BEFORE THE HON'BLE NATIONAL GREEN TRIBUNAL

PRINCIPAL BENCH, NEW DELHI

ORIGINAL APPLICATION NO. 548 OF 2024

In the matter of: -

Suo motu case based on news item titled "IIT Madras study reveals presence of, forever chemicals, in Chennai lakes drinking water" appearing in the Hindu dated 07.04.2024

Index

Filed by Advocate: -
Rajkumar
(On behalf of Central Pollution Control Board)

Dated: 27.08.2024
Place: Delhi

BEFORE HON'BLE NATIONAL GREEN TRIBUNAL,

PRINCIPAL BENCH, NEW DELHI

Original Application No. 548/2024

IN THE MATTER OF:

Suo motu case based on news item titled "IIT Madras study reveals presence of, forever chemicals, in chennai lakes drinking water" appearing in the Hindu dated 07.04.2024

REPLY ON BEHALF OF RESPONDENT NO. 1 CENTRAL POLLUTION CONTROL BOARD (CPCB)

1. That the Hon'ble NGT vide order dated 17.05.2024 has impleaded Member Secretary, Central Pollution Control Board (hereinafter referred as CPCB) as Respondent No.1 and issued Notice to the respondent CPCB for filing the response in the instant matter. Thereby, the reply is made in succeeding paragraphs.

2. That, CPCB is a statutory Board constituted under Section 3 of The Water (Prevention and control of Pollution) Act, 1974. It performs the functions under The Water (Prevention and control of Pollution) Act, 1974, The Air (Prevention and Control of Pollution) Act, 1981 and The Environment (Protection) Act, 1986.

NOTARY PUBLIC R.L. Chaudhary N.C.T. of Delhi Regd. No. 1513 GOVT. OF INDIA

3. That the Original Application No. 548 of 2024 (PB) is regarding the presence of 'forever chemicals' i.e. per and polyfluroalkyl substances in Buckingham Canal, Adyar river and Chembarambakkam lake in Chennai (Tamil Nadu) as per a recent study conducted by IIT Madras.

4. That it is humbly submitted that MoEF&CC, Delhi awarded the project "Strategic Approach to Management of Perfluoroalkyl Substances in India" to IIT Madras, and the main objective of this project is to state the necessity of Perfluoroalkyl and

Polyfluoroalkyl Substances (hereinafter referred as PFAS) related guidelines in India. The IIT Madras carried out the study and the project report was submitted to MoEF&CC in the month of June, 2022.The report included

i. History of PFAS production and phase-out
ii. Toxicity of PFAS and its related compounds
iii. Distribution and fate of PFAS in different environmental matrices
iv. Detected concentrations of PFAS in India and Worldwide
v. Overview of the treatment technologies to reduce PFAS in various matrices
vi. Guidelines, policies, and regulations formulated to mitigate the PFAS pollution
vii. Challenges in the PFAS monitoring in India
viii. Recommendations to frame the monitoring guidelines in India

The copy of the study conducted by the IIT Madras on "Strategic Approach to Management of Perfluoroalkyl Substances in India" is annexed herewith as **ANNEXURE-1**.

5. That, it is humbly submitted that from the executive summary of the report it has been learnt that in India, only limited number of monitoring studies have been conducted for Perfluoroalkyl Substances. The monitoring of Perfluorooctanoic acid (hereinafter referred as PFOA) and Perfluorooctane sulfonic acid (hereinafter referred as PFOS) in samples of tap water, drinking water, and surface water was done for a maximum of two years (2006-2008) only. Over the period of time (2006-2020), the concentrations of PFOA and PFOS have considerably increased. For instance, the concentration of PFOA in drinking water samples collected from South India was <0.005 nanogram/liter in 2008 (Mak et al., 2009) and 2 nanogram/liter in 2015 (Selvaraj et al., 2021). Similarly, the concentrations of PFOS have also risen from <0.033 nanogram/liter(Mak et al., 2009) to 1 nanogram/liter (Selvaraj et al., 2021). This gradual increase in the

concentrations of PFAS might be related to the shift of industrial production from developed countries to developing countries (e.g., China and India) due to the phase-out initiative taken by US EPA via the Stewardship Program.

6. That, it is humbly submitted for regulations of PFAS in India, India has set the first step to regulate PFOA and PFOS by adopting the standard methods used for sampling and analysis. That Considering the hazardous nature of PFOA and PFOS, accordingly, the BIS announced three methods for sampling and measuring PFOA and PFOS (BIU, 2020). The methods are:-

   i. ISO 3696:1987: Water for analytical laboratory use Specification and test methods,
   ii. ISO 5667-1 Water quality sampling- Part 1: Guidance on the design of sampling programs and sampling techniques,
   iii. ISO 8466-1:1990 Water quality– Calibration and evaluation of analytical methods and estimation of performance characteristics– Part 1: Statistical evaluation of the linear calibration function.

It is further submitted that according to the report "Strategic Approach to Management of Perfluoroalkyl Substances in India" submitted by IIT Madras to MoEF&CC in June,2022, the detected concentrations of PFAS in India are relatively lower than those in other countries.

7. That it is humbly submitted that there is no standard prescribed for PFAS in drinking water by Bureau of Indian Standard in Indian standard (IS 10500:2012).

8. That, it is humbly submitted the MoEF&CC is coordinating a project "Review and update of National implementation plan on persistent organic pollutants" through CSIR-NEERI. NEERI is carrying out countrywide survey/inventory of manufacturing and usage of Persistent organic compounds(POPS) by industries including PFAS manufacturing Industries.

9. That, the answering respondent craves leave of the Hon'ble Tribunal to file additional reply, in future, if required.

10. That in light of the above submission, it is respectfully submitted that this Answering respondent i.e. CPCB, shall abide by any order(s) or direction(s) passed by this Hon'ble tribunal in the instant OA.

NOTARY PUBLIC
R.L. Chaudhary
N.C.T. of Delhi
Regd. No. 1513
GOVT. OF INDIA

(Dinabandhu Gouda)
Scientist 'F'
Central Pollution Control Board

BEFORE HON'BLE NATIONAL GREEN TRIBUNAL,

PRINCIPAL BENCH, NEW DELHI

Original Application No. 548/2024

IN THE MATTER OF:

Suo motu case based on news item titled "IIT Madras study reveals presence of, forever chemicals, in chennai lakes drinking water" appearing in the Hindu dated 07.04.2024

AFFIDAVIT

I, Dinabandhu Gouda, working as Scientist 'F' in Central Pollution Control Board, Parivesh Bhawan, East Arjun Nagar, Delhi, the Respondent No. 1 in the above matter, do hereby solemnly affirm, declare on oath and state as under:-

1. That I, the deponent herein is authorized representative to represent the Respondent CPCB in the present case, and as such, I am well conversant with the facts and circumstances of the present case on the basis of the information derived from the official records, and hence, I am competent and authorized to verify, sign and swear this affidavit on behalf of the Respondent CPCB.

2. That the accompanying reply may be read part and parcel of the present affidavit as I am competent to swear this affidavit.

3. That the accompanying reply has been drafted and filed under my instructions and authority the contents thereof are true and correct on the basis of the record maintained during ordinary course of business of CPCB and available records and documents and the contents of the same are read over and explained to me and are not repeated herein for the sake of brevity.

NOTARY PUBLIC
R.L. Chaudhary
N.C.T. of Delhi
Regd. No. 1513
GOVT. OF INDIA

DEPONENT

दीनबन्धु गोडा /Dinabandhu Gouda
प्रभागीय प्रमुख, आई.पी.सी.-I/Divisional Head, IPC-I
केन्द्रीय प्रदूषण नियंत्रण बोर्ड
Central Pollution Control Board
पर्यावरण, वन एवं जलवायु परिवर्तन मंत्रालय, भारत सरकार
M/o Envt. Forest & Climate Change, Govt. of India
परिवेश भवन, पूर्वी अर्जुन नगर, दिल्ली-110032
Parivesh Bhawan, East Arjun Nagar, Delhi-110032

VERIFICATION:

Verified at New Delhi on this day of 27 AUG 2024 2024 that the contents of the above reply are correct and true on the basis of the records of the case as mentioned in the day-to-day affairs of the CPCB. Nothing has been concealed therefrom or mis-stated.

NOTARY PUBLIC
R.L. Chaudhary
N.C.T. of Delhi
Regd. No. 1513
GOVT. OF INDIA

DEPONENT

दीनबन्धु गोडा /Dinabandhu Gouda
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Parivesh Bhawan, East Arjun Nagar, Delhi-110032

ATTESTED
NOTARY PUBLIC
GOVT. OF INDIA
27 AUG 2024

Strategic Approach to Management of

PERFLUOROALKYL SUBSTANCES in INDIA

Report prepared by

Indian Institute of Technology Madras

Submitted to

Ministry of Environment, Forest, and Climate Change,

New Delhi, June 2022

Strategic Approach to Management of Perfluoroalkyl Substances in India

This report “Strategic Approach to Management of Perfluoroalkyl Substances in India” is a part of a larger project on the "Strategic Approach to International Chemical Management (SAICM)", supported by the Ministry of Environment, Forest, and Climate Change (MoEF&CC, GoI). Several experts have contributed to the report in varying capacities. Project coordinators Prof. Ligy Philip, Prof. B.S. Murty, and Prof. V.R. Muraleedharan, would like to sincerely thank all of them for their critical contributions to this report.

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EXECUTIVE SUMMARY

Poly and Perfluoroalkyl Substances (PFAS) are commonly used as a coating material in non-stick cookware and food packaging materials. Additionally, PFAS have been used in fire-fighting foam because of their water repellent feature. The superior physicochemical properties of PFAS have found their applications in many day-to-day products. Consequently, their concentrations in the different environmental matrices have increased. The tragedy brought about by PFAS producer DuPont made the world aware of the toxicity of PFAS. The release of untreated industrial Wastewater (WW) containing PFAS causes diverse chronic health impacts such as thyroid disease, liver damage, kidney cancer, testicular cancer, breast cancer, and so on. So, different governmental agencies (e.g., US Environmental Protection Agency (US EPA) and World Health Organization (WHO) have developed various guidelines and regulations to minimize PFAS pollution. However, policies related to the regular monitoring of PFAS in India are in the initial stages only. In this regard, PFAS has been included in the SAICM's framework so that national policies can be formulated to monitor and control the PFAS levels in India.

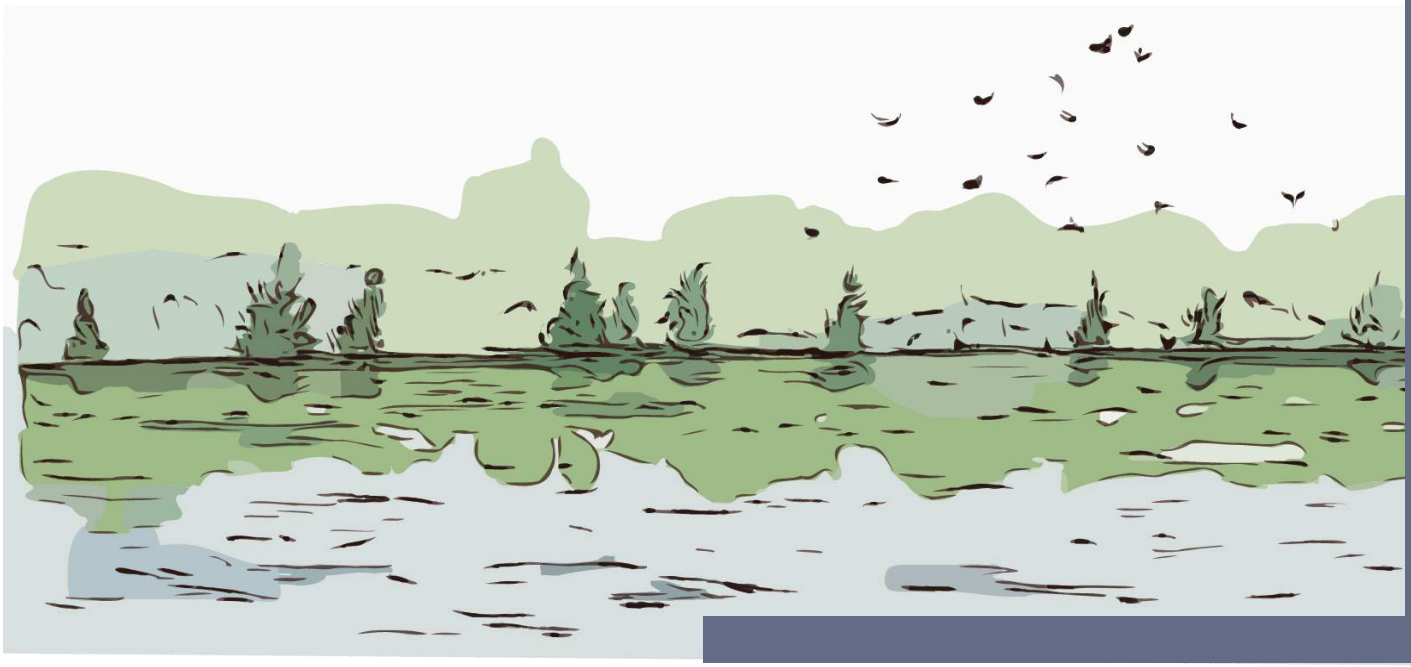
The main objective of this report is to state the necessity of PFAS related guidelines in India. The report includes,

- History of PFAS production and phase-out
- Toxicity of PFAS and its related compounds
- Distribution and fate of PFAS in different environmental matrices
- Detected concentrations of PFAS in India and Worldwide
- Overview of the treatment technologies to reduce PFAS in various matrices
- Formulated guidelines, policies, and regulations to mitigate the PFAS pollution
- Challenges in the PFAS monitoring in India
- Recommendations to frame the monitoring guidelines in India

According to the Chemical Abstract Service (CAS) registry, more than 4000 PFAS related compounds are available, and greater than 100 PFAS related compounds are continuously manufactured and used in industrial applications. The research dataset indicates that PFAS are persistent organic compounds, so minimizing their production is essential to cut down PFAS pollution in the long term. As the toxicity of two widely used PFAS (e.g., Perfluorooctanoic Acid (PFOA) and Perfluorooctane Sulfonate (PFOS)) was proven, efforts have been taken to phase out these compounds by 2002. According to the Stock-

holm convection, production and usage of PFOA and PFOS are banned in countries like the US, Europe, Japan, Korea, and Australia. Though the production restriction was initiated a decade ago, the global emission of the PFOA and PFOS have not reached any regulatory guideline values, indicating their persistent nature. Accordingly, the countries identified with high concentrations of PFAS, such as the US, Europe, and Australia, are using granulated activated carbon technology to treat water and WW contaminated with low (10 ng L^{-1}) to high (>1000 µg L^{-1}) concentrations of PFAS. Likewise, incineration is a commonly used technology to treat PFAS-contaminated soil, particularly in military-based sites where extensive artificial fire-frightening foam is used for training. In the US and Europe, the PFAS levels in the identified hotspot are monitored regularly and keenly. The health of the victims is also recorded periodically. Based on these datasets, the toxicity profiles of PFAS are updated by governmental agencies regularly.

In India, only a limited number of monitoring studies have been conducted. The monitoring of PFOA and PFOS in samples of tap water, drinking water, and surface water was done for a maximum of two years (2006-2008) only. Over the periods (2006-2020), the concentrations of PFOA and PFOS have considerably increased. For instance, the concentration of PFOA in drinking water samples collected from South India was <0.005 ng L^{-1} in 2008 (Mak et al., 2009) and 2 ng L^{-1} in 2015 (Selvaraj et al., 2021). Similarly, the concentrations of PFOS have also risen from <0.033 (Mak et al., 2009) to 1 ng L^{-1} (Selvaraj et al., 2021). This gradual increase in the concentrations of PFAS might be related to the shift of industrial production from developed countries to developing countries (e.g., China and India) due to the phase-out initiative taken by US EPA via the Stewardship Program. This kind of geographical shift resulted in the increased occurrences of PFAS and its alternatives in China, India, and Russia. For instance, the Chinese Ministry of Environmental Protection classified the PFOA under the Comprehensive Catalogue for Environmental Protection when China reported a high concentration of PFOA and PFOS after 2006. So, it is essential to map out the pattern of increase of PFAS in India too. Considering the hazardous nature of PFOA and PFOS, the Bureau of Indian Standards (BIS) developed a sampling and analysis methods for PFOA and PFOS in water samples (ISO 3696-1987). Apart from this, no phase-out or guidelines have been developed by the Indian government.



