Comment

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Unforeseen risks due to the use of fluorinated materials for per- and polyfluoroalkyl substance removal

Shuqin Liu, Robert J. Letcher, Miriam L. Diamond, Arlene Blum & Da Chen

The escalating regulatory pressures on per- and polyfluoroalkyl substances (PFAS) in drinking water highlight the critical and expeditious need for advanced PFAS removal technologies. While innovative fluorinated materials are reported to be a promising avenue for PFAS removal by exploiting fluorine-fluorine (F···F) interactions, their production and applications raise potential concerns about perpetuating the "forever chemicals" cycle.

The growing need for advanced PFAS removal technologies

Stringent standards for permissible PFAS concentrations in drinking water have been set in response to mounting concerns on the harmful effects of PFAS on human health and the widespread extent of PFAS contamination of source waters. For instance, the U.S. Environmental Protection Agency (EPA) announced in May 2025 that it would retain the Maximum Contaminant Levels (MCLs) in drinking water at 4.0 ng/L for perfluorooctanoic acid (PFOA) and perfluorooctane sulfonic acid (PFOS) under the PFAS National Primary Drinking Water Regulation originally finalized in April 2024[1]. Regulatory frameworks in the European Union, Canada, and China have also established permissible limits for PFAS in drinking water[2–6]. Stringent regulations are increasing pressures on water treatment industries. Although the existing water treatment techniques (e.g., granular activated carbon adsorption, ion exchange and high-pressure membranes) can meet some of the new standards, novel PFAS removal technologies remain needed to achieve compliance with the increasingly stringent drinking water standards and guidelines in a more cost-effective manner. In addition, new technologies are urgently needed to respond to an increasing number of alternative and non-regulated PFAS (e.g., short-chain PFAS such as trifluoroacetic acid, TFA) which may potentially raise global environmental and human health concerns.

The paradox of fluorinated sorbents

The development of sorbents that are highly efficient for PFAS removal represents a promising approach to mitigate PFAS contamination in not just drinking water but also groundwater, surface water, and wastewater that all serve as significant pathways for human exposure to PFAS and are often directly linked to contamination of drinking water systems. State-of-the-art sorbents exploit a variety of mechanisms to achieve efficient PFAS removal, including independent or synergistic inter-molecular actions including electrostatic interactions, hydrogen bonding, hydrophobic interactions, and fluorophilic interactions between PFAS19 molecules and the surface functionalities of the sorbents.

Recent studies have highlighted the potential of fluorophilic affinity based on non-covalent F···F interactions as an efficient approach for PFAS removal[7]. This has led to the development of a series of novel fluorinated sorbents (Fig.1), such as polymeric sorbents, framework matrices, and inorganic materials which are prepared by incorporating F-substituted moieties[8–11] or per-/polyfluorinated carbon chains[12–18] into the sorbent structures. The fluorinated carbon chains incorporated into the structural units of novel sorbents can reach a chain length of up to C8 or even longer (Fig.1).

Although fluorinated materials demonstrate efficient performance in PFAS removal, their development raises serious environmental safety concerns. The production, use, and disposal of these materials could inadvertently release per- or polyfluorinated substances including their by-products into the environment, perpetuating the "forever chemicals" cycle. This creates a paradox: developing materials containing PFAS structures (containing at least one perfluorinated methyl (-CF_3) or perfluorinated methylene (-CF_2-) group according to the 2021 OECD definition[19]) to remove existing PFAS contamination may ultimately create new forms of PFAS contamination. Regarding the paradox of fluorinated sorbents, two controversial issues warrant closer examination.

Controversy 1: Are F···F interactions indispensable for efficient PFAS removal? Many studies highlight that the perfluoroalkyl tails of PFAS can engage in specific F···F interactions with fluorinated moieties on sorbents, thereby enhancing adsorption selectivity and minimizing competitive adsorption of co-contaminants[7,12,13]. F···F interactions also act in concert with electrostatic, hydrophobic, and hydrogen-bonding forces to construct more stable and synergistic adsorption configuration, enhancing both PFAS adsorption capacity and stability[7]. However, these findings are not universal and as evidenced by the results reported in some recent studies. For example, the introduction of fluorinated sidechains into copolymers was found to reduce the binding with PFOA[14], while the addition of fluorinated building blocks to UIO-66 did not lead to an improvement in PFAS removal efficiency[8]. Furthermore, while the addition of fluorophilic functionalities enhanced the adsorption of short-chain PFAS (carbon chain length ≤6) in pure water, the adsorption efficiency drastically decreased in complex matrices[14]. This effect is more pronounced for PFAS with shorter carbon chains. The picture is further complicated by the demonstration of comparable or superior PFAS adsorption capacities in certain non-fluorinated metal-organic frameworks (MOFs) and covalent

