. Consequently, contaminated agricultural products represent a potential dietary exposure pathway for humans.

5. Strategies for PFAS Source Reduction

5.1. Chemical Substitution and Material Replacement Strategies.

Chemical substitution represents the most direct and widely adopted source reduction strategy for PFAS, as it aims to eliminate PFAS usage at the production stage. This approach aligns closely with green chemistry principles by prioritizing the development of safer materials that retain comparable functional performance while minimizing environmental persistence and toxicity. Instead of relying on fluorinated compounds, research has focused on identifying viable substitutes that can replicate key properties such as water, oil, and stain resistance. Current categories of PFAS alternatives include biopolymer-based materials, fluorine-free synthetic compounds, and short-chain PFAS substitutes [5]. Biopolymers such as cellulose and chitosan offer biodegradable properties, while fluorine-free alternatives aim to replace surface-active functions using hydrocarbon- or silicone-based chemistries. Short-chain PFAS have also

been introduced as transitional alternatives, although their environmental safety remains under evaluation. These material classes are further discussed in subsequent sections.

5.2. Regulatory Frameworks and Policy Interventions.

Regulatory actions at national and international levels have played a critical role in restricting and phasing out PFAS usage across various sectors. In the United States, the Environmental Protection Agency (EPA) introduced regulatory oversight of PFAS in drinking water through the National Primary Drinking Water Regulation (NPDWR), initially setting advisory limits for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS). Early guidelines recommended a threshold of 70 ng/L for both compounds; however, updated assessments in 2022 significantly lowered advisory values, reflecting increasing concern over their toxicity. Under the revised framework, the EPA established maximum contaminant levels (MCLs) of 4 ng/L for PFOA and PFOS, 2000 ng/L for PFBS, and 10 ng/L for PFNA, PFHxS, and GenX compounds [59]. These regulatory limits highlight the tightening control of PFAS exposure in drinking water systems. In parallel, the European Chemicals Agency (ECHA), supported by countries such as Norway, Germany, Sweden, Denmark, and the Netherlands, proposed a comprehensive restriction on the manufacture, use, sale, and import of PFAS under the European Commission framework in March 2023 [60]. This proposal represents one of the most extensive regulatory efforts targeting the entire PFAS class.

At the state level in the United States, stricter regulations have been introduced, particularly in the textile and consumer goods sectors. For example, Maine's Law LD 1503 enforces a phased prohibition of PFAS in most products by 2030, with exceptions only for essential uses approved by the Department of Environmental Protection (DEP) [61]. Maine also banned PFAS-containing food contact materials as early as 2020. Similarly, New York's Law S6291/A7063 mandates a ban on clothing containing PFAS beginning in December 2023 [62]. California has implemented progressive limits, restricting intentionally added PFAS in textiles to 100 ppm by 2025 and further reducing the threshold to 50 ppm by 2027 [63]. These regulatory measures collectively demonstrate increasing global momentum toward PFAS restriction.

5.3. Role of Consumer Behavior in Reducing PFAS Exposure.

Consumer behavior also plays an important role in reducing PFAS usage and exposure through informed purchasing decisions and lifestyle modifications. One of the major consumer exposure pathways is non-stick cookware, where PFAS-based coatings are used to prevent food adhesion and facilitate cleaning. However, degradation of these coatings during cooking or washing processes can lead to PFAS release into food and wastewater systems [64]. To reduce exposure, consumers are encouraged to select alternative cookware materials such as ceramic, cast iron, carbon steel, or stainless steel, which do not rely on fluorinated coatings and are generally more stable under high-temperature conditions [65].

Another important exposure source is food packaging, particularly fast-food containers and single-use materials that are treated with grease-resistant PFAS coatings. These coatings prevent oil and moisture penetration but can migrate into food under certain conditions, especially at elevated temperatures or during prolonged contact with fatty foods [66]. As a result, consumers can reduce exposure by limiting reliance on heavily packaged or processed fast foods, choosing fresh or minimally packaged alternatives, and using reusable containers

made from safer materials. In addition, selecting certified PFAS-free products in categories such as cosmetics, textiles, cookware, and food packaging can significantly reduce indirect exposure pathways [67]. The PFAS source reduction strategies, mechanisms, and key examples are summarized in Table 2.