The existing gaps in monitoring, controlling, and remediating PFAS in India,

* Standardized methods for sampling and analysis: The current implementation of BIS is focused on the PFOA and PFOS. Nevertheless, short-chain PFAS needs to be quantified as their concentrations gradually increase. Therefore, it is essential to develop a standardized sampling and analysis protocol for various classes of PFAS.
* Development of a guideline for PFOA and PFOS: Many countries have developed the guideline values for PFOA and PFOS in collaboration with their state governments or have adopted the guideline value recommended by the US EPA. In this respect, considering the chronic toxicity of PFOA and PFOS, framing guideline values for two predominant PFAS, namely, PFOA and PFOS, are essential.
* The toxicity, fate, and distribution of PFAS alternatives in diverse environmental compartments are unknown. In India, these compounds' concentration gradually increases over a period. Therefore, periodic monitoring of PFAS and identifying alternatives are vital to control the spread of PFAS in the environment.
* Standard protocols are necessary to identify the hotspot of PFAS: As a wide variety of PFAS-related components exist in the market, the current usage pattern of PFAS indicates its geographic-specific nature. In such a line, it is essential to formulate a protocol or questionnaire to identify the hotspot of PFAS.
* Transparency in the production, export, and import of PFAS-related compounds is needed.

1. INTRODUCTION

Lifestyle changes have increased the consumption of personal care products and packed food items. This increased the presence of PFAS in different environmental compartments. PFAS, which are hydrophobic and oleophilic compounds, are commonly used as a coating material for non-stick cookware, food packaging, wax, and fire-fighting foams (Vo et al., 2020). PFAS was first commercially used by DuPont® in the 1940s. Currently, more than 4,000 PFAS are in use in different industrial applications (e.g., surface coating and paper protection) (Bolan et al., 2021). Electrochemical fluorination (ECF), telomerization, and radical polymerization are the typical synthesis routes for PFAS (Prevedouros et al., 2006; Buck et al., 2011). The strong acidic nature of PFAS allows their complete ionization in water (high aqueous solubility, ranging from -570 to 9,500 mg L^{-1} which results in more stability in water, acids, and alkali (Martin et al., 2004). Likewise, the high thermal and mechanical stabilities of the PFAS increase with the number of carbon atoms (O'Hagan, 2008). Also, lesser intermolecular interactions between fluorine atoms increase the fugacity (volatility) of PFAS. These physicochemical properties of PFAS make them difficult to hydrolyze or degrade, persisting in soil/sediments and subsequently accumulating in biota (Ghisi et al., 2019).

The bioaccumulation in diverse environmental matrices causes the entry of PFAS into the food chain, subsequently affecting humans and other animals. The Stockholm Convention Committee conducted toxicity studies related to PFAS emissions in 2007 and 2017 (Convention, 2007; EPA, 2014). The report observed that exposure to PFAS via inhalation and dermal route results in heart diseases, asthma, semen quality reduction, fluctuated glucose, blood pressure, thyroid levels, ovarian diseases, low birth infants, abnormal menstrual cycle, female infertility, and cancer (Steenland and Winquist, 2020; Stone et al., 2021). PFAS is not only a “forever chemical” but also “a life-threatening toxic substance” for humans and animals. Therefore, Stockholm Committee recommended that global actions are required to mitigate the pollution of PFAS and their associated negative impacts. In response, several countries introduced PFAS-free products in their markets and attempted to replace long-chain PFAS with shorter ones. However, in India, no guidelines were made to regulate PFAS in various consumer and defense goods. On the other hand, research indicates a gradual increase in the concentrations of PFAS in samples in various environmental matrices. So, people are exposed to PFAS indirectly resulting in adverse health impacts.

Estimates of the global production of PFAS until 2003 varied from 44,000 to 75,000 metric tons, and the maximum production was observed from 1990 to 2000 (~4,500 metric tons per year) (Paul et al., 2009). The corresponding total emission was 3,200-7,300 metric tons (Prevedouros et al., 2006). This sharp increase in the use of PFAS increased their concentration in the environment, which in turn has been causing adverse impacts on human health. Therefore, as illustrated in Figure. 1, phase-out of the PFAS in several nations got initiated during the years 2000 to 2003 (Geueke, 2016). Europe, the US, and Japan implemented strict regulations to cut emissions. However, around the same time, emissions of PFAS peaked in the countries such as India, Poland, and China (Wang et al., 2014b). In 2013, the fire-fighting foams and food packaging materials showed a net profit of $130 million (Research, 2015). The annual demand for fluorinated compounds in 2020 was estimated at 47,500 metric tons, with a yearly growth rate of 12.5% (Research, 2015).

Although India allowed the imports of only PFAS-free non-stick pans, research on these products found the presence of PFAS (Link, 2019). PFAS are also found in water samples taken from different sources, biological samples, and foods. However, the number of studies on PFAS carried out in India from 2010 to 2021 is less than that carried out in other countries. Specifically, US and China conducted extensive research to resolve the problems associated with PFAS (Figure 2).

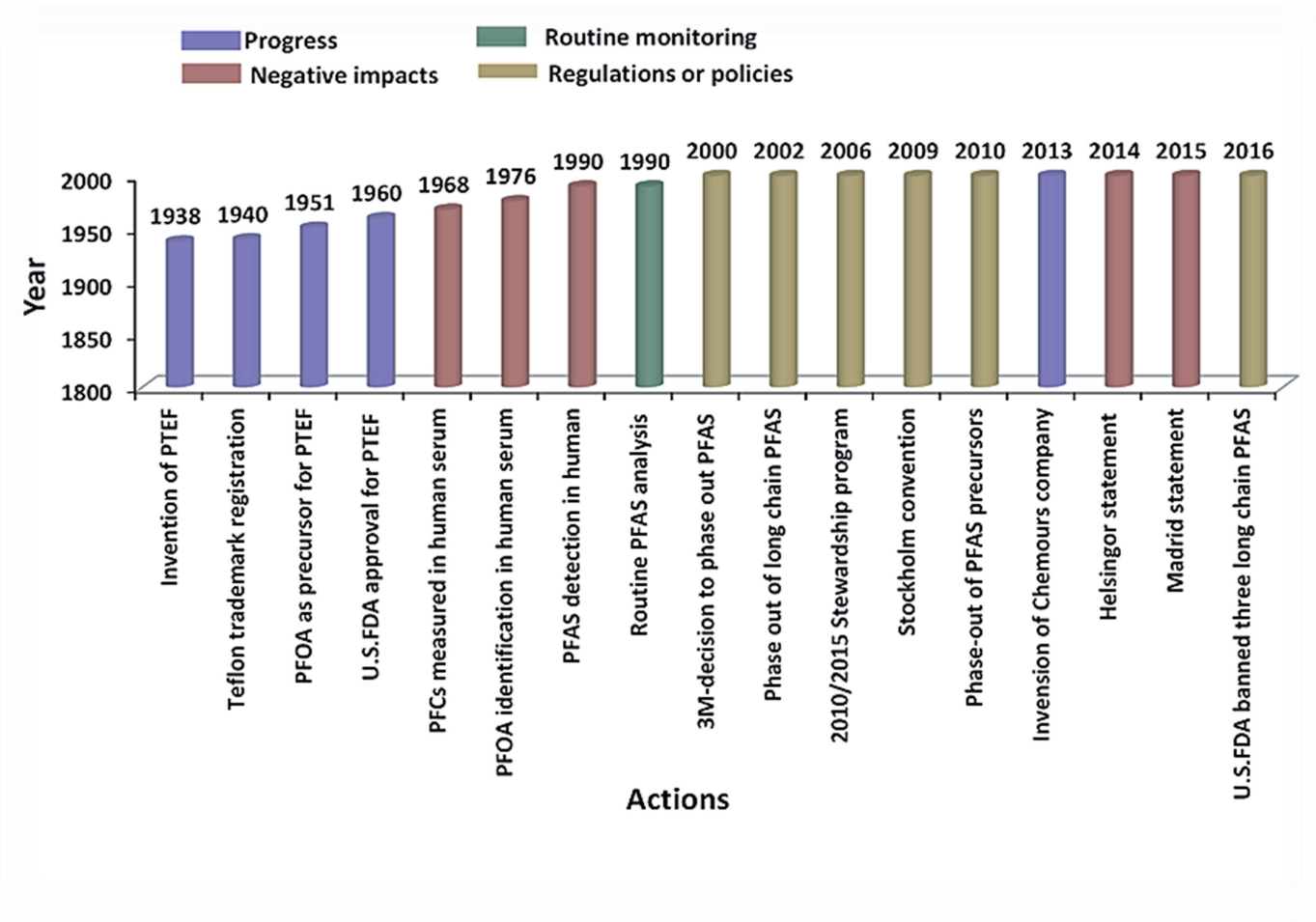


Figure 1. History of PFAS from the discovery to phase out (Geueke, 2016).

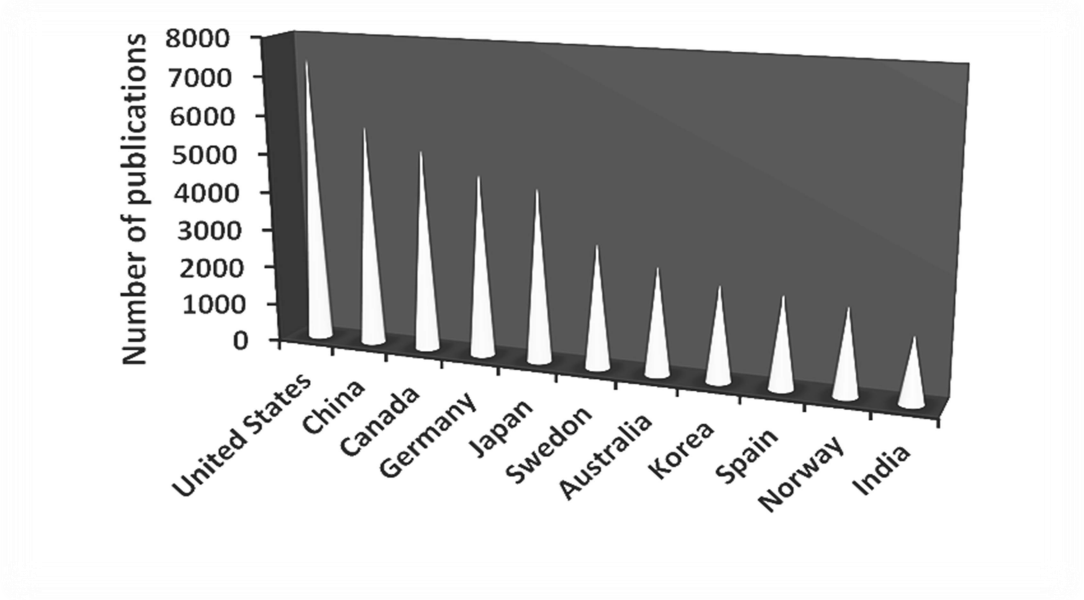


Figure 2. The total number of publications related to PFAS from 2010 to 2021 (Data extracted from Google scholar, July 2021 with the keyword of PFAS, country name).

Meanwhile, large numbers of newspaper articles have been published on the adverse impacts of PFAS. These articles highlighted the relationship between fast food, non-stick cookware, cosmetics, and carpets, and PFAS exposure (Table 1). All these articles point out that although India participated in the 2006 and 2009 Stockholm Convention, guidelines/regulations for PFAS have not yet been set.

This report is organized as follows: Section 1 covers a brief introduction and various aspects of PFAS pollution. The chemical classification and physiochemical features of PFAS are discussed in section 2. The toxicological impacts of PFAS are summarized in section 3. A brief history of PFAS production, phase-out, and emission profile is detailed in section 4. Sources and distribution of PFAS in the environment and human health have been evaluated in section 5. The prevalence of PFAS in different environmental compartments (e.g., Ground Water (GW), surface water, drinking water, and WW) are discussed in section 6. PFAS treatment technologies for industrial effluent, WW, point of entry treatment systems, and incineration-based PFAS removal from the soil are listed in section 7. The amended regulations and protocols to minimize the pollution of PFAS are summarized in section 8.

Table 1. Newspaper and magazine articles published in India related to PFAS exposure to humans

S. No	Publishing date	News Paper/ Magazine	Title	Key statement	References
1	05-Jul-10	The Telegraph Online	Trouble as you boil and bubble	Long-term exposure to non-stick cookware could present a health hazard	(Mitra, 2010)
2	21-Aug-11	The Times of India	Killers in the kitchen	Adverse impacts of PFAS used in non-stick cookware coatings	(India, 2011)
3	25-Oct-19	The Swaddle	Fast food packaging — not just the food itself — is harmful to our health	Accumulation of PFAS by consuming fast food and its health impacts highlighted	(Matta, 2019)
4	09-Feb-20	My India	Is it time to discard your non-stick cookware?	Briefly stated the health hazard related to PFOA and PFOS emitted from non-stick cookware	(Duhan, 2020)
5	07-Oct-20	The Swaddle	What you need to know about the chemicals we're exposed to every day and	PFAS occurrence in non-stick cook-wares, canned water, paint, and carpets briefed	(Desai, 2020)
6	23-May-21	The Week	India's infertility war: exposure to chemicals lowers sperm count, egg quality	Highlighted the relationship between infertility and PFAS	(Jaiswal, 2021)
7	16-Jun-21	The Swaddle	High levels of toxic chemicals found in top make□up brands' products: study	Cosmetics is a point source of PFAS exposure to human	(Rakshit, 2021a)
8	01-Sep-21	The Swaddle	Worrying' levels of toxic chemicals found in everyday objects, places: study	Air is also a distributor for PFAS-Indoor air pollution and its sources described	(Rakshit, 2021b)

2. CLASSIFICATION AND PROPERTIES OF PFAS

PFAS can be classified into the polymer and non-polymer substances (ITRC, 2020; Vo et al., 2020). Perfluoroalkyl Acids (PFAAs) and Perfluoroalkane Sulfonamide (FASAs) are the main subgroups of PFAS (Figure 3). Fluorotelomer, FASA substances, and Polyfluoroalkyl Ether Carboxylic Acids (PFCA) belong to PFAS. Polymer-based PFAS include Fluoropolymers (FP), Polymeric Perfluoropolyethers (PEPE), and Side-Chain Fluorinated Polymers. The most identified and studied PFAS are Perfluoroalkane Sulfonic Acids (PFSA) and Perfluoroalkyl Carboxylic Acids (PFCA). PFSAs typically have linear fluoroalkyl chains with sulfonic acid or sulfonate groups. Perfluorooctane sulfonic acid (PFOS) is the most widely used surfactant. Due to its extensive usage and associated toxicity, it was included in the "global voluntary phase-out" action of 3M Company in 2002 (Geueke, 2016). PFCA is another subgroup of PFAAs with the chemical formula of $CF_3(CF_2)_nCOOH$. PFOA is the widely used emulsion in the polymerization process of fluoropolymers. PFOS and PFOA are the most harmful, widely used, and persistent PFAS (Table 1) (Espana et al., 2015).