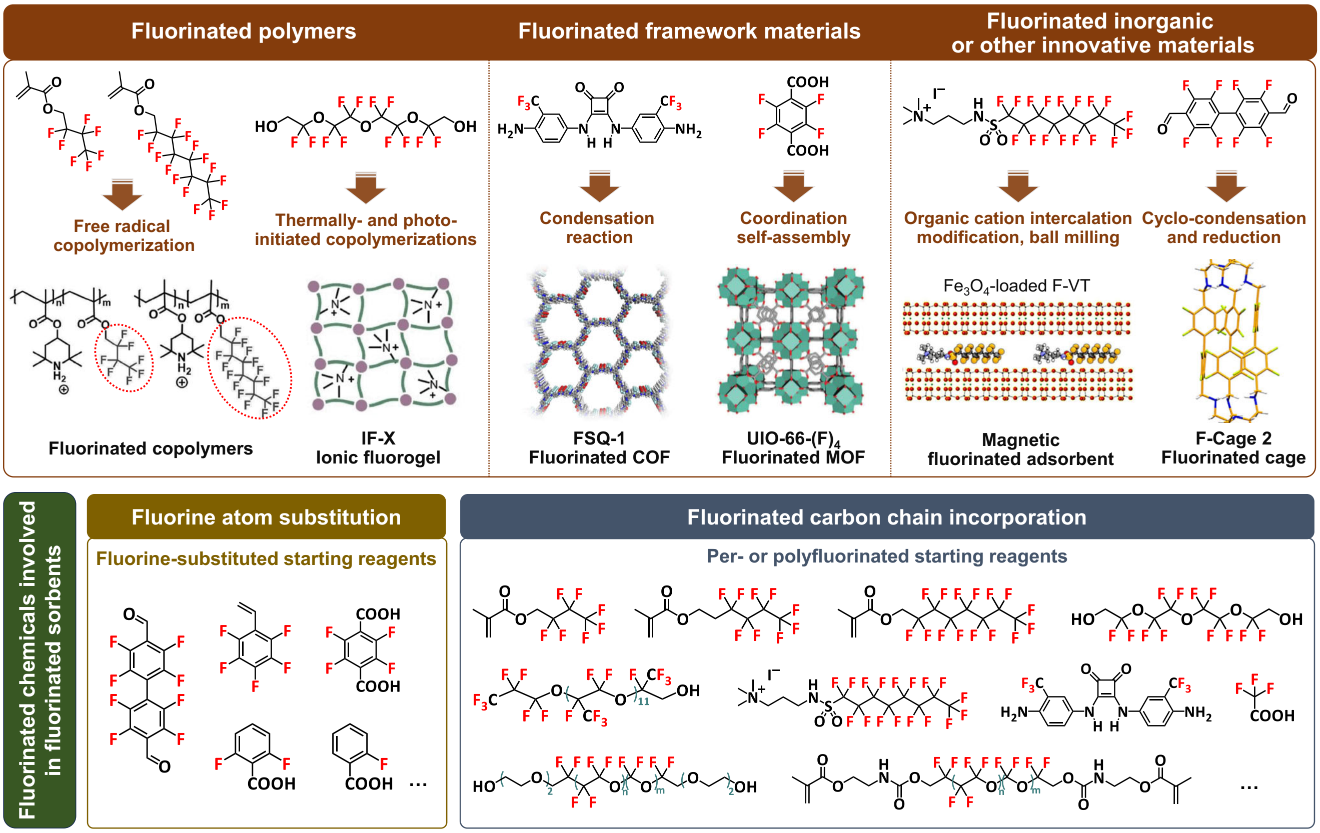

Fig. 1 | Representative structures of fluorinated sorbents and their synthetic fluorinated precursors. Top: Structures of typical fluorinated sorbents, including fluorinated polymers[14,16], fluorinated framework materials[8,18], and fluorinated inorganic or other innovative materials[9,15]. Bottom: Examples of fluorinated chemical reagents used in sorbent synthesis, including fluorine-substituted starting reagents and per- or polyfluorinated starting reagents. The sorbent structures are adapted with permission from refs. 9,15,18. Copyright 2024 American Chemical Society, 2017 American Chemical Society and 2022 Wiley-VCH, respectively.

organic frameworks (COFs). In lieu of F···F interactions, these materials leverage mechanisms such as electrostatic interactions, hydrogen bonding and hydrophobic interactions, which are dominated by physical adsorption, as well as chemically mediated coordination bonds formed through chemical adsorption, to achieve efficient PFAS removal[20,21].

