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An analysis of PFAS pollution and a comparison of potential treatment methods in the Middle Lower Yangtze Plain

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Abstract:
PFAS pollution has become a significant and global problem, posing threat to human beings. The Yangtze River also suffers from a high amount of PFAS pollution. While textile industries and fluorochemical companies pollute the water with PFAS, no specific water stations designed to tackle the pollution exists in the Yangtze River Plain, the most PFAS-polluted region of the river basin. While a few PFAS-treatment methods exist in the waste water treatment industry, many have been considered impractical for the future due to their low removal efficiency or high costs, or other such reasons. However, a few methods have been proven reliable in recent research papers. The super-fine activated carbon method (SPAC), the plasma chamber method, as well as the electrochemical oxidation method will be discussed in the article in terms of there effectiveness and cost, as well as their applicability in the Yangtze River Plain Region.

Keywords: PFAS, SPAC, plasma, electrochemical oxidation.

1. Introduction

The ubiquity of per- and polyfluoroalkyl substances (PFAS) has long been a problem remaining to be tackled due to the difficulty of PFAS degradation [1]. PFAS is a group of manufactured chemicals that exists almost everywhere, such as everyday drinking water, groundwater near landfills, and even personal care products such as dental floss. [2-4, 9, 12] Therefore, humans are unconsciously exposed to PFAS in many ways, such as drinking PFAS-contaminated water or breathing polluted air. [2-4] Researches have shown that exposure to PFAS may result in serious chronic diseases including cancer, liver dysfunction, and serious chronic liver diseases. According to the National Library of Medicine, the world's largest biomedical library, the recommended concentration of PFAS for human beings is below 2 ng/L, and potential adverse effects may start to take place as concentrations rise from 2 ng/L to 20 ng/L. [5-8]

The Yangtze River, the biggest river in East Asia, is also PFAS-contaminated. As the mother river of a large portion of Chinese citizens, the Yangtze is where the majority of tap water come from. PFAS pollution is greatest in the middle-lower Yangtze River Basin, which is also a densely populated area. The average PFAS concentration in water in the region is an astounding 191 ng/L, almost twenty times the amount considered healthy for humans. [9]

There, textile, paper, and fluorochemistry industries account for large portions of the total PFAS pollution discharge. [9] The Jiangsu Advanced Technological Industrial Park of Fluorochemistry, in particular, holds waste water with PFAS concentrations as high as 12 400 ng/L in its local treatment plants, posing a threat to nearby waterways. [9] Total alternatives to polyfluoralkyl acids (PFAA) range from 0.4 to 208 ng/L in the ground water around the Industrial Park. [9, 12] Home to many fluorochemical companies producing such compounds as polytetrafluoroethylene (PTFE) and polyvinylidene fluoride (PVDF), the industrial park contributes greatly to the emission of perfluorooctanoic acid (PFOA) and Perfluorononanoic acid (PFNA) in the region. [9] It is also worth noting that PFOA is not only the most concentrated PFAS strain in the region, but also the most concentrated PFAS pollutant in the downstream Yangtze River Plain Region. PFOA seems to be present in most, if not all of the 4300 kilometers downstream of the Min River and the city Chongqing situated at the upper stream of the Yangtze [9, 10, 13]. Moreover, PFOA is detected in all sediment samples collected from the water quality stations in the Yangtze River Plain that Zheng et al. chose and discussed in their research. [9, 10, 13] In fact, PFASs were emitted into the Yangtze River at an estimated 10.80 ton/yr. [10, 11]

Having recognized the huge amount of PFAS pollution in natural environment, many nations has made their own regulations to restrict it. For example, in China, government set limits for PFOA at 80 ng/L and perfluorooctane sulfonate (PFOS) at 40 ng/L in drinking water, a restriction that is much higher than its counterparts in the US and Europe. [13] In US, eleven states in total set up Maximum Contamination Levels (MCLs); other eleven states are preparing to apply regulations. [14] Specifically, in MCLs, the limits for PFOA and PFOS are 4 parts per trillion (ppt); and 10 parts per trillion for PFNA, perfluorohexane sulfonic acid (PFHxS), and hexafluoropropylene oxide dimer acid (GenX). [15] The European Food Safety Authority (EFSA) has suggested that combined exposure to four PFAS (PFOA, PFOS, PFHxS, and PFNA) at levels above 6.9 nanograms per kilogram of body weight per week could lead to adverse effects, particularly on the immune system and cholesterol levels. [24]