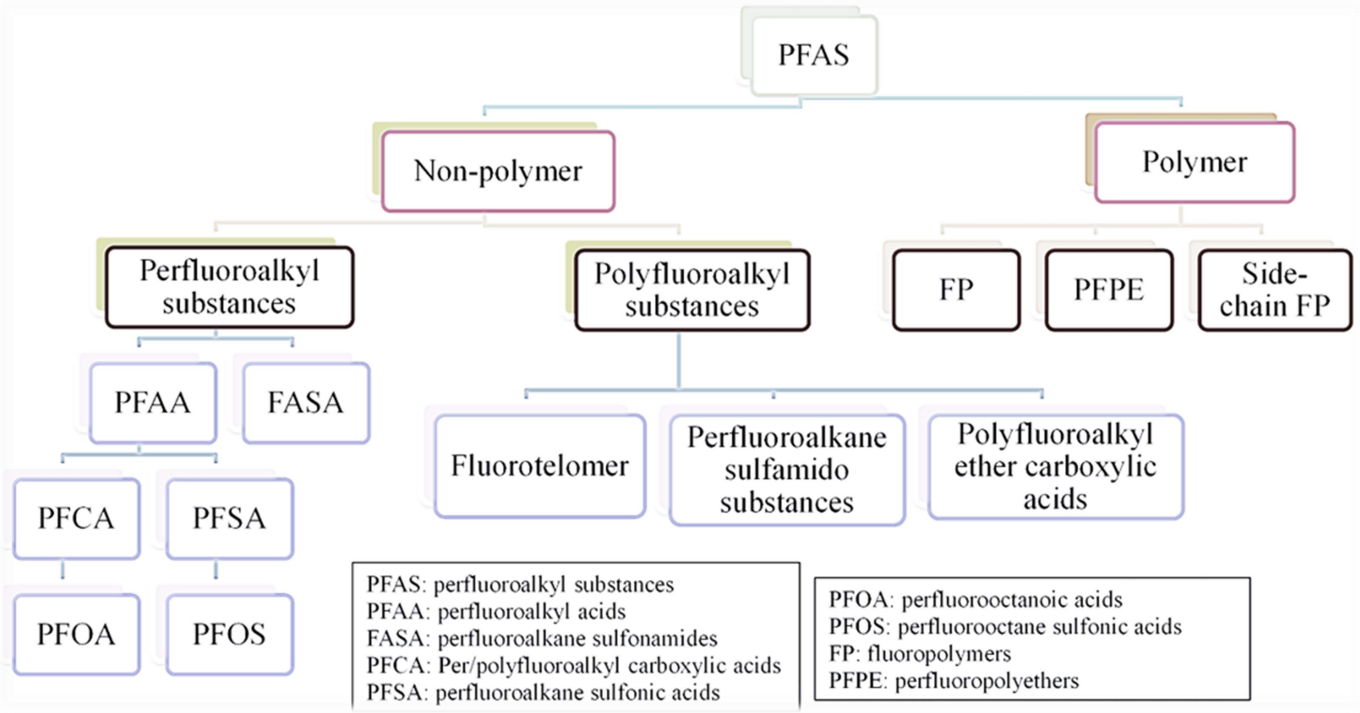
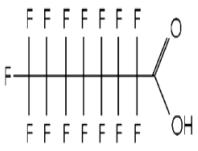
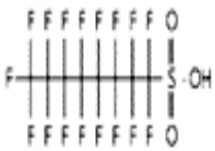
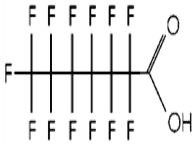
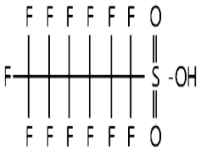


Figure 3. Family tree of PFAS (Modified from ITRC, 2020)

For naming FP, two different nomenclatures according to their: (a) functional group and (b) chain lengths have been used. In this regard, European Union (EU) authorities classified the PFCS as holding lesser than seven perfluorinated carbons belonging to the short-chain. PFCAs having seven or higher perfluorinated carbons are categorized as long-chain. Similarly, PFSAs are classified as short-chain and long-chain, depending on the number of perfluorinated carbons. US EPA regulates the short-chain PFCA as molecules containing eight carbon atoms and PFSA with six carbon atoms. Long-chain PFAS poses higher toxicity than short-chain PFAS (Abunada et al., 2020). A recent epidemiological study found that the toxicity of short-chain PFAS was 5-500 fold lower than long-chain PFAS (Menger et al., 2020). This higher toxicity of long-chain PFAS is due to their higher hydrophobicity (e.g., log K_{ow}: 3.16 to 5.3) than short-chain (e.g., log K_{OW}: 0.5 to 4.6) (Vo et al., 2020). The strong bioaccumulation nature of long-chain PFAS prompted their phase-out by governmental agencies, whereas short-chain PFAS have been used as alternatives to long-chain PFAS. The presence of a strong C-F bond (536 kJ mol^{-1}) restricts the removal of PFAS using traditional treatment (Simon et al., 2019). Physicochemical properties of commonly found PFAS in India are presented in Table 2.

Table 2. Physicochemical properties of most detected PFAS in India

Characteristics	PFOA	PFOS	PFHxA	PFHxS
CAS number	335-67-1	1763-23-1	307-24-4	355-46-4
Abbreviation	Perfluorooctanoic Acid	Perfluorooctane Sulfonate (PFOS)	Perfluorohexnoic Acid	Perfluorohexane Sulfonate
Molecular formula	$C_8HF_{15}O_2$	$C_8HF_{17}O_3S$	$C_6HF_{11}O_2$	$C_6HF_{13}O_3S$
Molecular structure				
Molecular weight (g mol^{-1})	414	500	314	400
Water solubility (25ºC) mg L^{-1}	3.3×10^3	680	15×10^3	6.25
Melting point (ºC)	40-50	>400		
Boiling point (ºC)	189-192	249	157	239
Vapour pressure at 25ºC (mm Hg)	0.53	2.48×10^{-3}	1.98	0.0046
Octonal-water partition coefficient ($logK_{OW}$)	4.81	4.49	3.48	3.16
Henry's law constant at 25ºC atm-cu m mol^{-1}	9.08×10^{-2}	NM	NM	4×10^{-4}
Half-life	Atm: 31 d water: >256 y (at 25ºC)	Atm.: 114 d water: >41 y (at 25ºC)	Atm.: 31 d	Atm.: 115 d

NM: Not measurable; Atm.: Atmosphere, The data source: PubChem.


Regular monitoring of
PFAS concentrations
in different
environmental media,
food products,
utensils,
household products,
and
cosmetics is needed

3. TOXICITY OF PFAS

PFAS in different aquatic organisms, humans, and mammals indicate the possible toxicity. In this context, suppression in algal growth, cell mutation in zooplankton, immunotoxicity in mollusk, and hepatotoxicity and immunotoxicity in mammals are reported (Liu et al., 2019). The research related to the toxicity of PFAS to humans is vast (Stahl et al., 2011; Fenton et al., 2021). This has been classified based on the circumstance of exposure (magnitude, duration, and routes of exposure) and factors connected with the exposed individual (e.g., age, sex, and health status). The major health impacts of PFAS include reduced immune function (Kvalem et al., 2020), thyroid dysfunction and autoimmunity (Inoue et al., 2019), liver disease and cancer (Salihović et al., 2020), lipid and insulin dysregulation (Qin et al., 2020), kidney disease and cancer (Rashid et al., 2020), reproductive and developmental disorders (Yuan et al., 2020), effects on breast milk, lactation, pre-eclampsia in pregnant women, infant birth weight reduction, tumor induction (NTP, 2020), and endocrine disruption (Gogola et al., 2019). Recently, the Centre for Disease Control and Agency for Toxic Substances and Disease Registry (ATSDR) indicated the potential connection between PFAS exposure and COVID-19 (ATSDR, 2020). Briefly, studies on humans and animals have already proven that PFAS exposure leads to reduced response to the vaccine by reducing the disease resistance capability (Looker et al., 2014; NTP, 2016; Grandjean et al., 2017b). Regular monitoring of PFAS concentrations in different environmental media, food products, utensils, household products, and cosmetics is needed. For example, the research conducted by Hairline International, Bangalore, India, stated that the hair loss in people aged between 18-44 using Teflon cookware was relatively higher (80%) than people using earthen, steel, and aluminum cookware (3%) (Bani Anand et al., 2016).

Understanding the PFAS' compound-specific toxicity is complicated by their diversity. Also, the fate pathway of PFAS is different for different organisms. For example, PFOS accumulates in aquatic organisms, while PFOA concentrates preferably in plants. Therefore, the toxicity and uptake pattern of each PFAS differs for animals, plants, and humans. The variation in the elimination kinetics complicates the toxicity evaluation in different species. Rats show sex-related bioaccumulation and chemical elimination patterns i.e., female rats excrete PFAS much faster than male rats. However, no differences were found between female and male mice in the elimina-

tion and bioaccumulation of PFAS. A similar observation was also noticed in humans. The estimated half -lives for long-chain PFAS in humans are longer (1 month to several years) than for other species (Fenton et al., 2021). Particularly, short-chain PFAS are excreted faster than longer ones.

3.1. Aquatic, terrestrial, vertebrates, and plants

PFOS enhanced the permeability of the cell membrane, which affected the growth of *Chlorella vulgaris* (Desjardins et al., 2001). PFOA caused dysfunction and abnormalities in the cell membrane of *Scenedesmus obliquus* (Feng et al., 2010) and significantly disturbed the growth chart of water fleas. The determined lethal concentration 50 (LC50) values for PFOS and PFOA of *Moina macrocopa* were 18 and 200 mg L^{-1}, respectively (Ji et al., 2008). Chronic toxicity of PFOS was found in *Chironomus tentans* (Chironomid) and damselflies at exposure concentrations less than 0.0023 and 0.01 mg L^{-1}, respectively. The significant growth reduction in *Daphnia Magna* at the exposure of 0.04 mg L^{-1} of Perfluorononanoic Acid (PFNA) was also reported (Lu et al., 2015). The hereditary effects of *Chironomus riparius* towards 0.01 µg L^{-1} of PFOA, PFOS, and Perfluorobutane Sulfonic Acid (PFBS) showed adverse effects (e.g., reduced growth) up to 10 generations (Marziali et al., 2019).

The PFAS compounds in surface water and sediment are toxic to benthic (aquatic) creatures. The benthic toxicity of PFAS lies in the range of moderate to high. However, there aren't enough studies, so it's impossible to conclude clearly how harmful these are. In terrestrial invertebrates, PFOS is highly toxic to honeybees (*Apis melifera*) (Beach et al., 2006). Detrimental reproductive disorders were observed in bumblebee (*Bombus terrestris*) at an Lethal Dose 50 (LD50) of 1.01 mg L^{-1} PFOS (Mommaerts et al., 2011). Exposure to PFOS, PFOA and PFNA by *Caenorhabditis elegans* decreased worm abundance up to the fourth generation (Tominaga et al., 2004). Oxidative stress and DNA damage were seen in earthworms (*Eisenia fetida*) after exposure to PFOS (Xu et al., 2013). Similarly, *E.coli* and *Caenorhabditis elegans* experienced stress profiles in the development and immunity (Stylianou et al., 2019). The calculated LD50 values for earthworm (*Eisenia fetida*) to PFOS were 84-447 mg kg^{-1}. Danish Ministry of Environment found that PFAS poses moderate to high toxicity to earthworms, particularly by causing reproductive disorders (PFOA and PFOS) (DMoE, 2015). PFBS decreased the lifespan of earthworms at concentrations of 1 mg kg^{-1} (Karnjanapiboonwong et al., 2018).

The toxicity of invertebrates was found to be dependent on the soil conditions (type of soil (sandy or clay), organic matter, and pH) (Princz et al., 2018). The LC50 values of PFOS for Rainbow trout (*Oncorhynchus mukiss*) and fathead minnow (*Pimephales promelas*) were 7.8-22 mg L^{-1} and 9.1 mg L^{-1}, respectively (Palmer et al., 2002). The saltwater acute survival toxicity for Rainbow trout was 13.7 mgL^{-1} (Palmer et al., 2002). Growth toxicity after exposure to PFNA (LOEC: 0.01^{-1} mg L^{-1}) was also reported (Zheng et al., 2012). The LC50 values in amphibians are dependent on the exposure duration. For example, LC50 of PFOS for leopard frog was 12.5 mg L^{-1} at one week and 6.2 mg L^{-1} at five weeks. Also, it causes growth inhibition and malformation (Ankley et al., 2004). PFOS is highly toxic for marine and aquatic organisms, but its toxicity was lesser than Perfluorohexane Sulfonate (PFHxS) for leopard frogs (Brown et al., 2021). Exposure to PFOA during the gestation period induces a delay in organ development in mice (Wolf et al., 2007), reduction in adaptive immune response (Wei et al., 2008), alters the sex hormones, and induces ovarian effects (Du et al., 2013). In contrast to PFOA, the toxicological impact of PFOS is much more prominent. For example, the toxicity of PFOS to *Brachionus calyciflo-*


The benthic toxicity of PFAS lies in the range of moderate to high. However, there aren't enough studies, so it's impossible to conclude clearly how harmful these are...

rus was ~2.5 times larger than PFOA, and the health impacts were also passed on to the next generation (Zhang et al., 2012b). PFOS also caused oxidative stress to emerald mussels and disrupted the function of thyroid glands via modulating the hypothalamic-pituitary-thyroid gene (Shi et al., 2009). Concentration-dependent inhibition of longer-chain PFAS (e.g., PFNA and Perfluorodecanoic Acid (PFDA) to p-gp protein transport in mussels was also reported (Stevenson et al., 2006). It also showed immunotoxicity in green mussels (Liu and Gin, 2018). PFAS also caused liver tumors in rodents by activating Peroxisome Proliferator-Activated Receptor-α (PPAR-α) (Takacs and Abbott, 2007). Specifically, long-term exposure to PFOS caused chronic toxicity in cynomolgus monkeys. The general effects of PFOS, PFOA, and PFHxS include growth and reproductive disorders, and their corresponding Lowest Observed Effect Concentrations (LOECs) ranges from 0.05-0.12 mg L^{-1} for toad, frog, and salamander (Abercrombie et al., 2021).

The LD50 for the acute exposure to PFOS by northern bobwhite quail and the mallard duck were 61 and 150 mg kg^{-1} d^{-1}, respectively (Newsted et al., 2006). Cumulative exposure of PFOA, PFOS, and PFHxS (0.00245-0.0031 mg kg^{-1} d^{-1}) resulted in female body weight reduction and reduced impaired hatching success (Newsted et al., 2006). PFOS also affects the liver in the early incubation stage of leghorn chicken. The field studies on tree swallows showed reduced hatching success, but the results showed a positive relationship in the areas contaminated with other micropollutants (Custer et al., 2019). The dispersion of PFAS in atmospheric sources also impacted the population of flamingos (De Vries et al., 2017). Bottlenose dolphin and wood mouse showed a significant correlation between liver PFAS concentrations and multiple immunological, hematopoietic, and hepatic function disorders (Hoff et al., 2004; Fair et al., 2013).

The PFOS exposure to diverse plant species (e.g., onion, soybean, tomato, flax, and lettuce) decreased the survival at the concentrations of 57 to 1,000 mg kg^{-1} (Brignole et al., 2003). *Triticum aestivum* was the most sensitive plant, with a 30-day No Observed Effect Concentration (NOEC) of 1 mg kg^{-1}. Zhao et al. (2011) found that PFAS toxicity is associated with soil organic matter; higher organic carbon content in the soil increases the phytotoxicity factor.

3.2. Humans

PFAS are potentially hazardous chemical compounds whose adverse health effects depend on two factors, circumstance (e.g., magnitude, strength, and route) of exposure and individual factors (e.g., age, sex, and genetic predisposition). Generally, PFAS increases disorders related to the immune system, thyroid function, reproductive, liver disease, and cancer (Figure 4) (Fenton et al., 2021). Recent studies also discovered a connection between PFAS concentrations and anemia and claimed that they cause diabetes (Conway et al., 2018). Researchers have also found a correlation between diphtheria antibodies developed from vaccines and PFOS in children. This kind of decreased immunological response is prevalent in children under 13 years of age (Grandjean et al., 2017a). PFOS also positively correlates with rubella, mumps, and hemophilus influenza vaccinations (Abraham et al., 2020). The increased concentration of PFOS in blood down-regulates the C-reactive protein and arrests the inflammation system in adults and children (Genser et al., 2015). Frequent exposure to PFAS in infancy and childhood results in increased chances of atopic dermatitis and respiratory tract infections (Kvalem et al., 2020). The increased serum PFOA concentrations showed a linear trend with the incidence of ulcerative colitis (Steenland et al., 2013). PFOA exposure causes hyperthyroid disease in women and hypothyroid dis-

ease in men (Rappazzo et al., 2017).