It is important to note that such capacities are often determined under disparate experimental conditions, making direct comparison across materials challenging. Also, the PFAS removal performance of novel sorbents is often evaluated under conditions that are not representative of practical scenarios relevant to water remediation or drinking water treatment, and the scalability of their performance remains unexplored in a large volume water processing facility. As a result, the theoretical saturation capacities of the sorbents reported in laboratory studies are rarely met in real-world environmental applications, as noted above. Thus, the lack of standardized testing protocols significantly hampers the ability to compare sorbent performance across studies[22]. There is also an urgent need for standardized methodologies and real source water treatment applications to enhance our understanding of the strengths and limitations of F···F interaction-based sorbents in PFAS removal. However, despite the concerns with the evaluation methodologies, it is clear that there is no unambiguous evidence showing that F···F interactions are indispensable for efficient PFAS removal. Additionally, as shown below, the use of fluorinated sorbents might result in secondary pollution.

Controversy 2: Are the environmental and human health risks of fluorinated sorbents fully understood? The satisfactory performance of fluorinated sorbents in PFAS removal may be accompanied by potential environmental risks[23]. During the production, use and end-of-life stages, these materials may release a variety of fluorinated chemicals (residues, impurities and degradation by-products) into the environment. The structures, toxicity and persistence of these derivatives originating from fluorinated sorbents are poorly characterized, leaving uncertainties about their long-term environmental exposure and health impacts. Therefore, environmental hazards need to be evaluated against the benefits, which could be achieved using alternative technologies for PFAS removal. Potential hazards may arise from three key stages during the lifecycle of fluorinated sorbents (Fig. 2):

(1) Production of fluorinated reagents and sorbents: The starting reagents of recently developed fluorinated materials for PFAS removal include different kinds of per- or polyfluorinated chemicals (Fig.1), such as TFA[10], 1H,1H-heptafluorobutyl methacrylate[13,14], 1H,1H-perfluorooctyl methacrylate[14], N,N,N-trimethyl-3-(perfluorooctyl sulfonamido) propan-1-aminium iodide[15], and even the low-molecular-weight fluoropolymer product perfluoropolyether (PFPE, with 77 fluorine atoms)[12], as well as others[11,16–18]. These compounds, along with

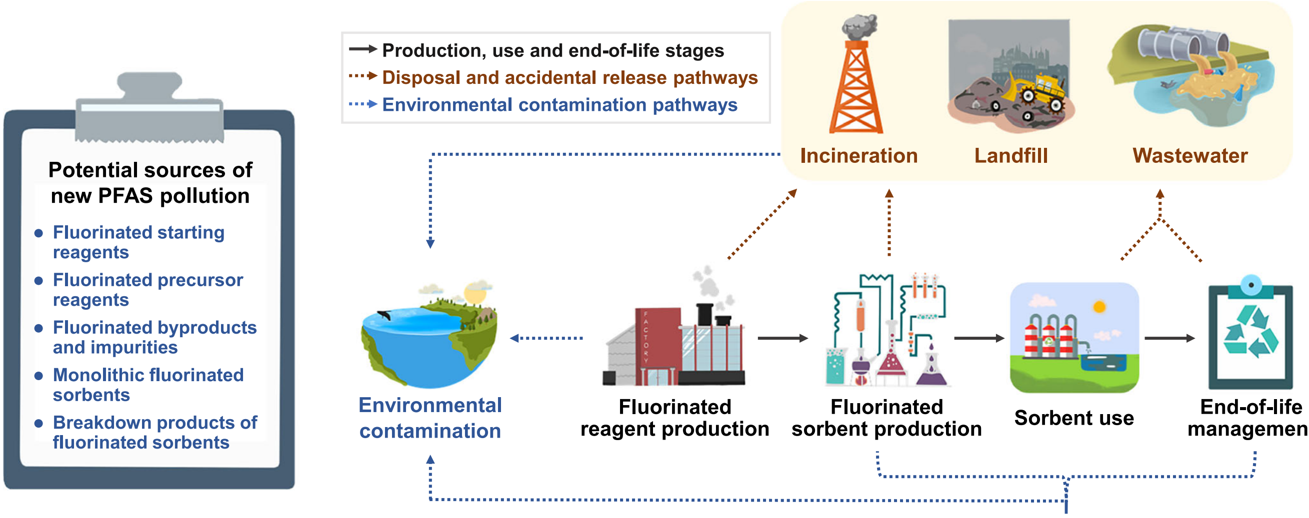

Fig. 2 | Potential release pathways of fluorinated compounds during the life cycle of fluorinated sorbents. Fluorinated compounds may be released at all stages of the life cycle, including reagent production, sorbent synthesis, use, and end-of-life management, leading to potential environmental and health risks.

