

# “Forever chemicals”: a sticky environmental problem

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## HIGHLIGHTS

- “Forever chemicals” are being redefined in terms of environmental lifespans.
- Novel degradation technologies offer promising PFAS remediation solutions.
- Global Collaboration in responding to the PFAS crisis is emphasized.

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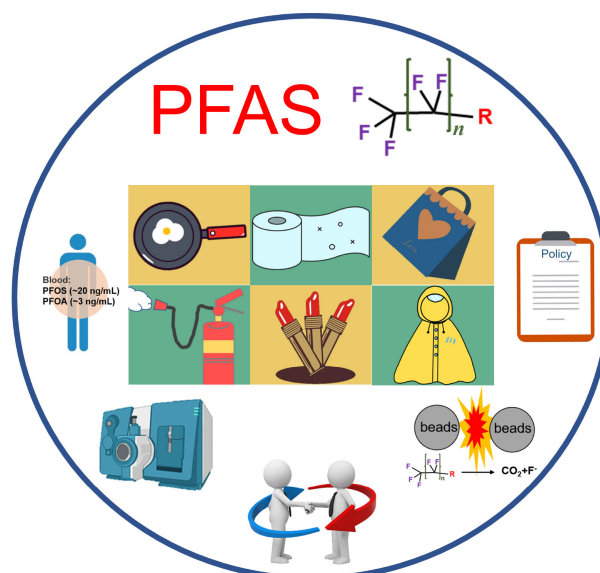
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## GRAPHIC ABSTRACT



## ABSTRACT

The discovery and widespread use of per- and poly-fluoroalkyl substances (PFAS) have exemplified the beneficial role of chemistry in modern life, yet they have also underscored significant environmental and health concerns. Termed “forever chemicals” due to their remarkable persistence, PFAS present formidable challenges in terms of contamination and toxicity. Efforts to address these challenges have led to the development of innovative degradation technologies, such as hydrothermal alkali treatment (HALT), low-temperature mineralization, and mechanochemical degradation, offering promising solutions to PFAS remediation. However, these advancements must be accompanied by robust investment in research, collaboration among stakeholders, and global responsibility to ensure effective management of PFAS contamination and mitigate its adverse impacts on ecosystems and human health.

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“Chemistry makes life better” has been a motto to numerous proud chemists and engineers, a shining

example is the discovery of per- and poly-fluoroalkyl substances (PFAS). This is a group of synthetic chemicals which have been used to produce many wonder products such as waterproof jackets and non-stick pans (Gluege et al., 2020; Lyu et al., 2022; Hamid et al., 2024). Unfortunately, PFAS have also attracted growing attentions for

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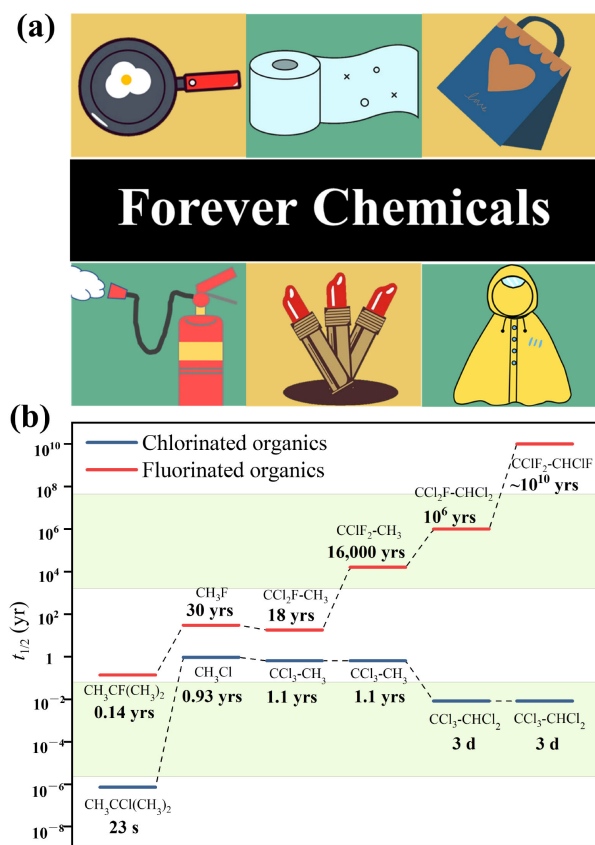
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a very different reason as they have been called “forever chemicals” in science literature as many believe that they are highly toxic pollutants but indestructible in the environment (Fig. 1(a)) (Lyu et al., 2022; Lim, 2023). Chemically, PFAS are fluorinated aliphatic compounds with multiple fluorine atoms that are attached to the carbon backbone. These molecules contain the strongest F–C bond, which makes these chemicals excel in repelling grease and water and renders them exceedingly resistant to degradation in the environment (Beans, 2021; Endo, 2023). Environmental lifespans of PFAS are estimated to be beyond thousands of years (Fig. 1(b)), well above the length of human lifespan (i.e., ~100 years), thus PFAS have frequently been referred to as the “forever chemicals” (Evich et al., 2022).