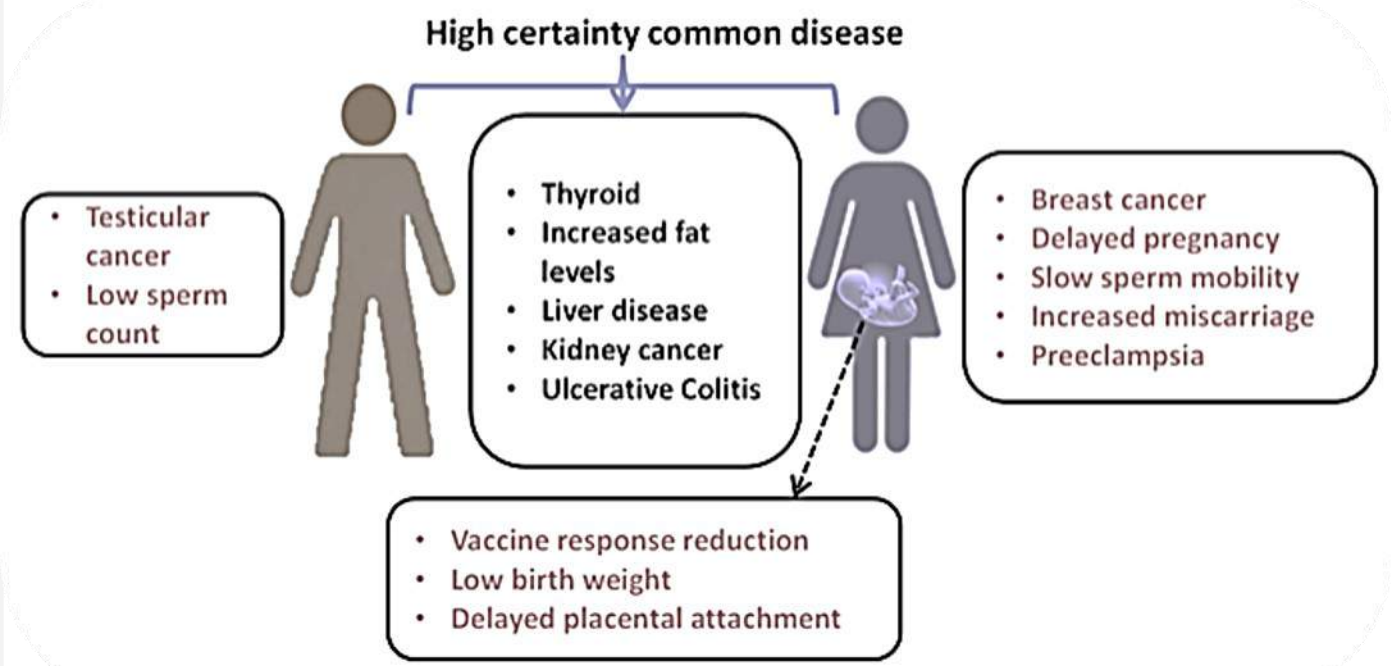


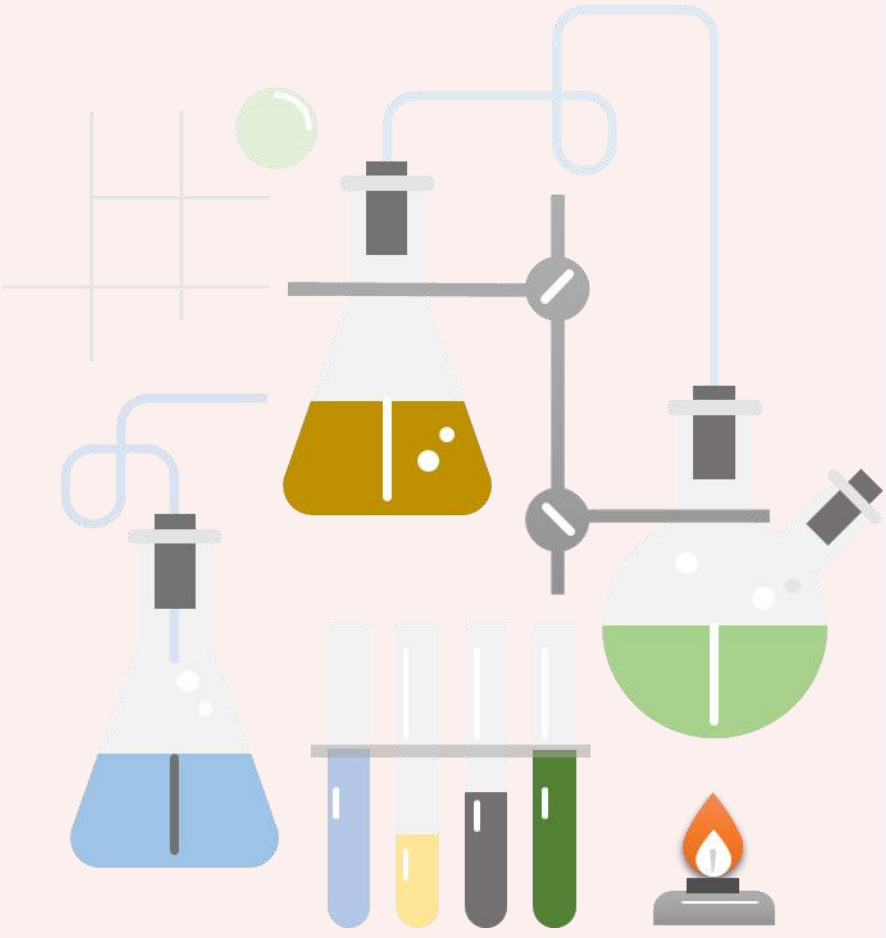
Figure 4. Human health effects of PFAS

The women and children affected by antithyroid peroxidase antibodies are at high risk. PFAS induces thyroid dysfunction and disease in children under 12 years (Lee and Choi, 2017). The mechanism linked with PFAS exposure and thyroid hormone interferes with thyroid peroxidase enzyme activity (Song et al., 2012). The increase in thyroid stimulating hormone during pregnancy transfers the PFAS to the fetus via maternal serum and cord blood (Forhead and Fowden, 2014). The liver is considered a primary storage place for PFAS. Specifically, long-chain PFAS alters the liver enzymes (e.g., alanine aminotransferase), and the effect was dominant in obese patients (Gallo et al., 2012). Non-alcoholic fatty liver disease is the common endpoint of PFAS exposure in humans, and in severe cases, it results in liver cancer (Bassler et al., 2019; Program, 2020). Workers heavily exposed to PFAS are more frequently diagnosed with cirrhosis and liver cancer than the general population (Girardi and Merler, 2019). Increased cholesterol levels and dyslipidemia are the two major lipid dysregulation outcomes of PFAS exposure (Alderete et al., 2019). Epidemiological studies related to human PFAS exposure indicated the association between reduced kidney function and chronic kidney disease. Severe exposure can result in kidney cancer (Vieira et al., 2013; Ferrari et al., 2019). Exposure to PFOA results in sperm mobility, less sperm penetration into viscous media, change in the length of the menstrual cycle, and progesterone imbalance (Lum et al., 2017; Salihović et al., 2020; Yuan et al., 2020). PFAS tends to travel to the placenta, to breast milk and increases serum PFAS concentrations in newborns more than in mothers (Gyllenhammar et al., 2018). PFOA exposure during pregnancy results in preeclampsia and low birth weight (Lam et al., 2014). Thus, PFAS pose deleterious effects in conception, pregnancy, and infant development.

3.3. Risk assessment

More than 4,700 PFAS are available in the market, and it is not possible to test the risks of each PFAS because it is a time and resource-intensive process (Cousins et al., 2020). In this regard, grouping strategies have been used to identify the risks associated with the particular set of PFAS to the environment and the general public, and biota. Two types of grouping approaches are commonly practiced to subgroup the PFAS; namely a) grouping based on the physicochemical properties of PFAS (e.g., molecular size, occurrence, solubility, persistence, toxicity, and mobility) and b) grouping based on the estimation of cumulative effects within particular exposure limits (Cousins et al., 2020). In this section, the risk assessment methods are discussed in detail.

Generally, long-chain PFAS are more bio-accumulative than short-chain. For example, the calculated bioaccumulation factor of PFOA in European carp (~3,200 L kg^{-1}) and rice-fish (13,600 L kg^{-1}) indicated their potential toxicity in biota (J-CHECK). Similarly, the bioaccumulation behavior of Perfluorotripropyl Amine, Perfluoro-2-[(Propoxy)]-1-Proponate, PFSAs, and PFCAs have also been reported (Meyer et al., 1992; Shi et al., 2016). The traditional bioaccumulation analysis is time-consuming, and without proper control experiments, the results lead to analytical bias. In such regard, computational and empirical methods based on the measurement of protein binding affinity have been developed (Cheng and Ng, 2018; Yang et al., 2020). These methods are beneficial for assessing the bioaccumulation potential of short-chain PFAS. Contrastingly, it often overestimates the risks of selected PFAS because the persistence duration of PFAS in biota and humans is shorter than in the environment. The German Environmental Agency developed a risk assessment approach based on the Persistence, Mobile, and Toxicity (PMT) of PFAS. Short-chain PFAS also considered in the risk assessment because they are Very Mobile (vM) and Very Persistent (vP). This type of PMT or vP-vM approach calculates the risk of PFAS via the persistent nature without considering the bioaccumulation and toxicity of PFAS.



Though different approaches have been developed for the risk assessment of PFAS, the arrowhead approach is the apt method to identify the cumulative risks of PFAS and its precursors. Initially, 3M company developed this approach, and it has been applied globally to regulate PFAS and its precursors (3M, 2000). Nevertheless, these approaches overlooked the risks from their

intermediate or by-products and precursors. For example, 6:2 Fluorotelomer Alcohols (FTOHs) were more toxic than Perfluorohexnoic Acid (PFHxA), and 6:2 FTOH is a precursor for PFHxA (Rice et al., 2020). Such disadvantages of the arrowhead method are balanced by the Total Oxidizable Precursor (TOP) method. The TOP assay is mainly used to identify the risks of PFAS and its precursors and transformation products in drinking water (Houtz and Sedlak, 2012). The TOP assay method is not supportive in simulating the environmental fate of these released PFAS and its metabolites. The TOP assay method is not yet standardized and warrens future research.

The simple additive approach helps frame the environment and health-based guidelines for PFAS via existing analytical methods. For example, US EPA drinking water guidelines for a sum of PFOA and PFOS were 70 ng L^{-1} and framed based on the additive approach (US EPA, 2020). Likewise, the Swedish government has amended 90 ng L^{-1} as a guideline value for 11 different PFAS. The main advantage of this method is that it provides cumulative toxicology information on PFAS belonging to the same subgroup (SFA, 2013). Contrastingly, it failed to predict the individual risk factor of each PFAS belonging to the same group. The precautionary grouping approach uses the intrinsic property of the PFAS and divides them into several subsets. Accordingly, it helps to reduce the "non-essential use of PFAS" (Cousins et al., 2019). However, the precautionary approach is similar to bioaccumulation, where a longer time is required to identify the intrinsic properties of each subset of PFAS. In consideration of the adverse effect of PFAS, many countries have phased out long-chain PFAS and continue to use short-chain or PFAS alternatives. Thus, government authorities can use one of these risk assessment approaches to identify the hazards of PFAS to biota, humans, and the environment.

3.4. Ecotoxicity of PFAS

Physicochemical properties (fluoroalkyl chain length, log K_{OW}, etc.) of PFAS decide the strength of bioaccumulation and magnification of PFAS in biota. For example, an increase in the fluoroalkyl chain length increases the K_{OW} and elevates the Biomagnification Factor (BMF) (Chen et al., 2021). The functional groups in the fluoroalkyl chain affect the strength of the binding capability of PFAS in the biota. Likewise, PFSA possesses a higher K_{OW} than PFCA with the same chain length. So, the BMF of PFSAs (e.g., PFOS accumulation in the liver of predators was 427 ng g^{-1}) higher than PFCAs (e.g., PFOS accumulation in the liver of predators was 5.98 ng g^{-1}) (Ng and Hun-

gerbühler, 2014). The PFAS initially interacts with membrane phospholipids and proteins (e.g., Serum Albumin), and depending on the physicochemical properties of PFAS, the BMF is varied. Generally, PFAS with lesser than 10^5 K_{OW} is eliminated from the liver of aquatic animals and extends their distribution in the surrounding (Kelly et al., 2007). In the case of lung-breathing animals and birds, the elimination of PFAS is not yet reported. Additionally, long-chain PFAS (C>6) possesses a slower elimination rate than short-chain. In consideration of the different elimination features, different models have been used to identify the BMF. For example, membrane-lipid models are employed to identify the BMF of aquatic animals. In the case of lung-breathing animals and birds, protein binding models were used.

The occurrences and toxicity of PFAS towards different species are provided in section 3.1. However, the occurrences of PFAS in animals and birds are related to the geography and food preferences. For example, the proportion of PFOS in marine apex predators (58.1%) was higher than in terrestrial apex predators (41.7%) (Chen et al., 2021). The food chain length of terrestrial apex predators is limited, and it is site-specific. Conversely, the food chain structure of marine apex predators covers a wide range, thus enriching the concentrations of PFAS. In the case of PFCAs, Perfluoroundecanoic acid (PFUnDA) (13%) and PFNA (20.3%) were dominant in apex predators. The occurrence of PFCAs with C>12 was not detected in any of the apex predators, and it is related to their lower usage and bioavailability. Accumulation of PFAS in the biota is different for different species. In animals, PFSA commonly accumulates in the kidney (Gui et al., 2019), whereas in birds, a higher concentration of PFAS noticed in the liver and feathers (Meyer et al., 2009). Among tropic animals (e.g., pinnipeds (liver 11.5-138 ng g^{-1})), polar bears registered maximum PFOS concentration (liver 350-3,270 ng g^{-1}) because of marine food habits (Müller et al., 2011). Recent reviews related to the ecotoxicity of PFAS indicated that the acute toxicity of PFAS is under the threshold value, while their chronic toxicity necessitates further investigation. For example, the bioaccumulation of PFAS in the brain of polar bears results in changes in progesterone production and distribution, and at chronic levels, it creates endocrine disruptions (Lau et al., 2007). So Hazard Quotient (HQ) is used to calculate the risk of pollutants on the ecosystem (Sinclair et al., 2020). It uses measured levels of contaminants in the environment and the concentration that causes an evident effect. The equation of HQ is

$$HQ = \frac{Measured\ environmental\ concentration\ (MEC)}{Predicted\ no\ effect\ concentration\ (PNEC)}$$

The HQ less than 0.1 indicates mild risk, and HQ higher than 1 indicates high risk. Accordingly, Sardiña et al. (2019) calculated the HQ of PFOS and PFOA from the WWTP effluent of different countries. The calculated HQ of PFOA and PFOS of the US, Europe, Australia, China, Hong Kong, and Taiwan were less than 0.1, indicating moderate risk. On the other hand, the PFOA HQ value for Singapore exceeds 1 and indicates a higher risk (Yu et al., 2009). Selvaraj et al. (2021) calculated the PFOA and PFOS HQ of riverine, mangrove, and estuarine ecosystems of Tamil Nādu, and the values lie in the range of 2.3×10^{-7} and

4.3×10^{-5}. This value is well below HQ 0.1, indicating a lower risk. The author has concluded that the detected concentrations of PFOS and PFOA in the study area (Pichavaram Mangrove and Gulf of Mannar) had no significant ecotoxicity on aquatic organisms.

While calculating the ecotoxicity of PFAS, another important parameter to consider is PFAS alternatives. The phase-out of long-chain PFAS (e.g., PFOS, PFOA, and PFHxS) increased the use of PFAS alternatives. In such a way, high concentrations of PFAS alternatives such as Perfluoro Butanoate (PFBA) (36 ng g^{-1}) were found in polar bears (Boisvert et al., 2019), and 6:2 Chlorinated Polyfluorinated Ether Sulfonic Acids (Cl-PEFSA) in finless porpoises (482 ng g^{-1}) (Wang et al., 2021). Although short-chain PFAS are known to cause less bioaccumulation than long-chain, it is more persistent than long-chain (Feng et al., 2018). Due to their higher persistence, it develops higher toxicity. For example, animal toxicity studies envisioned that 6:2 Cl-PEFSA might pose embryo, cardiac, and neurotoxicity (Wang et al., 2019b). These investigations inform those strict regulations are necessary to avoid the ecotoxicity developed by PFAS alternatives. On the other hand, treatment methods are needed to control the emission/distribution of PFAS from the contaminated sites to minimize the threat to biota.