impurities and by-products, will likely be released during the synthesis of fluorinated sorbents or the production of starting reagents themselves. The production of perfluoroalkyl chemicals via direct fluorination or oligomerization has long been associated with significant environmental releases and contamination[24]. Additionally, poor waste management during manufacturing processes could result in contamination of surrounding water, soil and air, with potential impact on both environmental and human health.

(2) Use of fluorinated sorbents: There is an increasing concern that PFAS or fluorinated derivatives may be released during the use of fluorinated materials, especially under long-term operations. The U.S. EPA found that fluorinated containers can leach parts-per-billion levels of total PFAS into water and methanol after just one week of exposure[25]. Whitehead et al. showed that directly fluorinated plastic containers used for food storage can release perfluorocarboxylic acids (PFCAs) into food products at ng/g levels[26]. In the case of fluorinated sorbents used for PFAS removal, Loukopoulos et al. observed the exchange of non-structural organic ligands during the capture of several PFCAs by the zirconium-based MOFs constructed from fluorine-containing monocarboxylic acids[10]. Specifically, the adsorption of PFAS triggers the exchange and subsequent desorption of the fluorine-containing monocarboxylic acids (i.e., 2-fluorobenzoic acid, 2,6-difluorobenzoic acid or TFA) from the fluorinated MOFs. Therefore, it merits concerns that accidental release of fluorinated chemicals from fluorinated sorbents would cause additional harmful pollution during water treatment.

(3) End-of-life management: Landfilling, incineration and improper disposal of fluorinated sorbents could further release fluorinated by-products or breakdown products into the environment[24,27–29]. Both laboratory and substance flow modelling studies have shown that fluorotelomer-based polymers can slowly degrade under conditions relevant to landfills, releasing fluorotelomer alcohols (FTOHs) and their oxidative derivatives (PFCAs) over time[27,28]. These studies suggest that landfilling of fluorinated sorbents could constitute a potential long-term source of PFAS and other fluorinated substances to surrounding air, soil and water through landfill gas and leachate. Moreover, landfill leachate transported to wastewater treatment plants that lack effective PFAS removal capabilities can ultimately contaminate surface waters distant from landfill watersheds[30]. Combustion of fluorinated materials is of particular concern, as the stability of C–F bonds can lead to incomplete thermal degradation under typical incineration conditions and may generate highly toxic byproducts such as fluoro-dioxins and fluoro-benzofurans, as well as fluorinated greenhouse gases (e.g., tetrafluoromethane and hexafluoroethane)[29]. Such combustion often occurs during solid waste incinerations, landfill fires or open burning, which may pose elevated risks to nearby residential areas, including vulnerable and underserved communities. This warrants particular attention from an environmental equity perspective.

A call for caution and sustainable initiatives

Given the challenges, it is imperative to adopt a forward-thinking and informed approach to PFAS removal technologies based on available evidence. Instead of pursuing fluorinated materials that may exacerbate existing PFAS problems, researchers and policymakers should prioritize the development of non-fluorinated, environmentally benign alternatives. Below are some specific recommendations:

(1) The design of non-fluorinated sorbents is highly encouraged for PFAS removal. Recent studies have reported fluorine-free materials, such as zirconium-based MOFs[20], cationic COFs[21], styrene-functionalized β-cyclodextrin[31], silylated clay[32] and others, as promising PFAS sorbents. These materials can achieve high PFAS removal efficiencies without inadvertently introducing additional fluorinated compounds into the environment. Promoting research into these alternatives is essential to identify materials with optimal performance, scalability, and environmental compatibility. Nevertheless, the design of efficient non-fluorinated sorbents for PFAS removal also relies on the establishment of standardized and environmentally relevant testing conditions for accurate evaluation of sorbent functions and practical applicability for real-world water treatment systems.