## 1 New definition of “forever chemicals”

Ideally, engineered chemicals, once released into the



**Fig. 1** (a) Forever chemicals are widely used in household products. (b) Estimated half-lives of some halogenated aliphatic compounds. The half-life values are calculated from measuring the hydrolysis rate constants and also derived from the Arrhenius parameters of neutral and alkaline hydrolysis reactions, see also Supplementary Materials. Data are from Mabej and Mill (1978), Jeffers et al. (1989), and Jeffers and Wolfe (1996).

environment, should break down into harmless end products. Unfortunately, the perfluorocarbon portions of PFAS decompose very slowly or do not decompose at all under natural conditions (Wang et al., 2023). Prior to the advent of the name of “forever chemicals,” some PFAS and chlorinated organics were generally referred to as persistent organic pollutants (POPs) (Muir et al., 2019; Cong et al., 2022; Han et al., 2023). Well known POPs such chlorinated hydrocarbons are common water pollutants with environmental half-lives ranging from a few days to hundreds of years. For example, environmental half-life of 1,1,1-trichloroethane ( $\text{C}_2\text{H}_3\text{Cl}_3$ ) is about one year (Jeffers et al., 1989), while the half-lives of  $\text{C}_2\text{H}_3\text{Cl}_2\text{F}$  and  $\text{C}_2\text{H}_3\text{ClF}_2$  jump to 17.9 years and 16,000 years, respectively. The environmental half-life of pentachloroethane ( $\text{C}_2\text{HCl}_5$ ) is a mere 3 d while those of  $\text{C}_2\text{HCl}_4\text{F}$  and  $\text{C}_2\text{HCl}_2\text{F}_3$  are estimated at one million years and as high as ten billion years (Fig. 1(b)) (Jeffers and Wolfe, 1996), respectively. The fluorine substitution increases the activation energy of both chemical and biological degradation and greatly reduces the reactivity of the organic compounds in the environment. As illustrated in Fig. 1(b), the half-lives of fluoroorganics are 1 to 13 orders of magnitude higher than those of chlorinated organics.

## 2 Health and environmental hazards

PFAS accumulate in the environment media including water, air, sediment and soil (Holder et al., 2023; Wanzek et al., 2023). Recently researchers discovered that PFAS have spread globally—even in remote areas like the polar regions, deep seas, and high mountains (Sonne et al., 2023; Oh et al., 2024). Human exposure to PFAS can occur throughout the life cycle of these chemicals and products, including during the production, manufacture, distribution, usage and disposal. Many PFAS compounds are now ubiquitous in the United States, Europe, and many other countries (Evich et al., 2022). Within the realm of PFAS, perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) have undergone the most extensive scrutiny in terms of biological activity and potential effects on human health (Butruille et al., 2023). The primary route of PFAS exposure to the human body predominantly occurs through the ingestion of contaminated food and drinking water. PFAS tend to accumulate in serum, liver, kidneys, lungs, and heart (Beans, 2021; Dai et al., 2022; Rhee et al., 2023).