Currently, a variety of treatment methods have been implemented to address PFAS pollution, each with its own set of advantages and limitations. Among these, granular activated carbon (GAC) stands out for its exceptional performance in absorbing long-chain PFAAs such as PFOS and PFOA. However, GAC's adsorption capacity for similar, shorter-chain compounds is significantly hindered by constituents like natural organic matter (NOM) found in natural water bodies. [16] The weaker

innate hydrophobicity of GAC also diminishes its efficiency in removing short-chain PFASs. Furthermore, the need for frequent replacement due to its limited adsorption capacity makes the method costly to sustain. [17,18]

The ion-exchange resin method (IX) performs better at all scales than GAC does, while still facing challenges. Basically, shorter carbon chains will lead to less hydrophobicity and less polarity of the PFAS molecules. So, like most adsorption methods, IX still shows limited removal efficacy for short-chain PFASs. [18]

Another prevalent treatment method, membrane filtration, needs substantially higher costs in comparison to both IX and GAC. [18] At last, all those methods mentioned above only collects the pollutants rather than degrades them, making those methods only brief respites.

Other more novel technologies such as degradation of PFAS using ultraviolet radiation or ultrasound are proven to be effective to remove pollution with a few compromises. These methods are not only energy-intensive but also fail to remove significant portions of the pollution itself. The sum removal rate of PFAS reaches a 26% peak level using UVA light and catalysts, which is but a small portion of more prevalent methods such as activated carbon, IX, and membrane filtration. [19, 20]

We have chosen super-fine powdered activated carbon (SPAC), plasma chamber, and electrochemical oxidation as methods to be discussed as they have high removal rates and efficiency. However, they all have their own drawbacks. The SPAC does not remove, but only passes the pollution from the liquid phase to the solid phase; the plasma chamber and the electrochemical oxidation methods does not fit the capacity of the Yangtze River, and is dependent on a range of factors from equipment to geographical location. [22, 25, 26]

This paper will aim to compare the technologies in their anility to remain both short-chain and long-chain PFAS, as well as their costs, energy consumption, and the applicability in the region in question. Ultimately, we will try to come up with a feasible solution to be implemented in the Middle Lower Yangtze Plain region.

2. Method 1 of 3 An analysis of PFASs removal using SPAC

Using granular activated carbon (GAC) to remove PFAS has been proven an effective, but not a very efficient method. [21, 22, 23] Recent research showcases that traditional GAC suffers in the removal of short-chain PFAS. [23] This analysis aims to discuss the problem of the low

PFAS removal rate of GACs by comparing a finer-powdered form of activated carbon, or the super-fine-powdered activated carbon technique (SPAC) with the traditional GAC technique in terms of their effectiveness in removing PFAS strains from different concentrations of contaminated water.

SPAC and a ceramic microfiltration system is used in the study alongside a GAC adsorption system. The effectiveness of the two systems in removing PFAS strains is demonstrated in two comparisons, each using a different water sample (Table 1). The effectiveness can be seen by the adsorption mass loading rate, or the average mass of PFAS absorbed by 1g of activated carbon, as well as the number of breakthrough triggered. In this study, researchers set the breakthrough point at 10 percent, meaning that if a concentration higher than 10 percent of the original concentration of PFAS in the influent is detected, a breakthrough is considered triggered. According to the study, SPAC/CMF system only triggered one breakthrough while the GAC triggered six breakthroughs. Moreover, the adsorption mass loading rate for the SPAC/CMF system exceeds that of the GAC in all seven sets of comparison done (Table 2, Table 3).

In the second comparison, the removal capacity of the SPAC/CMF system is further examined with the help of a highly contaminated water sample from a firefighting training area. In this sample, the concentration of particular strains of PFAS is roughly one thousand to ten thousand times the concentration of the sample used in the first study. The performance of SPAC/CMF is still measured in terms of adsorption mass loading rate as well as the number of breakthroughs triggered. According to the chart, the adsorption mass loading rate for specific strains of PFAS reach levels that are hundreds of times the rate in the first study. Furthermore, the system only triggered four breakthroughs under tough conditions. This indicated that the capacity of SPAC/CMF system exceeds that of the traditional GAC system by hundreds of times.