Table 2. PFAS Source Reduction Strategies, Mechanisms, and Key Examples.

Strategy Category	Key Approach	Description	Examples / Applications	Key Notes	Ref.
Chemical substitution and material replacement	Replacement of PFAS with safer functional materials	Eliminates PFAS at the production stage by replacing fluorinated compounds with alternative materials that provide similar functional properties	Biopolymers (cellulose, chitosan, starch), fluorine-free materials (hydrocarbon- and silicone-based systems), short-chain PFAS substitutes	Biopolymers are biodegradable; fluorine-free systems reduce persistence; short-chain PFAS are transitional but still environmentally concerning	[5]
Regulatory frameworks and policy interventions	Government and international restrictions on PFAS use	Establishes legal limits, bans, and phase-outs of PFAS in drinking water, industrial use, textiles, and consumer products	EPA drinking water limits (PFOA 4 ng/L, PFOS 4 ng/L); EU-wide PFAS restriction proposal; state laws in Maine, New York, and California	Strong global regulatory momentum; focuses on reducing exposure and industrial usage across sectors	[59–63]
Consumer behavior and product choice	Reduction of PFAS exposure through purchasing decisions	Encourages individuals to avoid PFAS-containing products and reduce exposure pathways through lifestyle choices	Use of ceramic/cast iron/stainless steel cookware; reduced use of fast-food packaging; selection of PFAS-free labeled products	Consumer action reduces exposure but cannot fully eliminate environmental PFAS contamination	[64–67]

6. Alternative Materials for PFAS Replacement

6.1. Biopolymer-Based Functional Materials.

Biopolymers have received increasing attention as environmentally favorable alternatives to PFAS due to their renewable origin and biodegradability. Common examples include chitosan, cellulose, and starch [68]. Chitosan is a deacetylated derivative of chitin and is the only naturally occurring polycation. Its charge density is governed by the pH of the surrounding medium and the degree of acetylation [69]. Structurally, chitosan contains amino ($–NH_2$) and hydroxyl (–OH) functional groups, which enable strong intermolecular interactions. Although chitosan is originally hydrophilic, its surface properties can be modified to increase hydrophobicity through methods such as lactic acid hydrolysis, radio-frequency (RF) plasma treatment, and esterification, which increase the water contact angle [70, 71]. For cellulose-based materials, several modification techniques, including cellulase pretreatment, oxalic acid treatment, and poly-D-lactic acid (PDLA) grafting, have been used to develop superhydrophobic coatings on paper-based substrates, improving resistance to water and grease [68]. In PDLA grafting, cellulose microcrystals (CMC) interact with poly-L-lactic acid (PLLA) to form stereocomplex polylactic acid (PLA), resulting in enhanced water and oxygen barrier properties due to a more ordered molecular structure and reduced intermolecular spacing [72]. The cellulase pretreatment mechanism is illustrated in Figure 1. Oxalic acid modification involves the reaction between cellulose hydroxyl groups and oxalic acid to form ester bonds [73]. This esterification reduces the number of hydrophilic hydroxyl groups, thereby improving

water resistance. Cellulase treatment is a biological process in which cellulase enzymes partially hydrolyze the cellulose surface, generating micro- and nanoscale roughness [74]. This surface structure enhances hydrophobicity through the Cassie–Baxter effect, where trapped air pockets reduce liquid–solid contact [75]. As a result, water droplets bead up and easily roll off the surface.