It is increasingly found that the PFAS exposure disrupts metabolism and immune pathways, induces reproductive and developmental toxicity, and elevates cancer risks (Steenland et al., 2020; Fenton et al., 2021; Blomberg et al., 2023). Long-chain PFAS, such as PFOS, PFOA, perfluorononanoic acid (PFNA) and perfluorodecanoic acid (PFDA), have been proved to change the immune

status of rodents and non-human primates (Fenton et al., 2021). In addition, PFAS exposure during infancy and childhood can lead to immunosuppression, increasing the incidence of atopic dermatitis and lower respiratory tract infections (Kvalem et al., 2020). Epidemiological studies have also linked serum PFAS levels to the tumor risks, including the cancers in the testis, liver, kidney, breast, and pancreas, particularly in populations residing in PFAS-contaminated environment (Boyd et al., 2022; Panieri et al., 2022). Recognizing these risks, the International Agency for Research on Cancer (IARC) has classified PFOA as a class-2B carcinogen (Pesonen and Vähäkangas, 2024).

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### 3 Barriers to PFAS prohibition

Governments are increasingly using more stringent administrative measures to control PFAS contamination and reduce the chance of the PFAS exposure to the public. For drinking water standards, the US Environmental Protection Agency (USEPA) has set lifetime health advisory concentrations for PFOA and PFOS at 70 ng/L (Hu et al., 2016); the European Commission recently adopted a "Total PFAS" directive limiting total PFAS to 500 ng/L (Uhl et al., 2023). In addition, several countries have implemented the laws and the regulations to phase out the PFAS production and use. In July 2021, five European nations (Germany, Denmark, Sweden, Netherlands, and Norway) announced plans to ban the PFAS production and use (Reinikainen et al., 2024). In the United States, Maine became the first state to halt the sale of PFAS products from 2023, and numerous other states also implement similar restrictions on cosmetics, textiles, and food packaging that use PFAS (Ruffle et al., 2024).

From an economic point of view, it is currently unrealistic to ban all the PFAS. As the legislation on PFAS production in developed countries become more stringent, the PFAS production moves to the developing countries with less stringent environmental regulations. For example, some companies in China began to mass-produce PFAS in 2003 following the phase-out by 3M Company, and China becomes one of the world's leading producers of C8 PFAS since 2004 (Wang et al., 2014; Li et al., 2015). At present, some developing countries still adopt the economic strategy "Grow First, Clean Up Later." If the PFAS are completely banned, the related industries could suffer significant economic losses, not to mention that the producers and downstream users may require costly investment and time-consuming work to develop and implement alternatives.

Ian T. Cousins, a professor at Stockholm University, recommends phasing out PFAS globally. They recommend categorize the PFAS products into three groups (Hogue and Enterprise, 2019). The first group is

those of non-essential uses that can be phased out immediately. For example, PFAS ingredients in cosmetics are not indispensable. The second group is those with available alternatives or of which alternative is of a high likelihood. A typical example of this is the PFAS on raincoats. Some companies are now developing PFAS-free coatings and paraffin wax for waterproofing uses. The third group those that are currently un-replaceable, including those of which suitable alternatives have not yet emerged. PFAS is indispensable in numerous fields, especially high-tech industries such as aerospace industry, where the boycott of the PFAS-containing products may have catastrophic consequences (e.g., spacecraft failure) (Hogue and Enterprise, 2019).

Studies have shown that PFAS, such as perfluoroalkanes, can persist for thousands of years (Ivy et al., 2012). Thus, PFAS will remain in the environment for centuries or more, even if environmental release stops immediately. If effective action is not taken against PFAS, these chemicals will continue to accumulate, causing harm to the health and ecosystems of future generations. Given the impossibility of eradicating PFAS production and usage at the source, efforts must focus on finding ways to eliminate its presence.