4. PRODUCTION AND EMISSION OF PFAS

4.1. PFOAs and related products

The production of PFCAs via ECF began in 1947. Between 1951 and 2004, 3,600-5,700 t of PFOA were produced in the chemical name Ammonium Perfluorooctanoate (APFO) (Prevedouros et al., 2006). The largest production sites of PFOA were in US, Belgium, and Italy. Japan had small-scale producers of PFOA (OECD, 2005). According to the survey conducted in 1999, globally,~260 t of PFOA was produced, and among those US contributed 20 t of PFOA production, which accounts for 5-10% of global production (EPA, 2002). From the toxicity studies, the adverse effects of PFOA were identified in 2000. Major PFOA manufacturers in the US, EU, and Japan announced the global phase-out for the PFOA products containing C_6-C_{10} between 2002-2013 (3M, 2002; Daikin, 2013; DuPont, 2013). Major PFCA and FP manufacturers participated in US EPA Stewardship Program (EPA2010/15), reducing the production and usage of PFCAs and their precursors by 2015 (EPA, 2015). On the other side, manufacturers also started using PFCAs-related alternatives and developed recovery technologies to reuse the WW streams (Yunzhi et al., 2009). These regulatory activities decreased the production rate of PFOA in the US, EU, and Japan. But the production of PFCAs and related products shifted to China and India. Accordingly, the PFOA production rate in China rose from 6.6 to 64 kt y^{-1} from 1999 to 2012 (Wang, 2006; Kaelin et al., 2012). PFOA production reached a maximum in China in 2009 (50-80 t) and hosted five PFCA production sites (Mei, 2008). At the same time, India also started to produce PFCAs in 2010, which gradually increased in the following years (e.g., in 2011 and 2012, the production of PFCAs was 2.3 kt and 7.5 kt, respectively) (Kaelin et al., 2012). In 2011, NDRC added PFOA-relevant technology to the Catalogue for the Guidance of Industrial Structure Adjustment and introduced the restrictions related to the new installation plants to produce PFOA and its precursors (NDRC, 2011). Chinese Ministry of Environmental Protection classified PFOA and its related products under the *Comprehensive Catalog for Environmental Protection* due to its high pollution and environmental risk (MEP, 2008). The global emission of PFOA didn't decrease, and only a geographical shift happened.

The estimated global emission of PFOA from the production site was 90-970, 30-430, and 0-630 t for the years of 1951-2002, 2003-2015, and 2016-2030, respectively (Wang et al., 2014b). Likewise, FP manufacturers with PFOA (i.e., PFOA-related products) emission inventory was 1,220-6,560, 660-3,870 and 0-4,520 t for the years of 1951-2002, 2003-2015, and 2016-2030, respectively. The use and disposal of PFOA and its related products may result in 90-320 t of emissions between 1951 and 2030. So, the overall PFOA-related production and usage emissions were 1,790-6,420 t between 1951 and 2030. This emission inventory analysis shows no decline in the global PFOA emission. US, EU, and Japan showed lesser emissions owing to their phase-out activities. On the other hand, the absence of legislative and

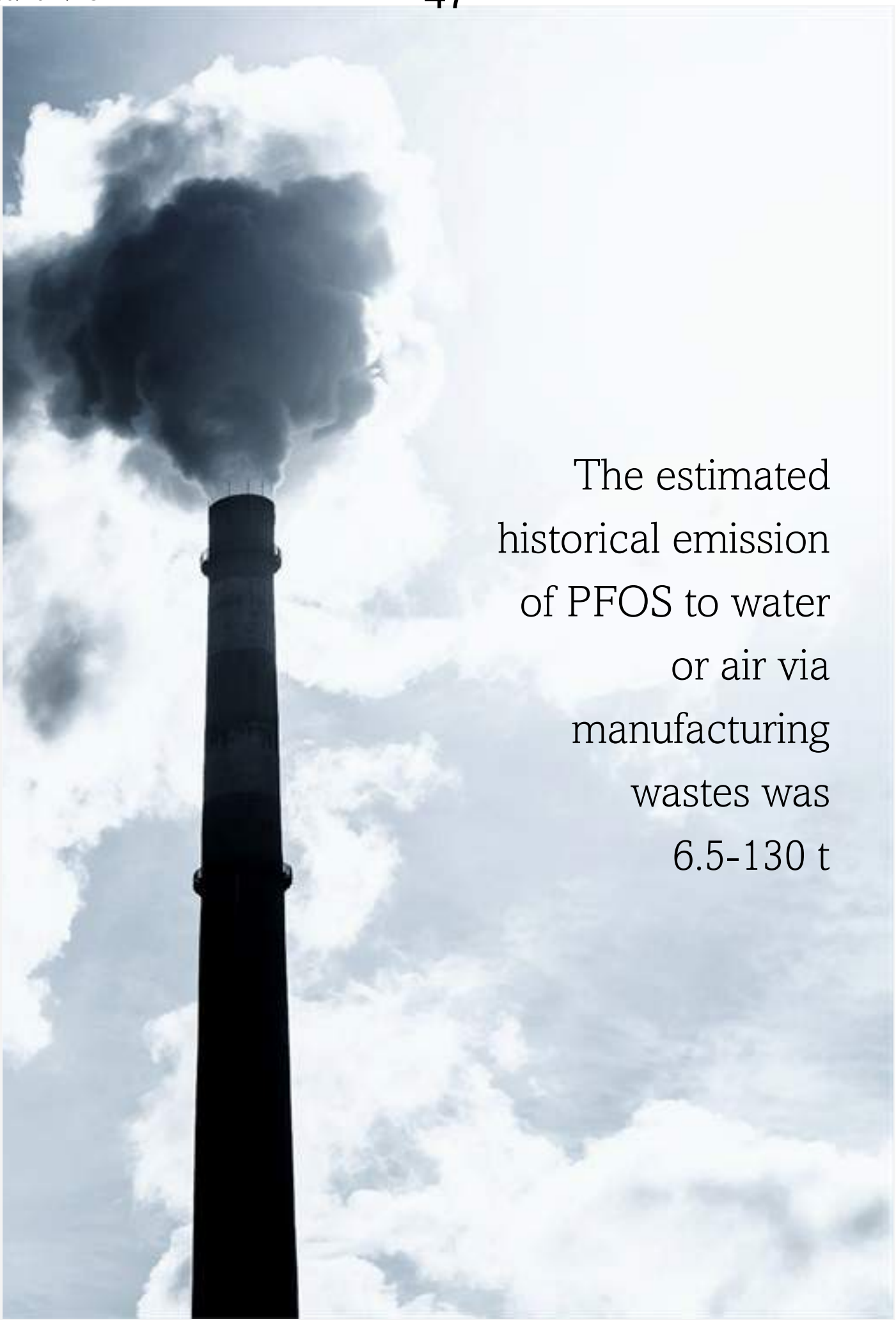
governmental policies caused the gradual increase in PFAS pollution in India, China, and Russia. Confidential Business Information has limited the transparency in producing PFOA and FP-related products.

4.2. PFOS and related products

Between 1970 and 2002, ~96,000 t of PFOS-related substances was produced, and 26,500 t were estimated as solid unwanted by-products. The production of PFOS peaked (5-fold enhancement) from 1975 to 1989 and relatively stabilized until 1990 (Paul et al., 2009). 3M is the main manufacturer of PFOS, and it has been estimated that 3M manufactured ~3,665 t of PFOS-related products in 2000 (OECD, 2002). The global production of PFOS was 4,650 t in 2000, and 3M contributed to 78% of the global PFOS-related chemicals production (OECD, 2002). According to the Organization for Economic Co-operation and Development (OECD) 2003 survey, the US, Belgium, and EU showed higher production/ import of PFOS related substances (OECD, 2005). Globally, 3M had PFOS-related plants in US and Belgium; other than 3M, six plants in the EU, six plants in Asian countries (e.g., Japan), and one in the southern part of America were produced PFOS related chemicals (Paul et al., 2009). A sharp drop in the production of PFOS related products was observed between 2000 and 2003. It is related to the voluntary phase-out in the production of PFOS by 3M. However, the OECD report showed a production volume of 303 and 3.5 t in 2005 and 2008, respectively (OECD, 2011). However, no such information is available for India and China.

Considering the adverse health and environmental impacts, 3M announced the phase-out in 2000 and stopped production in 2002 (Land et al., 2018). EU Marketing and Use Directive (2006/122/EC) banned PFOS and its related products in 2006. At the end of 2008, EU Directive allowed 0.005% of PFOS in PFOS -related products or alternatives (Land et al., 2018). In 2009 PFOS was listed under restricted chemicals for production and use by Stockholm Convention on Persistent Organic Pollutants. Meantime, China became a massive alternative production site for PFOS and fulfilled the global demand for PFOS and its related products. Accordingly, the production of PFOS related products in China increased from 30 to 247 t from 2002 to 2006 (Zhang et al., 2012a). However, the strict amendment of China's domestic Catalogue for the Guidance of Industrial Structure Adjustment related to PFOS decreased further PFOS production in China (e.g., 100 t in 2008) (NDRC, 2011).

The estimated historical emission of PFOS to water or air via manufacturing wastes was 6.5-130 t (Wang et al., 2014b). Industrial application of PFOS and its related substances caused high global production (420-2,700 t), followed by the use and disposal of consumer products (e.g., carpets: 205-1,000 t, apparel: 120-600 t, and artificial firefighting foam: 91-460 t) (Paul et al., 2009). Overall, the total emissions of PFOS in 1970-2000 were 450-2,700 t. Since 2002, a proper emission inventory analysis has not been conducted. So, the actual trend related to the current production, use, and emission of PFOS and related products is not known publicly.


The estimated
historical emission
of PFOS to water
or air via
manufacturing
wastes was
6.5-130 t

5. SOURCES AND DISTRIBUTION OF PFAS POLLUTION

PFAS possesses excellent repellent and stabilization properties. So, they are used in different products in day-to-day activities. This section details the possible sources of PFAS pollution and their fate in the environment.

5.1. Sources of PFAS

PFAS enters the environment via point and non-point sources. Industrial production, processing, transportation, and deposition of PFAS materials are some point sources (Liu et al., 2019), while non-point source includes emission of the PFAS and their precursor's through dry and wet deposition (Zareitalabad et al., 2013). The industries are preparing dispersants, fire extinguisher foam, floor polish, hair care products, fuel additives, fabric conditioners, and air fresheners act as primary point sources of PFAS emission (Liu et al., 2019) (Figure 5).

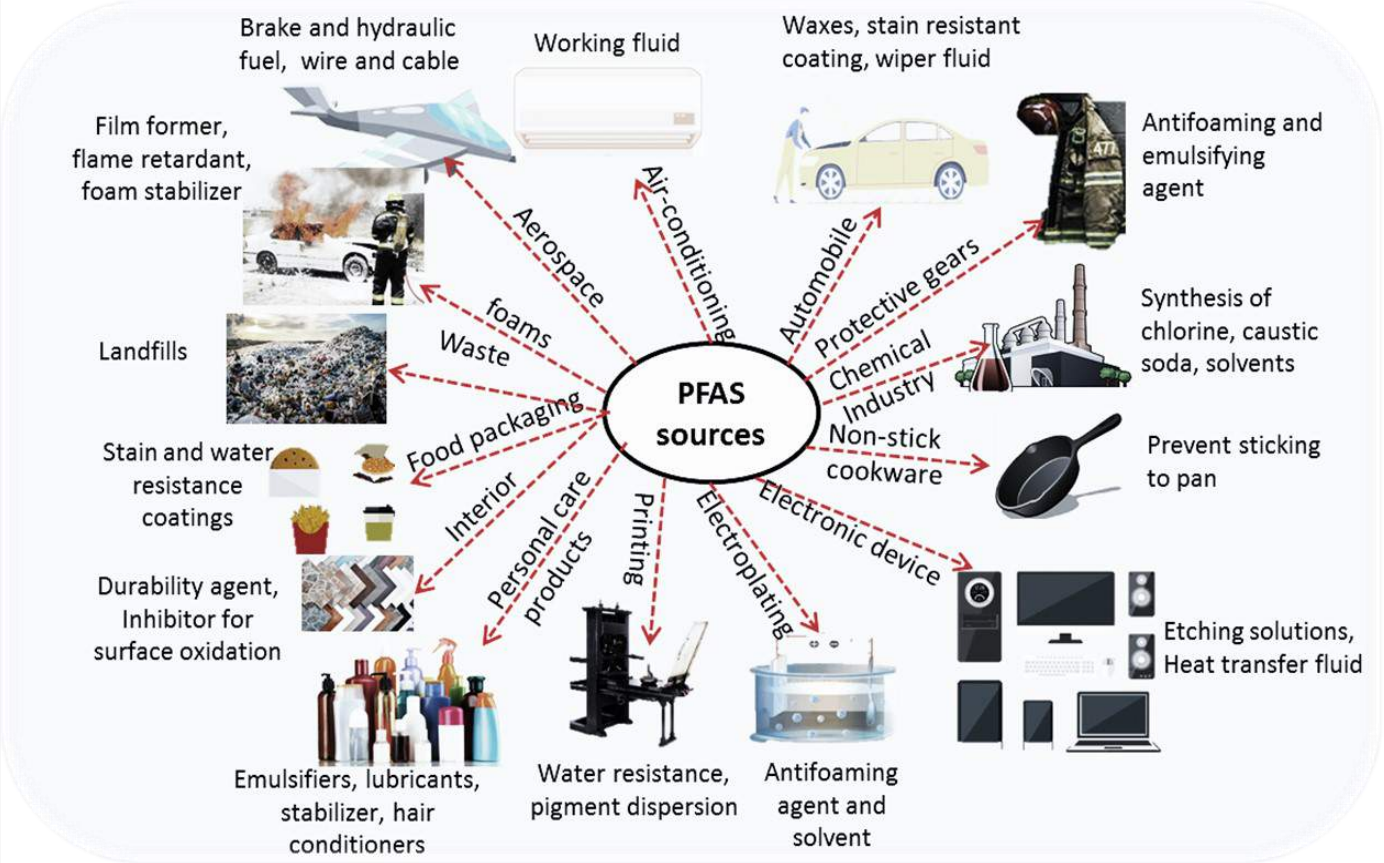


Figure 5. Sources of PFAS pollution in day-to-day activities (IIT Madras).

PFAS have been used for grease-proofing, water-resistance, processing aid, and surface-active agents in many industrial applications (Table 3). As a result, these industries listed in Table 3 contribute almost 65 -75% of point source PFAS pollution (ITRC, 2020). Fast food packaging materials are known as indirect point source exposure to PFOA, PFHxA, and PFBS (Schaider et al., 2017). This type of PFAS source creates a long-term (~40 years) negative impact on the environment and human health (US EPA, 2016d). PFAS used in the building and construction field is the source of indoor PFAS emissions. Short-chain PFCAs, Perfluoro Heptanoic Acids (PFHpA), C_8-C_{20}-PFAS, and Polytetrafluoroethylene (PTFE) are used to prepare oriented strand board, wood fiber insulation, lightweight concrete building blocks, and roof dome, respectively (Posner, 2013; EPA., 2015; Bečanová et al., 2016). Household products (e.g., water repellent agents, vessels, and decorative interiors) emit PFAS in the range of 0.9 to 1.8 ng g^{-1}. PFOS contributes 36% of the total emission caused by PFAS. The emitted PFAS from residential homes is considered an important sink for the atmospheric dispersal of PFAS (Wang et al., 2013b).

Table 3. Purpose of PFAS in different industrial applications (Source: ITRC, 2020).