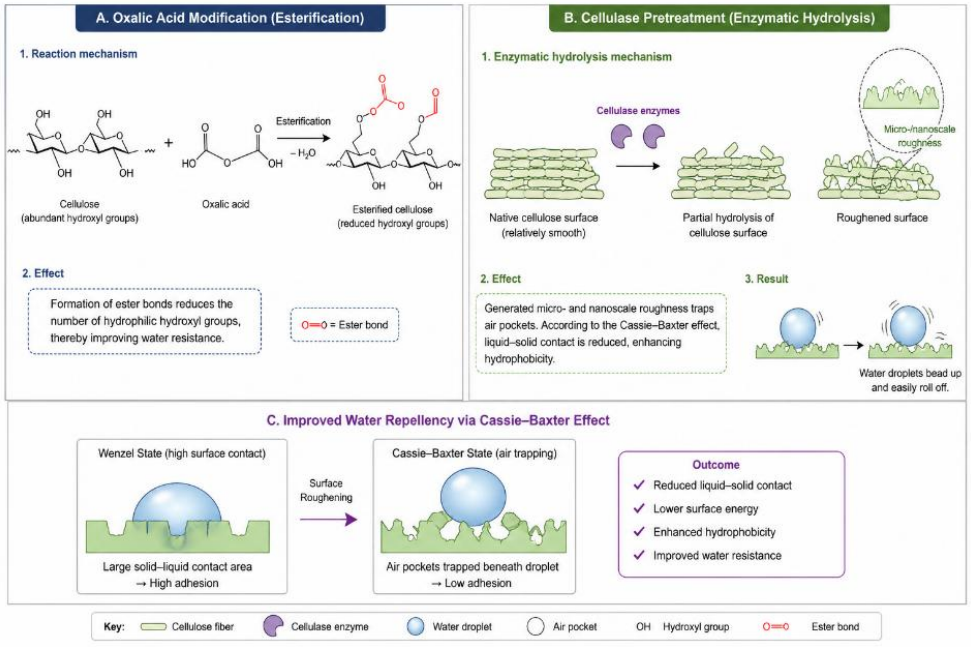

Figure 1. Mechanistic Illustration of Cellulose Surface Modification.

Edible coatings and films (ECFs), typically derived from polysaccharide-based biopolymers such as chitosan, cellulose, starch, alginate, and pectin, have emerged as promising sustainable alternatives to PFAS-containing food packaging materials [76, 77]. These thin biopolymer layers are applied directly to food surfaces to form a semi-permeable barrier that delays quality deterioration during storage. By reducing oxygen diffusion, moisture transfer, lipid oxidation, and microbial contamination, ECFs help maintain the sensory attributes, nutritional value, and overall quality of fresh produce, meat, seafood, dairy products, and bakery items, thereby extending product shelf life [76, 77]. In addition to serving as passive protective barriers, edible coatings can function as active packaging systems by incorporating natural antimicrobial agents, antioxidants, essential oils, probiotics, enzymes, or bioactive compounds that further improve food safety and preservation [76–79].

An important advantage of ECFs is that they are biodegradable, renewable, and generally recognized as safe for human consumption because they are produced from non-toxic and food-grade materials [78, 79]. Unlike fluorinated coatings, which persist in the environment and may pose potential health risks due to their persistence and bioaccumulation, edible biopolymer coatings naturally degrade after disposal and do not generate persistent pollutants. Consequently, their adoption supports the transition toward more sustainable food packaging systems and contributes to reducing plastic waste and PFAS contamination associated with conventional packaging materials.

Table 3 summarizes representative biopolymer systems, their modification strategies, and the resulting functional properties that have been developed to improve their suitability as PFAS alternatives. Various physical, chemical, and biological modification techniques,

including enzymatic treatment, plasma surface modification, esterification, polymer grafting, and blending with complementary biopolymers, have been employed to enhance water resistance, hydrophobicity, oxygen barrier performance, mechanical strength, and antimicrobial activity [68–75]. These modifications demonstrate that the functional performance of natural biopolymers can be substantially improved while maintaining their biodegradability and environmental compatibility.

Table 3. Biopolymers as Sustainable Alternatives to PFAS: Types, Modifications, and Functional Properties.