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### 4 PFAS removal and degradation

To address the pervasive PFAS contamination, various separation technologies have emerged for the PFAS removal from environmental media and some are promising (Meegoda et al., 2022; de Souza and Meegoda, 2024). These technologies focus on physically separating PFAS compounds from water, soil, or air matrices, thereby preventing their further dispersion and minimizing potential exposure risks. Common separation technologies include membrane filtration, adsorption, and extraction (Meegoda et al., 2022; Fang et al., 2024b). Membrane filtration, such as reverse osmosis and nanofiltration, relies on semi-permeable membranes to selectively remove PFAS molecules, and can effectively separate them from the water matrix (Chaudhary et al., 2023). Adsorption technologies involve the use of the sorbents, such as activated carbon or ion exchange resins, to attract and capture the PFAS compounds from aqueous environment. Some sorbents have large surface area and high PFAS affinity, enabling efficient PFAS removal through physical adsorption (Lei et al., 2023). The extraction methods utilize solvents or solid-phase materials to extract PFAS from aqueous environment, facilitating their subsequent separation and concentration for further treatment or disposal (Meegoda et al., 2022).

While many PFAS are notoriously persistent, efforts have been devoted to exploring degradation mechanisms for select compounds, indicating potential avenues for remediation (Lenka et al., 2021; Liu et al., 2022a; Jin

et al., 2023; Biswas and Wong, 2024). For example, degradation mechanisms of perfluoroalkyl carboxylic acids (PFCA) and perfluoroalkyl sulfonic acids (PFSA) include primarily nucleophilic attack of hydroxyl anions on carbon atoms of terminal carboxylic acid groups or sulfur atoms of the terminal sulfonic acid groups (Cui et al., 2020; Yamijala et al., 2020; Tan et al., 2022). This step requires the highest activation energy and is usually the rate-limiting step in the PFAS transformation (Bentel et al., 2019). After the cleavage of the carbonate/sulfate group, the second hydroxide anion can undergo further nucleophilic substitution of one terminal fluorine atom, resulting in the formation of fluoride and polyfluoroalcohols. The subsequent exchange of fluoride with hydroxide leads to the formation of water, carbonyl fluoride ( $\text{COF}_2$ ) and hydrogen fluoride (Fig. 2(a)) (Wang et al., 2022).

The number of reports is also growing rapidly about the remediation technologies of the PFAS in the environment, such as incineration, electrochemical oxidation, photochemical oxidation (Evich et al., 2022; Xiao et al., 2023). The recent innovative approaches such as hydrothermal alkali treatment (HALT), low-temperature mineralization using dimethyl sulfoxide (DMSO), mechanochemical degradation show promise to effectively degrade PFAS, because of their relatively high efficiency, low costs and low carbon footprints (Fang et al., 2024a).

HALT, a hydrothermal reaction under alkaline conditions, has been proposed for the treatment of PFAS, particularly for the treatment of cationic and zwitterionic PFAS (Hao et al., 2021; Hao et al., 2022). In an aqueous solution dosed with alkali (1–5 mol/L NaOH) at near-critical temperatures and pressures (350 °C, 16.5 MPa), rapid degradation was observed for all 109 PFAS compounds that are identified in two aqueous film-forming foams (AFFF) (at concentrations < 150 mg/L) (Hao et al., 2021). Most PFAS were degraded to undetectable level within 15 min, and the most stubborn perfluoroalkyl sulfonates was degraded within 30 min when treated with 5 mol/L NaOH (Fig. 2(c)). In 2022, HALT's potential as a method for treating AFFF-contaminated groundwater and soil was assessed (Hao et al., 2022). The results showed that all the 148 PFAS compounds that are found in the AFFF-contaminated groundwater and soil were mostly degraded to undetectable levels within 90 min when the soil and groundwater are treated with 5 mol/L NaOH at 350 °C (Fig. 2(d)). The mass balance of fluorine shows that the PFAS are almost completely defluorinated. Nevertheless, the requirement to elevate the temperature (350 °C) for this method remains a limitation, leading to a high energy consumption and costs.