S. No	Industry	Purpose	Main PFAS
1	Building and construction	Composite wood and oriented strand board	PFOA and PFOS
2	Cable and wiring	Water-resistant	PFCA
3	Metal finishing and plating	Mist suppressants	PFOS
4	Surfactants and FP production	Processing aid	PFOA
5	Paper products and packaging	Grease-proofing agent	PFOA, PFOS, PFHxA
6	Photolithography and semiconductor industry	Surface-active agent	PFAAs PFBS, PFHxS, PFOS, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA
7	Textiles, leather, and apparel	Stain, oil, and water repellent agent	PFAS

Besides the above sources, the usage of class B firefighting foams (e.g., aqueous film-forming foam) in military training bases is a well-known point source in the US and Australia (Darwin, 2011). Also, the slow release of stored PFAS-containing foam contaminates the soil and GW (ITRC, 2020). WWTP is the indirect point source for PFAS pollution. The detected concentrations of PFAS in the municipal and industrial WW were in the range of 465-638 and 10-1,000 ng L^{-1}, respectively (Kim et al., 2012; Arvaniti and Stasinakis, 2015). The conventional WWTP offers ~5-10% PFAS removals. Due to the phase transition (e.g., long-chain breaks into shorter-one), the negative removal efficiency was often observed for PFAS (Schultz et al., 2006; Ahrens, 2011; MWRA and MDEQ, 2019). The inefficiency of WWTP to remove PFAS results in the accumulation of PFAS in sludge. The lowest and highest concentrations of PFAS in sewage sludge were 241 and 7,304 ng g^{-1} (Kim et al., 2012). Also, the activated sludge and extended aeration systems result in atmospheric emission of PFAS near the WWTP in the range of 0.1 to 812 pg m^{-3}. Gaussian dispersion models estimated that an activated sludge system releases 110-320 g of PFAS

year^{-1} aeration tank^{-1}, and it is ~12 g year^{-1} tank^{-1} for extended aeration systems (Ahrens et al., 2011b; Shoeib et al., 2016). Further, using the PFAS containing solid sludge as biosolids and disposal in landfills induce soil contamination. This also provides a path to the food chain via plant uptake (Higgins et al., 2005). The indicator PFAS released from the landfill leachate is 5:3 Fluorotelomer Carboxylic Acid (FTCA), FTOHs, and PFBA (Ahrens et al., 2011a). Some other point sources of PFAS emission include discharge from automobile service (e.g., car-washing and waxing) facilities and sports areas (e.g., ski and snowboarding).

Industries manufacturing PFAS, improper deposition of activated sludge, and vapor deposition of fluorine alcohols are significant non-point PFAS sources (Ellis et al., 2004; Krusic et al., 2005) (Figure 6). Application of PFOS as mist suppressants in the fume hood of metal plating and finishing industry to reduce atmospheric pollution of toxic metal is the main source of atmospheric and WW/sewage sludge-related PFAS pollution (USEPA, 2012; OSHA, 2013). The release of high concentrations of PFOS from these activities resulted in their distribution in surface water, storm water, snow, and biota (e.g., fish) (ATSDR, 2008; MPCA, 2016).

5.2. Fate of PFAS in the environment

The relative environmental significance, discharge amount, and distribution of PFAS in the impacted

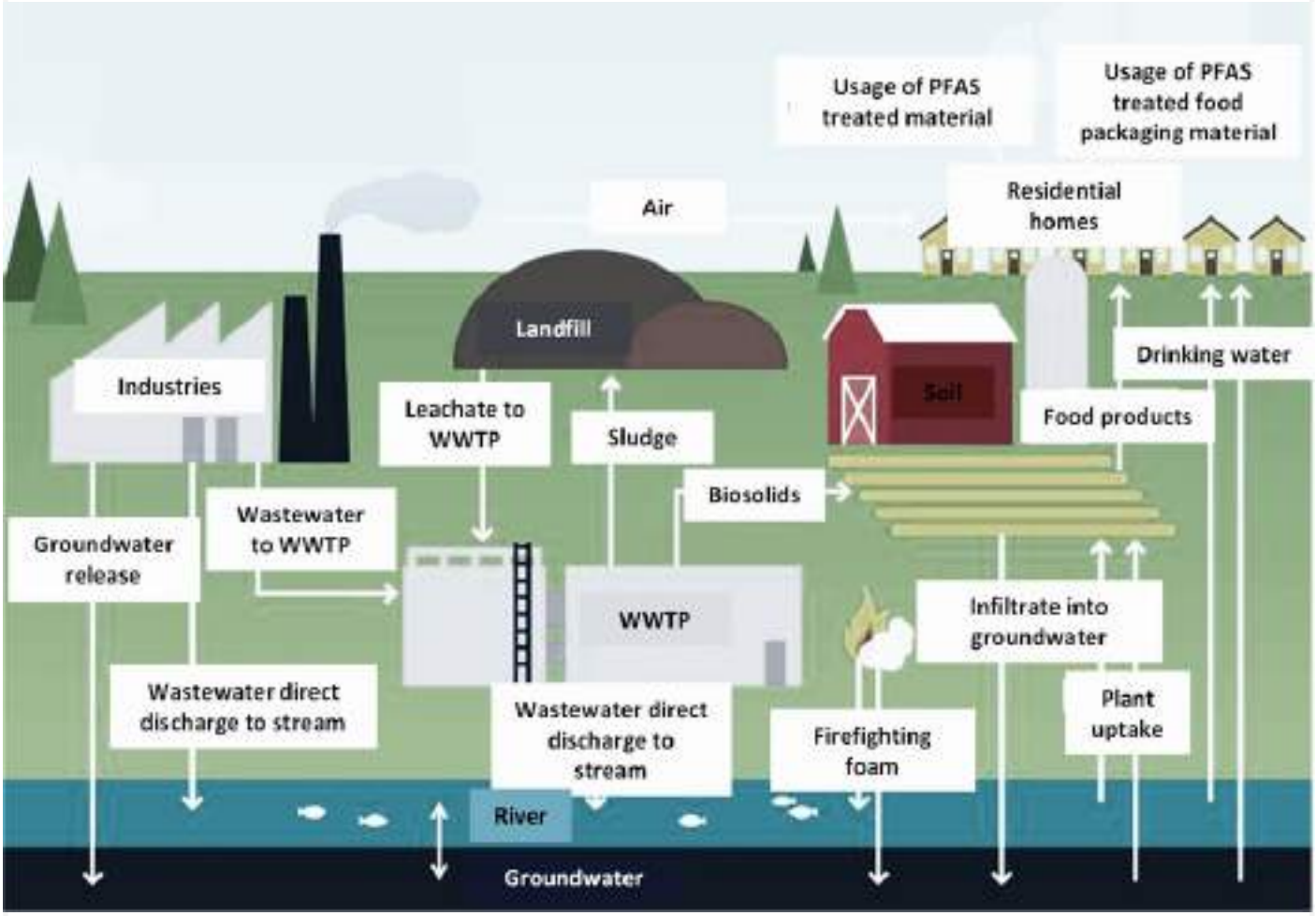


Figure 6. Mechanistic pathway of PFAS source cycle (Picture reused with permission under common creative license from Michigan Dept. of Environment, Great Lakes, and Energy (EGLE, 2019)).

media differ for each PFAS. Three dominant mechanisms: partitioning, transport, and transformation, are involved in the fate of PFAS (ITRC, 2020) (Figure 7). Hydrophobic-lipophobic effects, electrostatic interactions, and interfacial interactions are the evident mechanisms for the PFAS partition to soil, water, and soil/water or water/air interfaces (McKenzie et al., 2016; Brusseau, 2018). For example, the intensity of hydrophobicity of PFAS confirms their partition in the soil organic matter and their persistence in the contact medium. Likewise, the partitioning tendency of PFAS decides their distribution in the water/air or soil/water interfaces (McKenzie et al., 2016; Brusseau, 2018). The vapor pressure of the most commonly used PFAS (e.g., PFOA and PFOS) is low, and they possess high water solubility, so their partition to air is limited (EPA, 2000). Most PFAS are used in their anionic forms. So they are partitioned into the atmosphere as particulates (Ahrens et al., 2011a).

The main transport pathway of PFAS is physical transport because of their resistance to biotic and abiotic degradation (ITRC, 2020). Advection, dispersion, and diffusion are the fundamental and more evident transport pathways of the PFAS into the water sources, specifically the pollution related to surface water and GW. Theoretically, diffusion causes the atmospheric distribution of PFAS via air (Lohmann et al., 2013). Field studies near the fire training area showed penetration of PFAS up to the depth of 12 cm, which indicates the role of diffusion in the fixation of PFAS in the soil (Baduel et al., 2015). Deposition of PFAS from the air is solely related to the industrial release (e.g., stack emissions) because the low volatility of PFAS limits the long-range transport through the air (Ahrens and Bundschuh, 2014). Wet and dry deposition help to transfer the PFAS from the atmosphere to the soil (Rankin et al., 2016). Photooxidation also leads to the accumulation of PFAS intermediates in the soil and surface water through atmospheric deposition.

Leaching drives the transport of PFAS from one compartment to another. Leaching landfill waste containing PFAS products results in surface water, GW, and soil contamination (Benskin et al., 2012b).

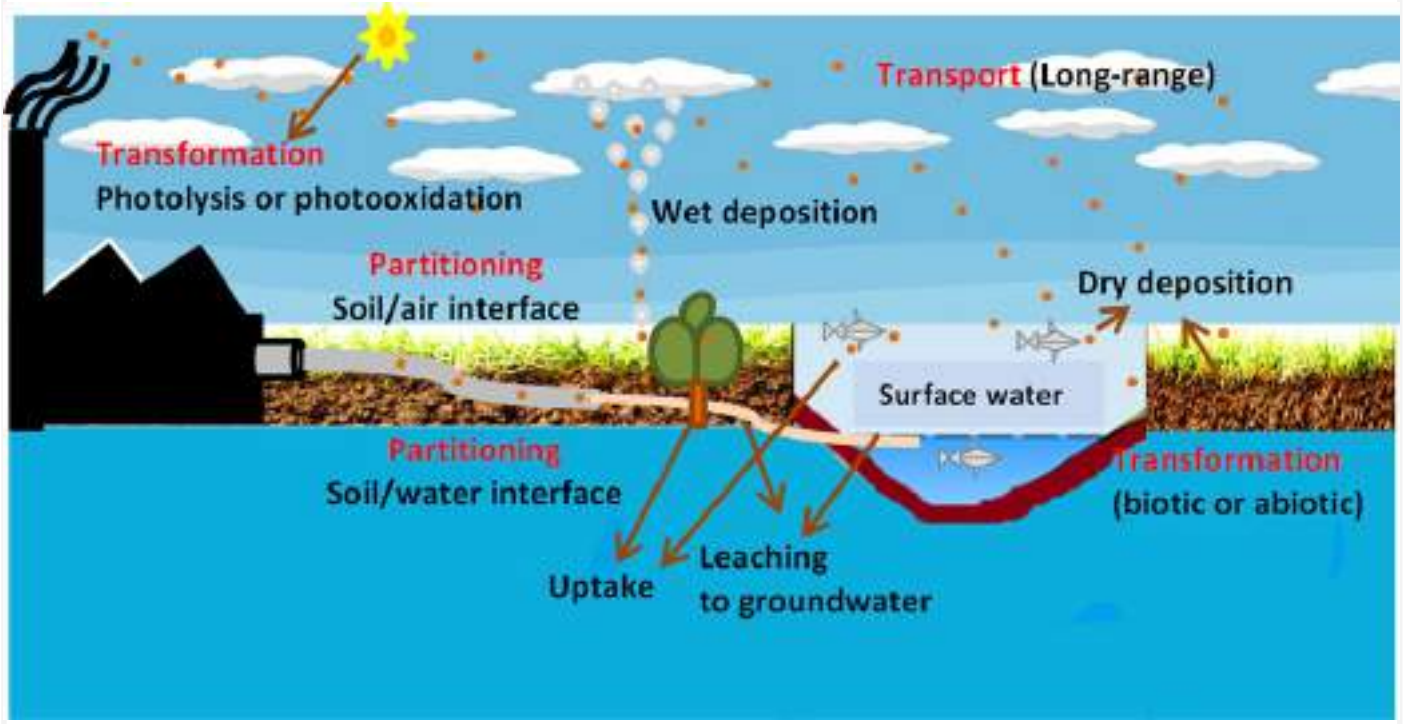
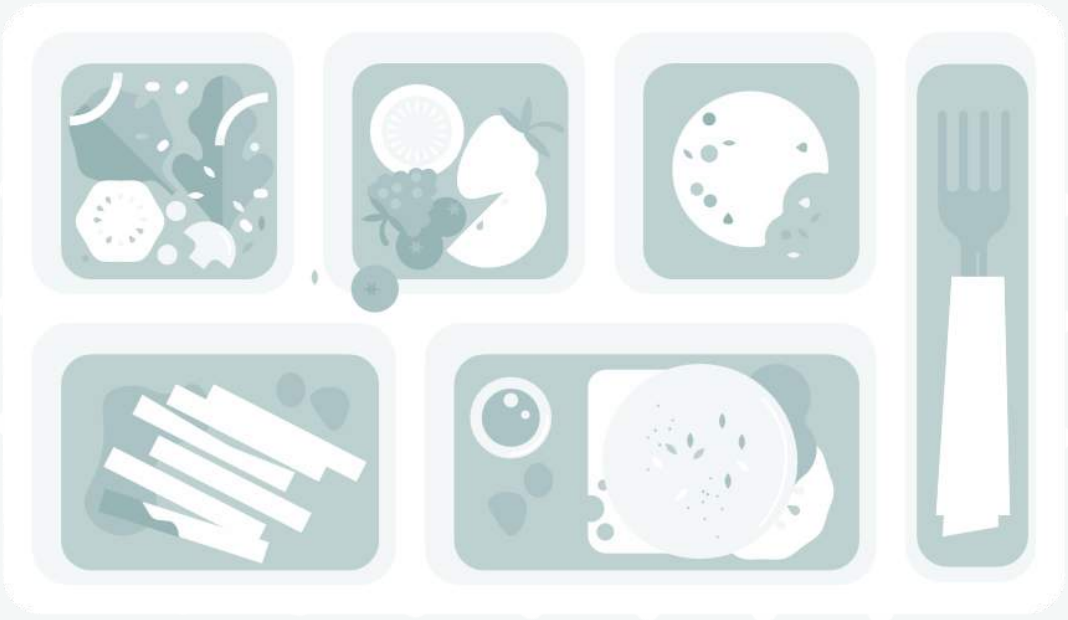


Figure 7. Environmental fate and transport of PFAS (IIT Madras)

Leaching is proved to reduce the exposure level of PFAS via dilution. However, it facilitates the long-term persistence of PFAS (Baduel et al., 2015). Long-chain PFAS are converted into PFAAs by biotic (e.g., aerobic biotransformation) and abiotic (e.g., hydrolysis and photolysis) reactions (Yarwood et al., 2007; Martin et al., 2010). Abiotic transformation of PFAS into their subgroups requires a longer time than biotic transformation. The degree of biotic transformation is site-specific and concentration-dependent (ITRC, 2020). Ultimately, PFAS are bioaccumulated in vegetables, crops, and animals. Consumption of any of these products results in the bioaccumulation of PFAS in humans. Using PFAS-containing products and working in industries manufacturing PFAS or products containing PFAS also result in PFAS accumulation in humans.