Biopolymer Type	Source / Nature	Modification / Technology	Resulting Functional Properties	Application / Description	Reference
Chitosan	Deacetylated derivative of chitin; natural polycation	Lactic acid hydrolysis, RF plasma treatment, esterification	Conversion from hydrophilic to hydrophobic; increased water contact angle	Used for surface coating and functional materials due to strong intermolecular interactions and tunable surface charge	[69–71]
Cellulose	Natural polysaccharide from plants	Cellulase pretreatment, oxalic acid modification, PDLA grafting	Improved water and grease resistance; enhanced hydrophobicity; increased barrier properties	Paper-based superhydrophobic coatings, food packaging enhancement	[68, 72–75]
Starch	Natural polysaccharide from plants	Blending and chemical modification (general use)	Improved film-forming ability and biodegradability	Used in biodegradable coatings and packaging materials	[68]
Poly-D-lactic acid (PDLA)-modified cellulose	Synthetic-biopolymer hybrid system	Grafting with PDLA to form stereocomplex PLA with PLLA	Increased water and oxygen barrier due to dense crystalline structure	High-performance biodegradable packaging materials	[72]
Cellulase-treated cellulose	Enzyme-modified cellulose	Enzymatic hydrolysis (cellulase treatment)	Formation of micro/nano roughness; enhanced hydrophobicity via Cassie–Baxter effect	Water-repellent surfaces for packaging applications	[74, 75]
Oxalic acid–modified cellulose	Chemically modified cellulose	Esterification with oxalic acid	Reduced hydrophilicity; improved water resistance due to decreased hydroxyl content	Hydrophobic coatings for paper and packaging materials	[73]
Edible coatings and films (ECFs)	Biopolymer-based edible films (mainly polysaccharides)	Film casting and blending with additives	Biodegradable, non-toxic, antimicrobial barrier properties; limited mechanical strength	Food preservation, shelf-life extension, oxidation and moisture barrier	[76–79]

6.2. Fluorine-Free Surface and Coating Systems.

Fluorine-free materials have been proposed as alternative systems to replace PFAS by eliminating fluorinated carbon chains while attempting to maintain surface functionality. Among these, hydrocarbon- and silicone-based coatings have been widely studied due to their lower toxicity and reduced environmental persistence. According to Lei et al. [80], silicone materials combined with surface roughness engineering can achieve superhydrophobicity through low-surface-energy methyl groups and siloxane (Si–O) backbones. The surface is enriched with non-polar methyl groups, which reduce surface tension. Similar to cellulase-treated cellulose systems, these structures generate the Cassie–Baxter effect [75], enabling water contact angles exceeding 150°, allowing droplets to roll off easily. Silicone-based

materials are commonly used in waterproof textiles such as swimwear and swimming equipment.

In addition, hydrocarbon-based surfactants reduce surface tension through van der Waals interactions; however, their oil-repellent performance is lower compared to PFAS [81, 82]. Highly branched hydrocarbon surfactants can achieve relatively low surface tension due to methyl group-rich structures with low surface energy [83]. García et al. [84] reported that hydrocarbon-based surfactants used in aqueous film-forming foams (AFFFs) are aerobically biodegradable in groundwater but show limited degradation under anaerobic conditions. The combination of silicone- and hydrocarbon-based surfactants represents a promising direction for developing fluorine-free foams for liquid fuel fire suppression [85].

6.3. Short-Chain Fluorinated Substitutes.

Short-chain fluorinated compounds have increasingly been adopted as industrial alternatives to long-chain PFAS due to regulatory restrictions and concerns regarding bioaccumulation. These compounds typically contain six or fewer carbon atoms and are used in applications such as aqueous film-forming foams (AFFF), where rapid vapor suppression and thermal stability are required. Modern fluorotelomer-based AFFF formulations may contain 50–98% short-chain fluorinated compounds, often alongside residual long-chain PFAS precursors, indicating a transitional substitution strategy rather than complete elimination of fluorinated chemistry [87].