The second experiment revealed several findings.

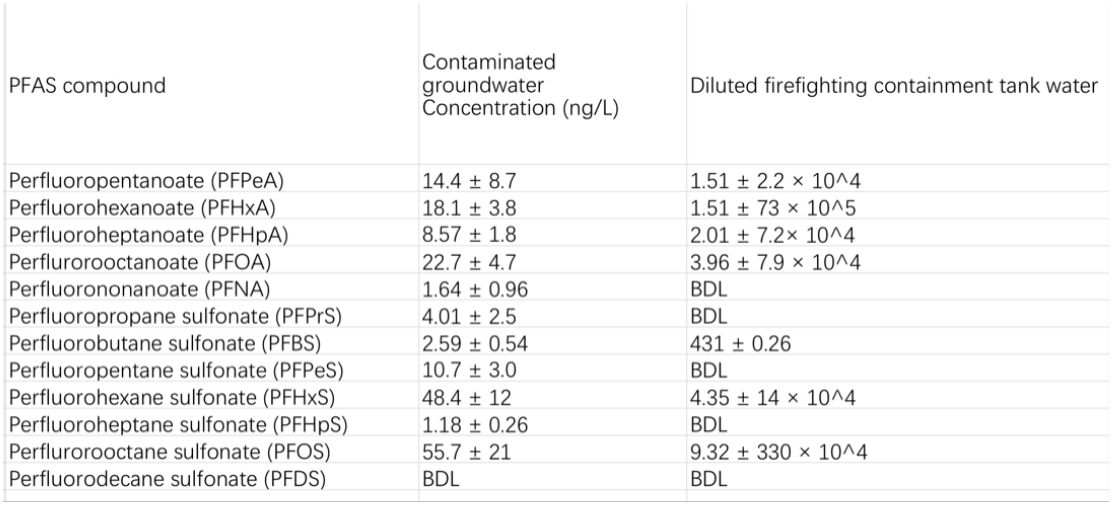
1. The capacity of the SPAC/CMF system rises as the concentration of pollutants in the water rises.
2. The capacity of the SPAC/CMF system can be potentially as high as a hundred times the capacity of the GAC system.

This paper also sheds light on a few other findings, which coincide with past and later findings of other researchers.

1. Long-chained PFAS are more easily absorbed by activated carbon than short-chained PFAS. The adsorption mass loading rate for longer-chained PFAS compounds generally exceeds that of the shorter-chained PFAS substances.
2. The real potential of the SPAC/CMF system is yet-to-be-determined, as the breakthrough is not reached in every single set of experiment in the second study, which means that system is still capable of handling more polluted waters than the sample in the second study.
3. Questions remain on the effectiveness of the removal of short-chain byproducts and other undetected compounds present in the waste waters.

Further study will be needed on the economic efficiency of the SPAC/CMF system, as well as the maximum flow rate it can handle, which will determine its effectiveness in the region in question. More importantly, further treatment of the PFAS polluted activated carbon is needed, as the adsorption system discussed in the paper does not degrade PFAS.

Table.1 Source water characterization of twelve initial PFAA concentrations for the contaminated groundwater and water from a firefighting training are.



PFAS compound	Contaminated groundwater Concentration (ng/L)	Diluted firefighting containment tank water
Perfluoropentanoate (PFPeA)	14.4 ± 8.7	1.51 ± 2.2 × 10^4
Perfluorohexanoate (PFHxA)	18.1 ± 3.8	1.51 ± 73 × 10^5
Perfluoroheptanoate (PFHpA)	8.57 ± 1.8	2.01 ± 7.2× 10^4
Perflurorooctanoate (PFOA)	22.7 ± 4.7	3.96 ± 7.9 × 10^4
Perfluorononanoate (PFNA)	1.64 ± 0.96	BDL
Perfluoropropane sulfonate (PFPrS)	4.01 ± 2.5	BDL
Perfluorobutane sulfonate (PFBS)	2.59 ± 0.54	431 ± 0.26
Perfluoropentane sulfonate (PFPeS)	10.7 ± 3.0	BDL
Perfluorohexane sulfonate (PFHxS)	48.4 ± 12	4.35 ± 14 × 10^4
Perfluoroheptane sulfonate (PFHpS)	1.18 ± 0.26	BDL
Perflurorooctane sulfonate (PFOS)	55.7 ± 21	9.32 ± 330 × 10^4
Perfluorodecane sulfonate (PFDS)	BDL	BDL

BDL indicates below detection limit for a particular analyte.