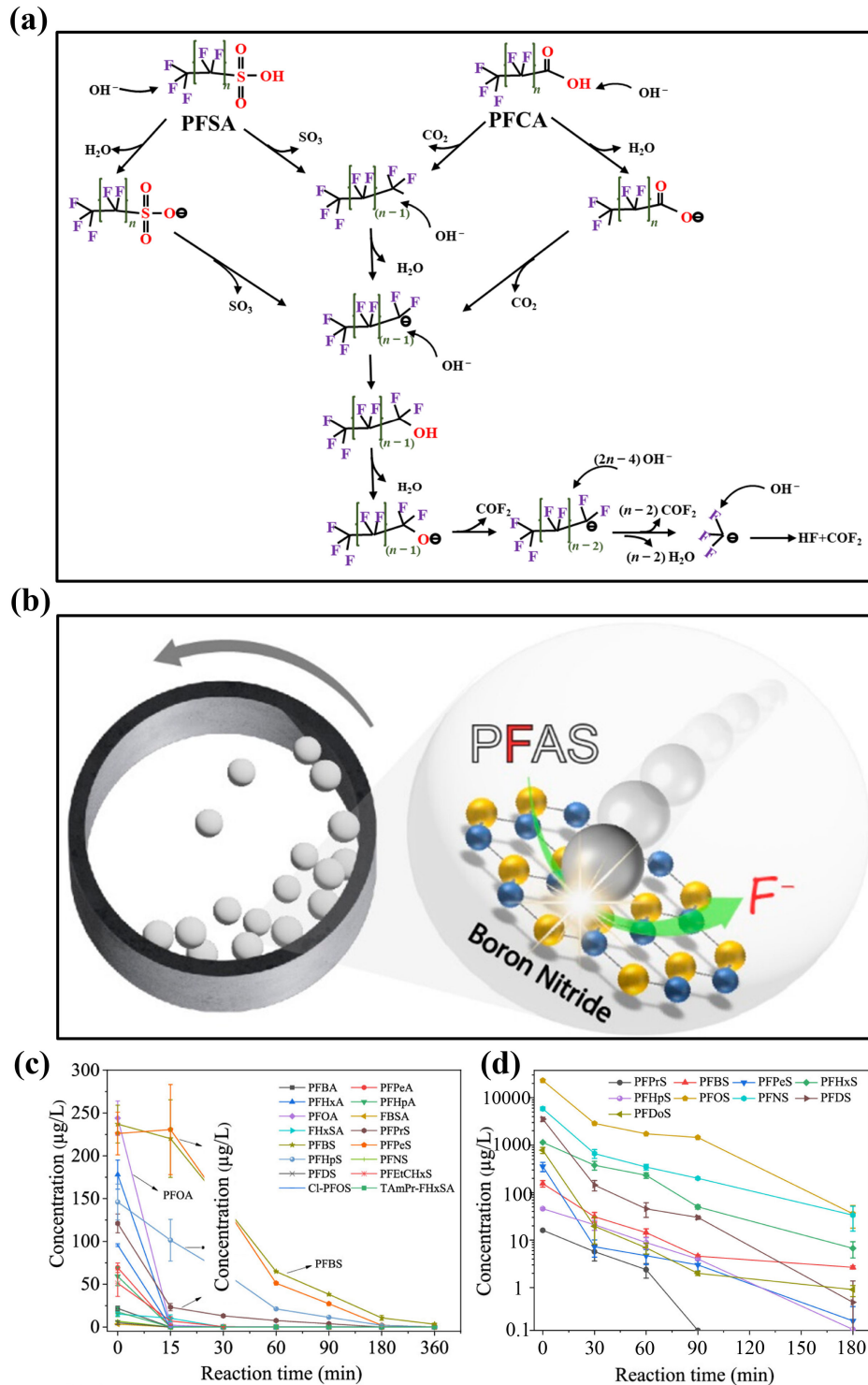
Low-energy pathways for decomposing PFCA have been proposed (Trang et al., 2022). For example, under mild temperatures (ranging from 80 to 120 °C) and ambient pressure conditions, in a mixture of water and DMSO, PFCA carboxylic acid molecules first lose the