A widely studied example is hexafluoropropylene oxide dimer acid (GenX), which was introduced as a replacement for perfluorooctanoic acid (PFOA). GenX exhibits shorter biological half-lives and lower bioaccumulation potential compared to legacy PFAS, initially suggesting improved environmental safety [88, 89]. However, emerging toxicological evidence indicates that GenX may exhibit comparable or even higher toxicity and environmental persistence than PFOA, raising concerns about its suitability as a safer alternative [90, 91]. Compared with non-fluorinated alternatives, short-chain PFAS still provide superior surface tension reduction and oil–water repellency. However, their environmental behavior remains problematic due to high mobility in aquatic systems and extreme persistence driven by strong carbon–fluorine bonds. This has led to concerns regarding “regrettable substitution,” where restricted chemicals are replaced with structurally similar compounds that later exhibit similar environmental risks.

In contrast, biopolymers, silicone-based systems, and hydrocarbon coatings generally demonstrate improved environmental profiles but suffer from functional limitations such as reduced oleophobicity, lower durability, and weaker chemical resistance. This highlights a key challenge in PFAS replacement: achieving an optimal balance between performance and environmental safety remains unresolved (Table 4).

Table 4. Comparative Properties, Advantages, and Limitations of PFAS Alternatives Including Short-Chain PFAS.

Alternative Class	Key Examples	Performance	Characteristics	Reference
Biopolymers	Chitosan, cellulose, starch	High surface energy; poor hydrophobicity and oleophobicity	Renewable, biodegradable, and generally non-toxic. Sustainable and widely available but require chemical or physical modification.	[92,93]
Silicone-based materials	Polydimethylsiloxane (PDMS) and derivatives	Low surface energy; good hydrophobicity but poor oleophobicity	Low toxicity and excellent thermal stability, flexibility, and water repellency.	[80,94]
Hydrocarbon-based materials	Alkanes, waxes, hydrocarbon coatings	Moderate surface energy; moderate hydrophobicity and poor oleophobicity	Lower persistence and toxicity than fluorinated materials, with relatively low cost.	[94,95]
Short-chain PFAS	GenX (HFPO-DA), C6 fluorotelomers	Very low surface energy; excellent hydrophobicity and good oleophobicity	Lower bioaccumulation than long-chain PFAS while maintaining high performance.	[87–91,95]

7. Key Challenges and Future Research Directions in PFAS Replacement

7.1. Performance Limitations And Environmental Trade-offs of PFAS Alternatives.

The substitution of PFAS with alternative materials remains challenging due to the difficulty in replicating their exceptional surface-active properties. PFAS are characterized by extremely low surface energy, which results in superior hydrophobic and oleophobic performance [4]. As reported by Glüge et al. [4], PFAS-based fluorosurfactants can reduce the surface tension of water to below 16 mN/m, which is approximately half the minimum achievable by hydrocarbon-based surfactants. Although fluorine-free coatings have demonstrated promising water-repellent performance in textile applications, their durability remains limited. In particular, their water-repellent properties often degrade after repeated washing cycles due to weak adhesion to fiber surfaces [96]. This limitation restricts their long-term applicability in high-performance textile systems.

Similarly, fluorine-free firefighting foams have been developed to reduce PFAS-related environmental contamination. However, emerging studies suggest that some of these alternatives may still exhibit toxicity toward aquatic organisms, highlighting the need for comprehensive environmental risk assessments prior to large-scale adoption [97]. Short-chain PFAS have also been introduced as transitional substitutes; however, their use presents the risk of "regrettable substitution" [98]. Due to their higher water solubility and polarity compared to long-chain PFAS, these compounds exhibit lower hydrophobic interactions with water molecules, allowing them to dissolve more readily and migrate faster through soil and aquatic systems. Furthermore, their high mobility and chemical stability make them difficult to remove using conventional adsorption and water treatment technologies [99].