Table.2 Adsorption mass loading rate (ug PFAA/g activated carbon) for SPAC/CMF and GAC tested with contaminated groundwater.

	PFOS	PFHxS	PFOA	PFHxA	PFHpA	PFPeA	PFBS	Sum PFAAS
GAC	⩾2.4	2.2*	0.86*	0.43*	0.0033*	0.12*	0.015*	6.2`
SPAC/CMF	≥ 3.8	3.3`	1.5`	1.2`	0.49`	0.9`	0.043*	11`

Table.3 Adsorption mass loading rate (ug PFAA/g activated carbon) onto SPAC tested with source water from a firefighting training area.

	PFOS	PFHxS	PFOA	PFHxA	PFHpA	PFPeA	PFBS	Sum PFAAS
SPAC	≥ 2630	121`	111`	95.9 ± 61*	31.3`	3.28 ± 3.2*	0.338 ± 0.074*	2990`

*Indicates that 10% breakthrough was observed

3. Method 2 of 3

Plasma-based water treatment is a treatment that effectively degrades PFAS compounds using a plasma zone. Unlike some other methods on PFAS treatment which either solely remove PFAS or degrade PFAS in low efficiency, plasma-based water treatment shows its huge potential in high rate PFAS degradation. During our research, we figured out two kinds of lab setups that may function more effectively. First lab setup, which is Fig. 1, proposed a model that combines a plasma chamber with a ozonation chamber using a novel cascade reactor. In that reaction cycle, the degradation process happens in the plasma chamber which particularly happens in that plasma zone. While degradation process happens in the plasma chamber, the ozonation chamber functions as a temporary storage space that can keep useless substances away from plasma reactor which increases the degradation efficiency. This first setup has a very high degradation rate especially on PFOA, which is the main PFAS in Yangtze River Plain.

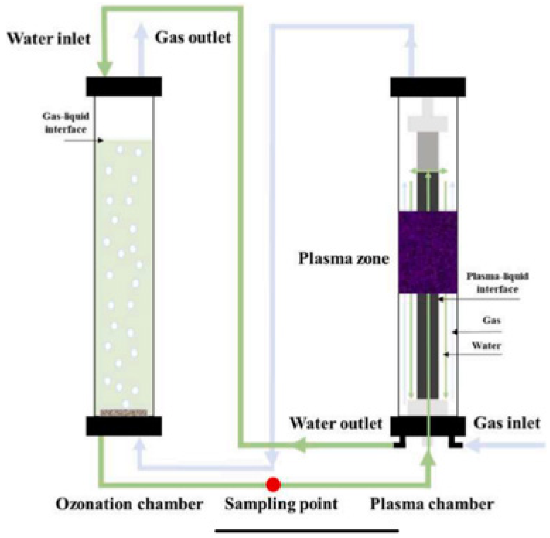


Fig.1 Plasma chamber lab setup

The degradation percentage of PFAS treated by plasma & ozonation is PFBA 11 %, PFHxA 32 %, PFOA 79 %, PFDA 90 %. (1) Direct comparison can be shown in Fig. 3. Such a high degradation percentage is the biggest advantages of plasma-based water treatment, but it still maintains some problems that might need to be solved in the future. Considering the cost of energy used for plasma to form, below equation has been used for the total energy cost. EE/O is the amount of electric energy cost to achieve 90% of PFAS degradation rate in one cubic meter wastewater, P is the discharge power, kW, t is the time required to achieve 90 % degradation of PFAS, min, and V is the wastewater volume, L. (2)

$$EE/O = P \times t \times 1000 / (V \times 60 \times \log(C0/C1))$$

The EE/O of PFBA, PFHxA, PFOA and PFDA is 1600 ± 450, 800 ± 230, 160 ± 11 and 90 ± 8 kWh/m3, respectively, in synthetic wastewater; while being 2900 ± 1400, 690 ±90, 180 ± 15 and 100 ± 10 kWh/m3 in secondary effluent as shown in Fig. 4. (3)