carboxylic acid group located on their heads. This leaves behind a highly reactive tail of perfluoroalkyl ions. It further degrades into inorganic fluoride as well as a mixture of carbon dioxide, formate and other small molecules that no longer contain the organic fluorine units within a few hours. The significance of this reaction lies in its mild nature, as well as the use of common and cost-effective reagents such as DMSO solvents, water, and sodium hydroxide. The end product resulting from this treatment contains inorganic fluoride ions and small organic molecules, which pose no harm (Joudan and Lundgren, 2022). However, it's worth noting that this method exhibits a relatively lower treatment efficiency for PFOS due to its inability to initiate the initial step of degrading PFOS by cleaving off the sulfate group.

Mechanochemical degradation is also considered to be a promising technology to degrade PFOA and PFOS (Leung et al., 2022; Yang et al., 2023). This innovative approach harnesses mechanical forces generated via high-energy ball milling to initiate chemical reactions, offering a unique method for PFAS degradation (Turner et al., 2021). The process involves the addition of co-grinding reagents, such as silica, potassium hydroxide, piezoelectric-material (PZM) or calcium oxide, to the milling equipment (Zholdassov et al., 2023). For example, in a milling experiment using PZM (boron nitride), it was successfully demonstrated that solid PFOS and PFOA were completely destroyed and de-fluorinated after 2 h of the milling (Fig. 2(b)) (Yang et al., 2023). This milling process is also used to treat the sediments contaminated with PFAS. About 80% of the 21 targeted PFAS were destroyed in 6 h of the treatment. During the milling, the colliding of the stainless-steel grinding beads in the rotating milling chamber generates free radicals and heat, which promotes PFAS defluorination (Zholdassov et al., 2023). This leads to the cleavage of the carbon-fluorine bonds in the PFAS molecules, breaking them down into smaller, less harmful byproducts.

A notable advantage of mechanochemical degradation is its adaptability and versatility in diverse environmental contexts (Patel et al., 2023). This technique can be applied to the PFAS-contaminated soil, water, and even solid waste (Battye et al., 2022). It offers a valuable alternative for the situations where conventional treatment methods such as incineration or chemical oxidation is impractical or cost-prohibitive. Importantly, mechanochemistry does not rely on the introduction of external chemicals, thereby minimizing the risk of generating secondary pollutants (Do and Friscic, 2017; O'Neill and Boulatov, 2021). Additionally, the mechanochemical process generates reactive species *in situ*, enabling the degradation without the need to elevate the temperatures of entire reactor or use complex machinery (Liu et al., 2022b; Zholdassov et al., 2023). As the research and development of mechanochemical degradation continues, optimization of the process parameters, identification of





**Fig. 2** (a) Degradation pathways of perfluoroalkyl carboxylic acids (PFCA) and perfluoroalkyl sulfonic acids (PFSA) (Wang et al., 2022). (b) Process for PFAS degradation using piezoelectric material-assisted ball milling (PZM-BM) (Yang et al., 2023). Reproduced with permission of American Chemical Society, Copyright of © 2023 Environmental Science & Technology Letters. Degradation of AFFF-contaminated groundwater (c) and soil (d) using HALT technology. Reaction conditions: 350 °C, 1 mol/L NaOH (c), 5 mol/L NaOH (d). Reproduced with permission of American Chemical Society, Copyright of © 2021 Environmental Science & Technology (Hao et al., 2021). Reproduced with permission of American Chemical Society, Copyright of © 2022 Environmental Science & Technology (Hao et al., 2022).

efficient co-grinding reagents, and exploration of scalable industrial applications will be essential to fully unlock its potential in addressing this persistent global issue of PFAS contamination.