7.2. Material Design, Sustainability Assessment, and Environmental Uncertainty.

A major challenge in developing PFAS alternatives lies in achieving comparable multifunctional performance. Current alternatives often fail to simultaneously match PFAS in terms of hydrophobicity, oleophobicity, thermal stability, and mechanical durability. One promising approach is the development of composite materials that integrate multiple functional properties, including low surface energy, high mechanical strength, and thermal resistance [96]. The incorporation of nanoparticles into composite systems has been proposed as an effective strategy to enhance multifunctionality. For example, zinc oxide (ZnO) nanoparticles provide antibacterial properties, while silicon dioxide (SiO_2) nanoparticles improve wear resistance and mechanical durability [97, 98]. Such hybrid systems may offer improved performance compared to single-component alternatives.

In addition, future research must prioritize comprehensive life cycle assessment (LCA) to evaluate the true environmental impact of PFAS alternatives. Substitutes that require higher energy input, consume more resources, or generate hazardous byproducts may not provide a net environmental benefit despite eliminating fluorinated chemistry. Therefore, sustainability assessments must integrate environmental, economic, and social dimensions to ensure balanced decision-making [99]. Another critical research gap involves understanding the environmental fate and long-term behavior of emerging PFAS alternatives [98, 99]. Although many bio-based and fluorine-free materials are promoted as safer options, their degradation pathways, persistence, and ecological impacts remain insufficiently studied. Without long-term environmental evaluation, there is a significant risk of unintended consequences, including the emergence of new pollutants through regrettable substitution.

8. Conclusion

Per- and polyfluoroalkyl substances (PFAS) have remained a major global environmental concern due to their extensive use, extreme persistence, and ability to accumulate in natural and biological systems. Their strong carbon–fluorine bonds make them highly resistant to degradation, resulting in widespread distribution across water, soil, air, and even remote regions such as the Arctic and Antarctic. Consequently, PFAS contamination has become a long-term issue affecting ecosystem stability, wildlife health, and human exposure.This review has shown that PFAS originate from several major sources, including aqueous film-forming foams (AFFFs), food contact materials, and textile manufacturing processes. These sources continuously release PFAS into the environment through industrial applications and consumer product use. The associated impacts are severe and include oxidative stress, endocrine disruption, metabolic disorders, carcinogenic effects, thyroid dysfunction, and uptake into agricultural systems, demonstrating their broad toxicological and ecological significance. To address these issues, multiple source reduction strategies have been implemented, including chemical substitution, regulatory controls, and consumer-driven actions. Among these, substitution has been widely explored through the development of biopolymers, fluorine-free materials, and short-chain fluorinated compounds. Biopolymers such as cellulose, chitosan, and starch provide renewable and biodegradable options, while fluorine-free coatings based on silicone and hydrocarbons offer lower environmental persistence. However, short-chain PFAS, although initially considered safer, still raise concerns due to their high mobility, persistence, and potential for “regrettable substitution.” Despite progress in material development,

significant challenges remain. Most PFAS alternatives still exhibit lower hydrophobicity, oleophobicity, and durability compared to conventional PFAS. In many cases, achieving comparable performance requires complex modifications or composite systems, which may limit scalability and industrial adoption. In addition, the long-term environmental behavior and toxicity of many alternatives remain insufficiently understood. Future research should focus on enhancing the performance and sustainability of PFAS alternatives through advanced material design, including composite and nanostructured systems, while ensuring environmental safety through comprehensive life cycle assessments. Long-term studies on degradation pathways, toxicity, and ecological fate are also essential to avoid unintended environmental consequences. Overall, reducing PFAS pollution requires continued scientific innovation, improved material design, and effective regulatory and societal engagement to achieve a balance between functional performance and environmental sustainability.

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Author Contributions

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Competing Interests

The authors declare no competing interests.

Data Availability

All data supporting the findings of this study are available within the article and its supplementary materials. Additional data are available from the corresponding author upon reasonable request.

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