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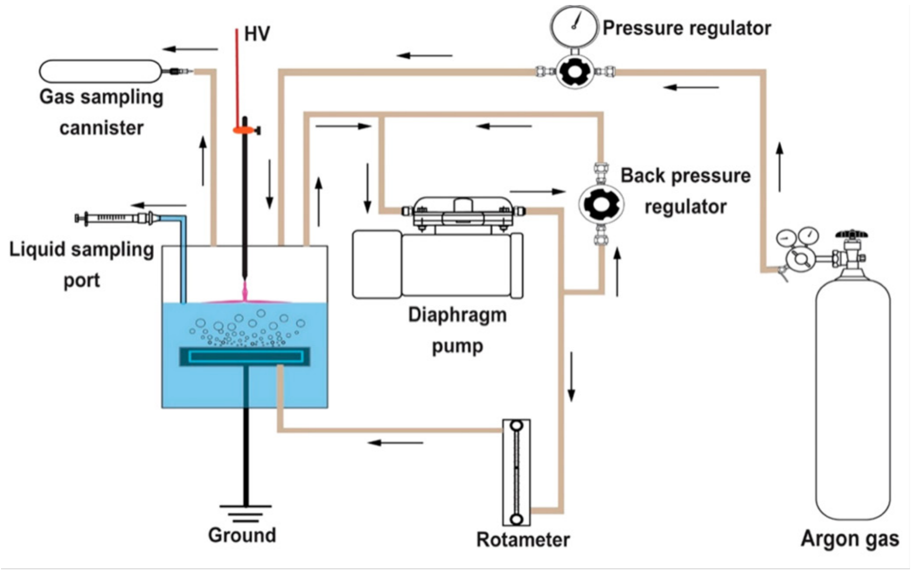

Fig.2 Plasma water treatment lab setup 2

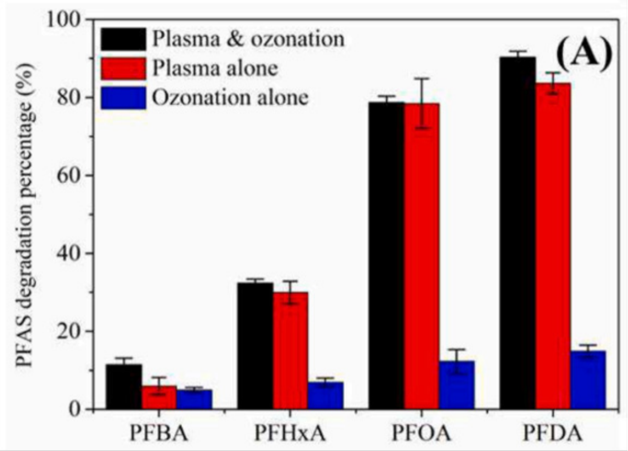

Fig.3 PFAS degradation percentage

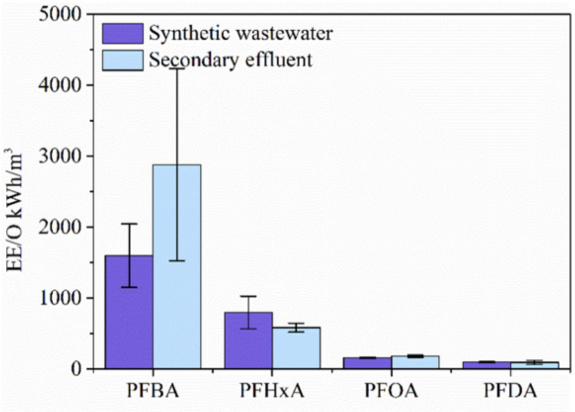

Fig.4 Plasma chamber and ozonation chamber energy cost

In lab setup 2, The system is formed as shown in Fig. 2. The two main components of this system are nickel-chromium rod (diameter = 2.2 mm) as a high voltage (HV) electrode, and an aluminum ring (outer diameter = 9.8 cm, inner diameter = 6 cm) as a ground electrode. The high voltage generated by the system is used to create plasma zone while argon gas is inputted into the system to promote the interaction between plasma and PFAS. Similarly, the efficiency of PFAS degradation is also far higher than some old methods as shown in Fig. 5. In this study, 90% of the PFOA was removed in 60 min, which is even higher than previous lab setup. However, the whole process of this experiment requires the system to maintain a 30 kW negative voltage and 0.24 J/pulse discharged energy. (4) Such huge amount of energy cost brings huge disadvantage when compare with some other methods.