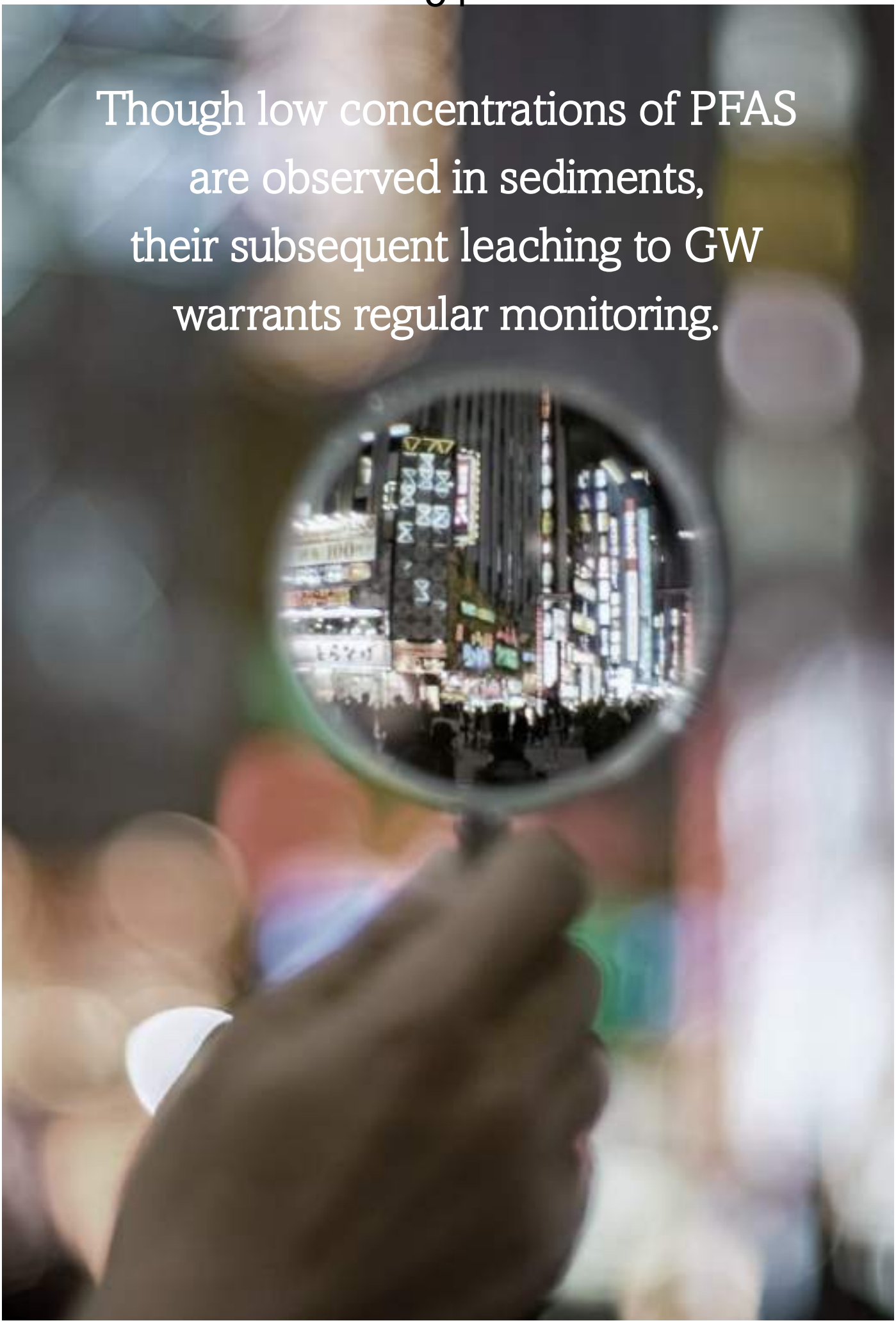


6. OCCURRENCE OF PFAS IN DIFFERENT ENVIRONMENTAL MATRICES

6.1. Worldwide distribution

The worldwide occurrence of PFAS during the period 2009 to 2021 is presented in this section. This period was chosen because the first report on the occurrence of PFAS in India was published only in 2009 (Mak et al., 2009). Worldwide reported concentrations of PFAS over the decade to lie in the range of pg L^{-1} to mg L^{-1}. The concentrations of PFAS-related compounds detected in different environmental matrices of countries except India are listed in Table 4. Detected concentrations of PFOA and PFOS were found to be low in recent years (after 2015), indicating the successful implementation of the policy on the phase-out of long-chain PFAS. However, many rivers are still contaminated with high amounts of PFOA and PFOS (Table 4). For instance, the detected concentration of PFOA in the Bohai Coastal region, China, was 4,534 ng L^{-1} (Wang et al., 2014a). Concentration of PFOS in the Llobregat River and its tributaries in Spain was 27,110 ng L^{-1} (Campo et al., 2015). These concentrations are relatively higher compared to the concentrations of other short-chain PFAS. High concentrations of short-chain PFAS have been detected in China: PFHpA (146 ng L^{-1}) (Sun et al., 2011), PFBS (1,256 ng L^{-1}) (Lin et al., 2010), and PFHxS (2,516 ng L^{-1}) (Lin et al., 2010). These concentrations are consistent with the production of short-chain PFAS in China (Liu et al., 2019).

Among the PFAS, higher concentrations of PFBS and PFHxS were detected compared to concentrations of PFOS. PFBS and PFHxS are the transformation products of Fluorotelomer olefins (FTOs) (a precursor in PFOS synthesis). Thus, transformation pathways might be leading to the increase in the concentrations of the PFOS and PFHxS. The reported concentrations of PFHpA (146 ng L^{-1}) and PFNA (174 ng L^{-1}) were lower than the concentrations of other short-chain PFAS (Sun et al., 2011; Castiglioni et al., 2015). Temporal and spatial variations of concentrations of PFAS in surface water are influenced by the type of surface water. In China, the Liao River had a PFAS concentration of 147 ng L^{-1}, whereas Taihu Lake recorded 461 ng L^{-1} (Yang et al., 2011). The reason for the low concentration of PFAS in the river was dilution. The partial degradation of PFAS into their short-chain or other compounds is possible in the river system because of the flow, while this is limited in the Lake. Similar observations have been recorded in other countries (Hong et al., 2013). Developed and highly industrialized countries like the US, U.K, Japan, South Korea, and China registered higher concentrations of PFAS than countries like Vietnam and Australia. Section 6.3 provides details on the presence of PFAS in India.


Though low concentrations of PFAS
are observed in sediments,
their subsequent leaching to GW
warrants regular monitoring.

The GW is an important sink for PFAS. Detected concentrations of PFAS in GW are relatively higher than in surface water due to the direct influence of upstream sources (Table 4). More noticeably, the detected concentrations of PFOS in the GW are relatively higher than in surface water due to their diffusion and partition phenomenon. For example, a low concentration of PFOS was detected in the surface water, whereas GW sampled at the same site showed 50 ng L^{-1} (Boiteux et al., 2012). The occurrence of high concentrations of short-chain PFAS is associated with their hydrophilic nature, as they can leach from the soil and get stored in GW.

WW is the dominant source of PFAS pollution in the environment. Domestic WW (e.g., Food packaging materials, dust, and household equipment), industries using PFAS containing products (e.g., chrome plating), and industries manufacturing PFAS rich goods (e.g., waterproofing agents) are identified as the major contributors. The approximate concentrations in these sources were in the ranges of <100, 100-1,000, and >1,000 ng L^{-1}, respectively (Vo et al., 2020). In Europe, the detected concentrations of PFHxA, PFOA, PFNA, PFHxA, and PFOS were 23,800, 2,962, 15,900, 2,735, 922, and 2,101 ng L^{-1}, respectively (Loos et al., 2013). Industrial WWTPs (Hsinchu Science Park) in Taiwan also showed high concentrations of PFAS (Lin et al., 2010). PFOA and PFOS were found to be the dominant pollutants. Unlike in surface and GW, the difference in short-chain and long-chain PFAS concentrations is not evident for WW. This may be due to the influent quality and treatment efficiency of the WWTP. In some cases, partial destruction of the long-chain PFAS results in increased concentrations of short-chain PFAS in the effluent than in the influent (Sun et al., 2011). The reason is that secondary treatment is known to remove >50% of long-chain PFAS and is inefficient for removing shorter PFAS. Many researchers indicated that discharge of partially treated WW and improper segregation of industrial, domestic, and municipal WW is the basis for subsequent environmental pollution.

Concentrations of PFAS in the sediment samples from different countries ranged from 0.01 to 3.7 ng L^{-1}. More noticeably, concentrations of PFCAs were higher than PFOS due to their physicochemical properties. Maximum concentrations of PFOA (3.7 ng L^{-1}), PFHxA (3.2 ng L^{-1}), and PFOS (7.3 ng L^{-1}) were observed in sediment samples collected from Haihe River, China (Li et al., 2011). Concentrations reported in France and Australia are one and ten orders of magnitude lesser than concentrations reported in China. Though low concentrations of PFAS are observed in sediments, their subsequent leaching to GW warrants regular monitoring.

Table 4. Worldwide occurrences of PFAS in different environmental compartments reported between 2010 and 2021

S. No.	Sample type	Sample Period	Location	Concentration of PFAS (ng L^{-1})							References
				PFHxA	PFHpA	PFOA	PFNA	PFBS	PFHxS	PFOS	
Surface water [A] China											
1	River	2009	Hun, China	1.26-11.3	0.56-4.0	1.83-10.7	ND-1.55	-	ND-1.50	0.40-3.32	(Sun et al., 2011)
			Lake, China	1.26-25	1.48-3.92	4.41-15.6	0.78-1.41	-	ND-1.50	0.40-1.09	
			Urban and drainage canal	1.26-36.7	1.28-146	9.23-24.5	ND-5.17	-	ND-76.1	0.66-16.3	
2	Reservoir	2006-2008	Sub-regions of Guanting Reservoir, China	ND	ND-35.0	0.43-82	ND-4.9	ND	ND-5.8	ND-31.0	(Wang et al., 2012)
3	Surface water Lake		Liao River, China Taihu Lake, China	- -	ND-18.4 ND-23.3	ND–27.9 10.6–36.7	-	-	1.4-94.5 ND-6.5	ND–6.6 3.6–394	(Yang et al., 2011)
4	Surface water	2010	Wuhan, China	-	-	ND-256	73.6	-	-	ND-88.9	(Wang et al., 2013a)
5	Pearl River		China	0.17-2.58	0.23-1.45	0.71–8.7	0.03-1.4	0.42-23	0.03-0.81	0.52–11	(Zhang et al., 2013)
6	Haihe River		China	-	2.2-42	4.4-42	-	-	-	2.0-7.6	(Li et al., 2011)
	Dagu drainage canal			-	14-124	14-65	-	-	-	1.2-73	
7	River Basin		Huai river basin Taihu lake	-	-	18 56	-	-	-	4.7 15	(Yu et al., 2013)
8	Coastal region		Bohai, China	-	57.5	0.96–4,534	-	-	-	-	(Wang et al., 2014a)
9	Lake		Taihu, China	-	-	136	-	-	-	29.2	(Pan et al., 2014)

S. No.	Sample type	Sample Period	Location	Concentration of PFAS (ng L^{-1})							References
				PFHxA	PFHpA	PFOA	PFNA	PFBS	PFHxS	PFOS	
Surface water											
[B] Taiwan, Vietnam											
11	River		Nanmem-Keya confluence, Taiwan	47.7-67.8	-	34.9-717	-	1,256	2,516	61.9-4,330	(Lin et al., 2010)
12	River	2013	Four cities (Hanoi, Ho Chi Minh) of Vietnam	ND-3.1	ND-2	ND-18	ND-0.93	-	ND-6.6	ND-5.3	(Duong et al., 2015)
13	River		Red river, Vietnam	-	-	ND-0.52	ND-0.53	-	0.21	-	
[C] Japan											
14	Bay		Tokyo, Japan			0.78-17				2.7-63	(Sakurai et al., 2010)
15	River	2010	41 rivers in Hyogo perfecture, Japan	ND-16,000	ND-27	ND-360	ND-39	ND-49	ND-8.4	ND-97	(Takemine et al., 2014)
16	Tap water		Tokyo, Japan	-	1.0-4.6	4.7-12	3.6-8.5	-	0.68-2.7	1.7-11	(Kuroda et al., 2014)
17	N. basin of Lake Biwa	2009	Japan	-	-	7.0-10	-	-	-	0.8-16	(Tsuda et al., 2010)
	S. basin of Lake Biwa					8.3-12				0.9-1.7	
	Akanoi Bay					9.1-17				1.4-2.8	
[D] South Korea											
18	Surface water	2008	Estuarine and coastal of Korea	ND-47	ND-47.2	2.95–68.6	1.38–14.3	ND-39.8	0.38-41.8	4.11–450	(Naile et al., 2010)
19	Artificial lake		Yongsan river	1.5-3.0	7.0-110	4.1-7.1	1.4-1.9	3.4-5.1	2.0-5.0	1.7-7.1	(Hong et al., 2013)
	Inland creek			1.3-6.9	1.6-8.3	2.6-10	0.62-3.2	2.1-8.6	1.0-16	0.38-68	
	Estuarine area		Nakdong river	0.81-2.7	2.4-9.3	1.3-3.6	0.45-2.1	1.0-4.6	0.83-4.3	0.57-5.8	
	Artificial lake			2.4-3.5	3.3-5.7	44877	4.6-5.2	1.3-2.1	3.7-7.1	6.5-12	
	Inland creek			3.1-17	4.7-21	8.9-28	2.5-12	3.7-12	0.83-17	5.3-66	
	Estuarine area			4.4-10	4.5-34	44888	3.6-7.7	2.1-15	1.5-14	2.5-26	
20	Rivers	2014-2015	South Korea	<0.20-95	<0.117-21	<0.12-2	<0.14-64	<0.11-2.32	<0.096-50	<0.12-33.2	(Lam et al., 2016)

S. No.	Sample type	Sample Period	Location	Concentration of PFAS (ng L^{-1})							References
				PFHxA	PFHpA	PFOA	PFNA	PFBS	PFHxS	PFOS	
Surface water											
[E] Singapore and Malaysia											
20	River	2009	Langat, Malaysia	-	-	0.2-5.94	-	-	-	0.71-43.5	(Zainuddin et al., 2012)
21	Surface water	2006-2007	Singapore	-	-	5.7-91.5	-	-	-	2.2-87.3	(Hu et al., 2011)
	Coastal water					2.4-17.8				1.9-8.9	
[F] Australia, Spain, Germany, France and Italy											
22	Harbour and surroundings		Sydney, Australia	2.8-3.2	1.4-2.0	4.2-6.4	0.60-2	1.2-1.5	2.7-4.3	7.5-21	(Thompson et al., 2011)
	Brisbane	2011	Australia	0.06-6.2	ND-3.7	0.08-11	ND-0.54	0.62-2.6	ND-17	ND-34	(Gallen et al., 2014)
23	Coastal water		Catalonia, Spain		<0.06-1.62	<0.03-3.93	<0.06-0.77	<0.07-0.24	<0.03-0.47	<0.03-3.93	(Sánchez-Avila et al., 2010)
	Surface water					0.79-9.63	<0.06-1.62	<0.07-0.74	<0.03-0.64	<0.03-9.56	
24	Llobregat and its Tributaries	2010	Spain	0.63-25.2	0.63-30.9	0.07-146	0.77-52.4	0.41-4.10	14.2-33.2	0.01-27,110	(Campo et al., 2015)
25	Surface water	2010-2012	Spain	0.23-31	0.23-27	0.16-68	0.03-52		0.06-37	0.04-2,709	(Llorca et al., 2012)
			Germany	0.23-13	0.23-24	0.16-6.5	0.03		0.06-5.6	0.04-4.6	
26	Tap water		Spain	0.23-11	0.23-16	0.16-35	0.03-22	0.11-36	0.06-28	0.04-258	
			Germany	0.23-1.8	0.23-24	0.16-1.9	0.03	0.11	0.06	0.04-0.4	
27	River water, Danube	2013	EU	ND-8.5	ND-18.8	ND-36.5	ND-3.3	ND-.37	-	ND-26.2	(Loos et al., 2013)
28	Welland	2008-2010	Canada	4.3-177	1.2-70.6	5.3-62.4	0.5-17.3	2.7-28.6	ND	6.1-392	(De Solla et al., 2012)
29	Surface water		France	139	<4	7	52	5	8	62	(Boiteux et al., 2012)
30	River		Orge, France	11.3	4.5	9.4	1.3	4.4	13.6	17.4	(Labadie and Chevreuil, 2011)
31	Surface water	2009	France		21	11	5	<4	<4	Traces	(Boiteux et al., 2012)
32	Po river tributaries	2011-2012	Italy	ND-62	ND-93	3-303	ND-174	ND-66	ND-5	ND-43	(Castiglioni et al., 2015)