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## 5 Challenges and outlook

PFAS pose severe challenges due to their persistence and adverse effects on human health and the environment. Addressing these challenges requires a multifaceted approach that includes the robust investment in fundamental research, the technological advancements, and the enhancement of the collaboration across various stakeholders. This comprehensive perspective aims to delve into some critical aspects, including the development of advanced detection methods, identification of the sources of PFAS and the exposure pathways, and the design of the effective treatment technologies targeting specific PFAS compounds and their transformation products.

**Advancing detection methods.** The USEPA recently proposes that the Minimum Contaminant Level (MCL) of PFOA in drinking water is 4 ng/L. Nonetheless, the 4ng/L concentration is a serious challenge to the accuracy and the sensitivity of the PFAS detection methods. One foremost challenge in dealing with PFAS contamination is the development of more sensitive and accurate methods for PFAS detection. Conventional methods are hampered by inherent constraints, compounded by the ever-changing complexity of PFAS compounds. To overcome these challenges, it is imperative to invest in research aimed at augmenting analytical techniques, notably mass spectrometry and chromatography, to bolster sensitivity and accuracy. Additionally, exploring emerging technologies such as sensors and biosensors holds promise for real-time and *in situ* detection, enabling swift responses to environmental incidents of PFAS.

**Identifying sources and pathways.** Understanding the sources and pathways of PFAS exposure is crucial for effective contamination prevention and management. Research efforts should focus on tracing the origins of PFAS, including industrial discharges, waste disposal sites, and potential atmospheric sources. Robust analytical methods, such as environmental forensics and isotope analysis, can aid the trace of the sources of the emission. Moreover, studying the transportation mechanisms through air, water, and soil is essential to predict and mitigate the spread of PFAS contamination. Banning the PFAS from the source is the first step to solve the PFAS problem. For example, on February 28th, 2024, the US Food and Drug Administration announced that the anti-grease materials containing PFAS for food packaging will no longer be allowed to be sold in the United States. This means that the main sources of PFAS from food packaging, microwave popcorn bags, take-away cardboard containers and pet food bags are being eliminated.

**Designing targeted treatment technologies.** Developing effective treatment technologies is paramount in addressing PFAS contamination. Conventional water treatment methods are unable to remove PFAS compounds effectively. Hence, there is an urgent need to invest the research and development of the advanced treatment technology for PFAS, such as the abovementioned HALT, low-temperature mineralization, mechanochemical technology, and the targeted technologies of high selectivity for specific PFAS compounds.

**Collaboration and Coordination.** PFAS contamination is a complex challenge that necessitates collaborative efforts among diverse stakeholders. Policymakers, industry players, researchers, and affected communities must work together to formulate comprehensive strategies. Establishing national and international frameworks is essential to harmonize regulations and standards, ensuring a global unified approach to PFAS management. Emphasizing data sharing and transparency fosters a more informed and collective response to the contamination crisis.

**Global responsibility and technology transfer.** In 2023, the 3M Company announced that it will provide 10.3 billion US dollars (74 billion yuan RMB) to the public water supply system in the next 13 years to solve the water pollution related to the “forever chemicals” PFAS. Equally important, the industrialized countries should and can offer more investment and transfer the technologies of PFAS treatment and remediation to developing countries. After all, these “forever chemicals” were first invented and produced in a few industrialized countries and just a few companies in those countries have made massive profits from selling the “forever chemicals” and related products all over the world. It is morally imperative for the developed countries and those companies to take more responsibility for the environmental damage.

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## 6 Conclusions

In conclusion, addressing the challenges of PFAS contamination requires a proactive and collaborative approach. Investing in cutting-edge research and technology is essential for advancing detection methods and treatment technologies. Identifying sources and exposure pathways provides a foundation for targeted interventions. Collaboration and coordination among stakeholders, coupled with a sense of global responsibility, will pave the way for a more sustainable and effective response to the persistent issue of PFAS contamination. Through shared efforts and a commitment to environmental stewardship, we can mitigate the impact of PFAS and strive for a cleaner and healthier future.

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