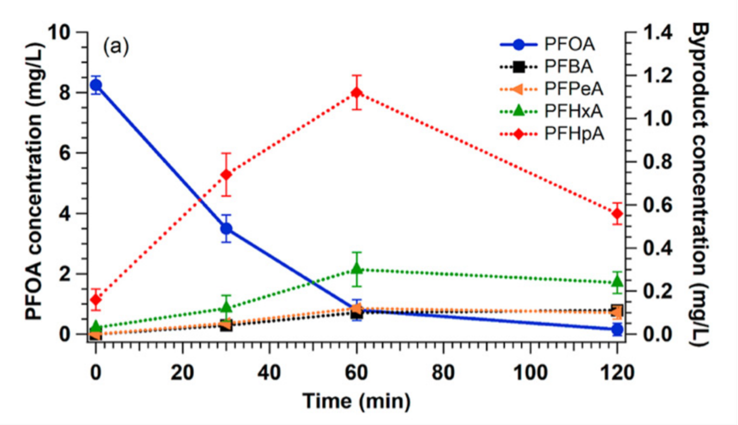

Fig.5 PFOA concentration change

Apart from the problem of huge amount of energy cost, the efficiency for Yangtze River PFAS degradation is still considerable. In first lab set up, only 600 ml wastewater can be added into the system each time, which only a liter wastewater can be added into lab setup two each time. Compare to the volume and flow rate of Yangtze River, plasma-based water treatment is too weak to contain the work.

4. Method 3 of 3

The electrochemical oxidation (EO) method is a novel treatment technique that stands out for its efficiency in breaking down per- and polyfluoroalkyl substances (PFAS). Unlike methods that focus solely on the absorption and transfer of PFAS, EO can effectively decompose these molecules with high efficiency. One of the key advantages of EO is its cost-effectiveness; most reactions occur under ambient temperature, and the process requires minimal or no chemical additives.

Furthermore, the byproducts of EO, such as fluorine, hydrogen fluoride, and oxygen, are less toxic compared to those produced by other methods. This makes EO technology highly energy-efficient, scalable, and modular. However, like any technology, EO has its drawbacks. For instance, it is less effective in treating shorter-chain PFAS, with removal efficiencies ranging from as low as 45% to an average of 80%, depending on the electrodes used [1]. Additionally, the process may produce toxic byproducts from the electrolyte or electrodes, and the current models may not fully explain the degradation pathways, which do not always follow the expected mechanisms.

Despite these challenges, the benefits of EO generally outweigh the disadvantages, making it a viable treatment option for real-world applications. The primary mechanism of EO involves ‘unzipping’ the PFAS molecule through desulfurization, decarboxylation, and the detachment of CF2 units [2]. With sufficient voltage, the carbon chain is progressively shortened due to the action of hydroxyl radicals. However, this process can also generate shorter-chain PFAS molecules, some of which may escape into the gas phase, reducing the overall degradation efficiency. The choice of electrodes and voltage can significantly influence this outcome, making the selection of materials a focal point of current research.

Given the pressing issue of PFAS pollution in the Yangtze Middle Lower Basin, there is an urgent need for effective solutions. While EO offers a promising theoretical approach, its practical application is still in its infancy. The field anticipates the emergence of more practical methods in the near future to address this critical environmental challenge.

5. Discussion and Conclusion

PFAS had been threatening the ecosystems in the Middle and Lower Yangtze Plain, while the mainstream treatment methods remain highly empirical. Here we report, that throughout the discussion above, we have examined the applicability of each method in the Middle and Lower Yangtze Plain, reaching out the conclusion that none of the three methods above except for SPAC is applicable. The research has also shown that SPAC has sufficient capacity to deal with the waste water in the Yangtze River, yet practical feasibility remains to be examined due to the potential high costs. Therefore, the findings of this study have a number of important implications for future practice: more field experiments need to be conducted.

In conclusion, our review concludes that there are much inadequacy in the field of practically treating PFAS, especially for degradation of PFAS rather than the respite of collecting pollutants through adsorption mediums. Overall, these studies highlight the need for further examination of, specifically, the economic efficiency of the treatment methods. Considering the infancy of all the edge-cutting treatment methods of PFAS degrading, the world still expects more novel techniques.

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Yichen Lu, Ziming Wang, Yuanming Taoliao contributed equally to this work and should be considered co-first authors.

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