(2) As part of development and pre-testing, life-cycle assessment (LCA) is recommended for PFAS removal techniques. This includes qualitative and quantitative assessment of the production and release of impurities, by-products, or breakdown products during the

manufacturing, use, and end-of-life management of fluorinated or non-fluorinated sorbents, as well as the environmental release of fluorinated starting reagents and their derivatives. This can be achieved through targeted and non-targeted mass spectrometer-based analysis. Precautionary evaluations of environmental hazards associated with the releases from sorbents and their derivatives should constitute an important component of the pre-testing system. In addition, the scalability of developed sorbents depends on not only the performance of PFAS removal but also integrated consideration of their economic costs and gains, energy consumption, and carbon release. Therefore, LCAs would facilitate better evaluation of the industrial perspectives and environmental costs of developed sorbents for PFAS removal. Regardless, it is recommended to avoid the production altogether of fluorinated materials for PFAS removal.

(3) Regulations and stakeholder collaborations should be strengthened for fluorinated sorbents. The Stockholm Convention on Persistent Organic Pollutants lists PFOA, its salts and PFOA-related compounds for global monitoring and regulations, where PFOA-related compounds are defined as any substances that degrade to PFOA, including any substances with a linear or branched perfluoroheptyl group with the moiety $(C_7F_{15})C$ as one of the structural elements[33]. The same concept may apply to other long-chain PFCAs, as well as PFOS and PFHxS that are already listed by the Stockholm Convention. Precaution indicates that these efforts to restrict PFAS manufacturing and release be further extended to short-chain PFAS. Several fluorinated starting reagents used for synthesis of fluorinated sorbents fall within the definition of PFOA- or PFOS-related compounds according to the Stockholm Convention. For example, 1H,1H-perfluorooctyl methacrylate[14] and N,N,N-trimethyl-3-(perfluorooctyl sulfonamido) propan-1-aminium iodide[15] are structurally related to PFOA and PFOS (Fig. 1), respectively. Therefore, the use of such reagents in sorbent development could contradict the objectives of the Convention if they are not adequately controlled. Complying with this international treaty, parties to the Stockholm Convention should consider including fluorinated sorbents into regional monitoring, risk assessment, or even regulations. While taking steps to limit future PFAS contamination, collaborations between stakeholders, including the researchers, industry and policymakers, should be promoted to seek optimal solutions to mitigate PFAS contamination. By strengthening regulations, fostering dialogue and sharing knowledge, stakeholders can work together to address the PFAS challenge in a holistic and sustainable manner.

Toward a sustainable future

In the quest to combat environmental pollution, it is crucial to ensure that the solutions we implement do not inadvertently cause greater harm than the problems they aim to solve. The development of fluorinated materials for PFAS removal represents a double-edged sword between potential benefits and unforeseen risks. While these materials could offer solution to PFAS contamination problems, their potential to perpetuate the "forever chemicals" cycle should also be considered. Rather than using a fluorinated chemical to remove another fluorinated chemical, safer non-fluorinated and sustainable alternatives are needed. As global communities continue to grapple with the PFAS crisis, innovations of cutting-edge technologies should be balanced with environmental responsibility toward a sustainable future. Achieving this balance requires a multifaceted strategy: fostering collaborations among researchers, industry leaders, and policymakers; strengthening regulatory frameworks to ensure the safe and sustainable use of PFAS removal technologies; and promoting public awareness to encourage informed decision-making.

Shuqin Liu [1], **Robert J. Letcher** [2], **Miriam L. Diamond** [3,4], **Arlene Blum** [5] **& Da Chen** [1] ✉
[1]Guangdong Key Laboratory of Environmental Pollution and Health, College of Environment and Climate, Jinan University, Guangzhou, China. [2]Wildlife Science and Landscape Directorate, Environment and Climate Change Canada, National Wildlife Research Centre, Carleton University, Ottawa, ON, Canada. [3]Department of Earth Sciences, University of Toronto, Toronto, ON, Canada. [4]School of Environment, University of Toronto, Toronto, ON, Canada. [5]Green Science Policy Institute, Berkeley, CA, USA.
✉e-mail: dachen@jnu.edu.cn

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Author contributions

S. L., conceived and wrote the first draft of the paper. D. C., helped clarify key ideas and edited the paper, with substantial inputs and amendments from R. J. L., M. L. D., and A. B. All authors read and approved the submitted version of the paper.

Competing interests

The authors declare no competing interests.

Additional information

Correspondence and requests for materials should be addressed to Da Chen.

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