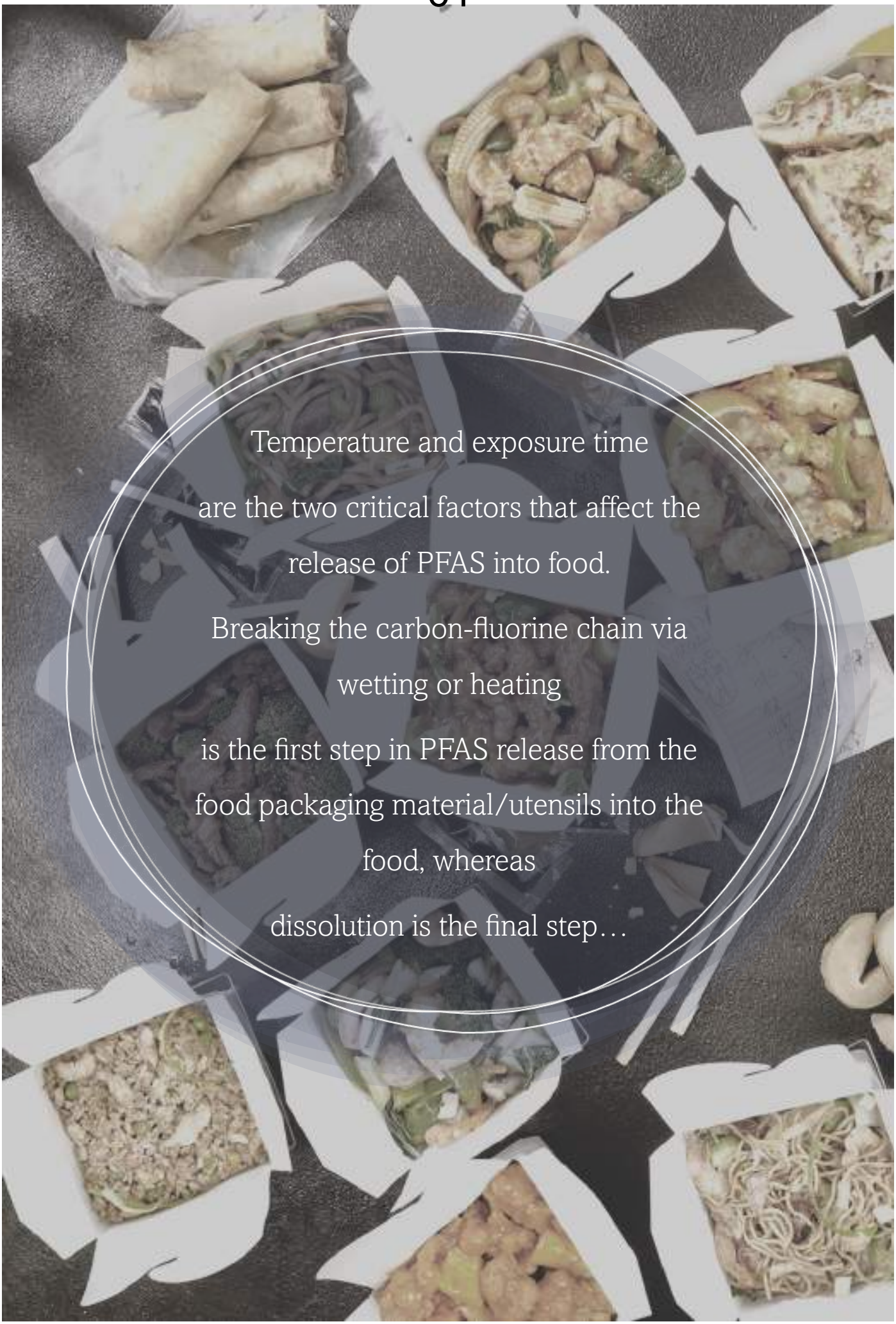
S. No.	Sample type	Sample Period	Location	Concentration of PFAS (ng L^{-1})							References
				PFHxA	PFHpA	PFOA	PFNA	PFBS	PFHxS	PFOS	
GW											
1	GW (Well water)	2013	Four cities (Hanoi, Ho Chi Minh) of Vi-etnam	-	ND-1.3	ND-4.5	ND-0.45	-	ND-6.0	ND-8.2	(Duong et al., 2015)
2	GW		France	28	39	16	14	9	32	50	(Boiteux et al., 2012)
Wastewater											
1	MWWTP Effluent		Taipei	155-181	<0.1	19.3-25.4	<0.1-0.3	2.6-4.8	6.3-35	163-265	(Lin et al., 2010)
2	IWWTP		Hsinchu Science Park, Taiwan	71.1	14.5	480	10.4	960	2227	5663	
3	WWTP (IN)	2009	Shenyang, Chi-na	15.7-33.4	3.14-17.9	26.2-71.1	ND-4.53	NA	8.06-22.3	2.88-12.8	(Sun et al., 2011)
	WWTP (EF)			10.7-11.3	1.60-24.2	18.4-41.1	ND		ND-4.32	1.69-3.85	
4	WWTP		Europe	1-23,800	387,890	1-15,900	0.5-2,735	-	1-922	0.5-2101	(Loos et al., 2013)
5	WWTP		Germany	-	2.1-63	0.04-1.8	0.09	-	0.13	0.22	(Llorca et al., 2012)
			Spain	-	-	0.04-17	0.09-18	-	0.13-15	0.22-501	
6			Singapore	-	-	7.9-1,060	-	-	-	5.8-35.3	(Hu et al., 2011)
7			Catalonia, Spain	-	-	3.47-61.9	<0.06-14.1	<0.07-2.03	<0.03-25.3	<0.03-72.1	(Sánchez-Avila et al., 2010)
Sediment											
1	Sediment			-	-	2,000 ng/kg	-	-	-	89,700-1,59,400	(Lin et al., 2010)
2	Sediment		Liao River, Chi-na		<0.01-0.13	0.02-0.18	0.01-0.07	ND-0.13	<0.02-0.10	0.04-0.48	(Yang et al., 2011)
			Taihu Lake, China		ND-0.33	<0.02-0.52	<0.01-0.51	ND-0.27	ND-0.03	0.06-0.31	
3	Haihe River		China	0.6-3.2		0.9-3.7				1.8-7.3	(Li et al., 2011)
	Dagu drainage canal		China	0.13-3.5		0.27-1.7				0.09-2.3	
4	Sediment		Orge, France	0.06	0.03	<0.07	0.05	<0.05	0.1	4.3	(Labadie and Chevreuil, 2011)
5	Sediment		Sydney, Austral-ia	ND	<0.02	0-0.16	<0.10-0.11	ND	<0.10-0.10	<0.10-0.20	(Thompson et al., 2011)

6.2. Release of PFAS from food contact materials

Food packaging became an essential element in maintaining the hygienic properties of the food. Temperature and exposure time are the two critical factors that affect the release of PFAS into food. Breaking the carbon-fluorine chain via wetting or heating is the first step in PFAS release from the food packaging material/utensils into the food, whereas dissolution is the final step. Investigation of PFAS release from the pans, bakeware, electric rice cookers, grills, and muffin papers indicated the release of short-chain PFAS (e.g., PFNA and PFODA) in the range of 2.12-3.05 μg L^{-1} (Choi et al., 2018). Another study on paperbacks and packed milk also showed high concentrations of PFAS (Elizalde et al., 2018). The release of PFAS from the non-stick pans while cooking acidic foods (e.g., tomato sauce) was slightly higher (PFOA: 18.3 μg kg^{-1} and PFOS: 16.6 μg kg^{-1}) than the release while cooking white beans (PFOA: 18.1 μg kg^{-1} and PFOS: 16.0 μg kg^{-1}) (Mohamed et al., 2019). Similarly salt in the food also increases (with salt PFOA: 16.0 μg kg^{-1} and without salt: 9.28 μg kg^{-1}) the release of PFAS from utensil to food (Mohamed et al., 2019). The temperature and cooking time affect the PFAS migration to food. For instance, muffin paper heated to >150ºC released 25% more PFAS than the food material cooked under 100ºC. The repeated use of PFAS-containing utensils also increases the PFAS migration to food material; e.g., the first use migrated 18.3 and 16.6 μg kg^{-1} of PFOS and PFOA, and 10 repeated usages increased the released concentrations to 60.3 and 54.2 μg kg^{-1}, respectively (Mohamed et al., 2019). Thus, food packaging materials increase the concentration levels of PFAS in consumers. On the other hand, disposal of the PFAS-coated paper into the soil environment releases PFAS into the soil. For instance, natural degradation or composting of the PFAS-containing paper results in the entry of PFAS into the biosolids, and subsequently, it is transferred to the soil after application. In this regard, the composite prepared from the waste paper biomass showed a PFAS concentration in the range of 3.4 to 35 μg kg^{-1} (Brändli et al., 2007). The application of PFAS-loaded composite to the cropland results in the entry of PFAS into the crops. The consumption of crops by livestock results in PFAS entry into the milk and meat; finally, it ends up in humans (Kowalczyk et al., 2012). The persistence of PFAS in soil depends on the chain length. For example, PFAS with carbon number greater than 8 are lipophilic and possess a longer bioaccumulation factor. PFAS with a carbon number less than 8 (short-chain PFAS) are hydrophilic and possess more mobility in water (Sun et al., 2016). So, the plants easily take up short-chain PFAS, which acts as a source of PFAS contamination in livestock and humans. Therefore, controlling the PFAS concentrations from the food contact material is a simple and solitary way to reduce PFAS pollution in the consumers. Accordingly, Denmark, and the cities of Maine, Washington, and San Francisco in US have introduced a ban on the entire group of PFAS in food contact materials (McRobert, 2020). The State of California is regularly biomonitoring the concentrations of PFAS in different food materials. Similar regulations are necessary to safeguard people from PFAS pollution in India also.

6.3. Indian scenario

The presence of PFAS in India has been investigated after the Stockholm Convention in 2006. PFAS was detected in tap water (Mak et al., 2009), surface water (Yeung et al., 2009), rain water (Kwok et al., 2010), sediment (Corsolini et al., 2012), GW (Lapworth et al., 2018), and biological samples (e.g., breast milk (Tao et al., 2008), blood samples (Ganesan and Vasudevan, 2015), pig meat (Watanabe et al., 2010), fish (Murakami et al., 2011), dolphin/shrimp (Yeung et al., 2009), and human hair (Ruan et al., 2019). The detected concentrations were in the range of 0.005 to 10 ng L^{-1}. In terms of region, the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southern part of India (e.g., Chennai) has shown higher concentrations of PFOS (8.4 ng L^{-1}) and PFOA (2.0 ng L^{-1}) (Mak et al., 2009) compared to the northern part (e.g., Varanasi (PFOS: 1.73 and PFOS: 1.2 ng L^{-1}) (Sharma et al., 2016). Similarly, biological samples (e.g., breast milk and human hair) collected from Chennai showed high concentrations (Human hair: 0.25 (PFOS)-2.13 (PFOA) ng g^{-1} and breast milk: 1.11-335 ng L^{-1}) (Tao et al., 2008; Ruan et al., 2019). The presence of well-established industrial hubs and income-associated growth in the lifestyle accounts for higher usage of PFAS-related products in the Southern part of India are the main reason for the hike in PFAS concentration. However, the frequency of detection of short-chain PFAS (e.g., PFHxA, PFHpA, PFNA, PFBS, and PFHxS) was higher for the northern region (especially near the River Ganga) in comparison with the southern part. So, other than industries, WW from municipal/domestic sources also contributed to PFAS pollution (Sharma et al., 2016). The detected concentrations of PFAS in India are relatively lower than those in other countries.

6.3.1. Occurrence in different environmental compartments

In comparison with the world scenario, fewer reports/studies about the detection of PFAS in various environmental media are made. This may be due to low awareness of the persistent nature of PFAS. Low concentrations of PFAS in different environmental media in India could also be a reason. For example, short-chain PFAS like PFHxA, PFBS, and PFHxS are predominantly detected in surface water samples of India. At the same time, higher concentrations of long-chain PFAS (PFOA and PFOS) are reported from other countries. Table 5 and Figure 8 present the concentration levels of PFAS detected in different environmental matrices in India.

The first study (referred to in Table 5) related to the occurrence of PFAS in tap water/drinking water samples was conducted from 2006 to 2008 (Mak et al., 2009). The detected concentrations of PFOA and PFOS were in the range of < 0.17-0.40 ng L^{-1}. In comparison with the worldwide occurrence of PFAS, the measured concentrations in Indian cities were lower. The estimated concentrations of PFAS in the blood samples collected from India were also lower (e.g., PFAS: 2,530-3,000 ng L^{-1}) compared to those reported from the US (e.g., PFAS: 1,300-9,17,000 ng L^{-1}) and Japan (e.g., PFAS: 2,600-40,300 ng L^{-1}) (Kannan et al., 2004). However, a high concentration (81 ng L^{-1}) of short-chain PFAS (e.g., PFHxS) was detected in Patna. The frequent occurrence of PFHxS indicates the increased use of PFAS substitutes in India. Compared to developed countries (e.g., US, Japan, EU), the limited usage of carpets and food wrappers reduces the occurrence of long-chain PFAS.

The presence of PFAS in tap water leads to testing the surface water to identify the exact pollution source for PFAS contamination (Yeung et al., 2009). The measured concentrations of PFOS (<0.04-3.91) and PFOA (0.04-49.7 ng L^{-1}) in surface and untreated sewage water in India are lesser than those reported in other countries. High concentrations of PFOS (3.91 ng L^{-1}) were observed in the Cooum river samples. Also, untreated sewage water samples in the southern part of India (Goa, Coimbatore, Chennai) registered high PFOA (49.7 ng L^{-1}) concentrations (Mak et al., 2009). PFOS was detected in shrimp, fish, and dolphins, whereas PFOA was detected only in fish samples (75%). Apart from this, PFUnDA and PFDA were predominantly detected in biota in the concentration range of 0.06-1.0 ng g^{-1}. Untreated sewage water contributed to the maximum PFOS (1.81 ng L^{-1}) and PFHxA (2.29 ng L^{-1}) contamination in the River Ganga. The WW released from coal-fired power plants, food processing, and electroplating industries also contaminates the River Ganga. The least concentrations of PFAS in the southern part of

Table 5. Occurrences of PFAS in different environmental matrices in India

S. No	Source	Sampling period	Sample No.	Location	PFAS detected (ng L^{-1})							References
					PFHxA	PFHpA	PFOA	PFNA	PFBS	PFHxS	PFOS	
1	Tap/drinking water	2006-2008	9	Goa Coimbatore Chennai Patna	- - - -	- - - -	<0.83 <0.005 2.0 0.051	- - - -	- - - -	- - - 81	<0.083 <0.033 8.4 <0.04	(Mak et al., 2009)
2	River water	April-May 2008 (prior monsoon)	42	Ganga	<0.092-2.29	-	<0.04-0.20	<0.04-0.18	-	-	<0.04-1.81	(Yeung et al., 2009)
3	Precipitation	June 2006- Sep 2008	2	Patna, India	12%	-	13%	-	-	-	-	(Kwok et al., 2010)
4	Sediment samples	Dec-Jan 2011	13	Ganga (Hugli)	-	-	<0.50-14.1 ng g^{-1}	-	-	-	<0.50 ng g^{-1}	(Corsolini et al., 2012)
5	Ganga	Feb-Apr 2014	14	Varanasi	0.4-4.7	-	0.1-1.2	0.027	<0.04-10.2	<0.017-0.8	<0.01-1.73	(Sharma et al., 2016)
	Drinking water		14		0.8-4.9	0.5-3.5	0.018-0.8	<0.027.2	<0.04-4.9	<0.017-0.08	<0.01-1.1	
6	River	2014			-	-	0.4-0.5	-	-	-	BDL	(Sunantha and Vasudevan, 2016)
7	GW	Mar-16	3	Varanasi	-	-	-	5.4	-	-	0.4-3	(Lapworth et al., 2018)
	Ganga		3		-	-	-	-	-	-	2.6-25	
8	River water (Drinking water category)	2013-2014 and 2014-2015 Dec-Jan and Mar-July		Kaveri, Vellar, Tamiraparani	ND-3.16	ND-1.21	ND-2.07	ND-0.47	ND-2.45	ND-7.02	ND-1.10	(Selvaraj et al., 2021)

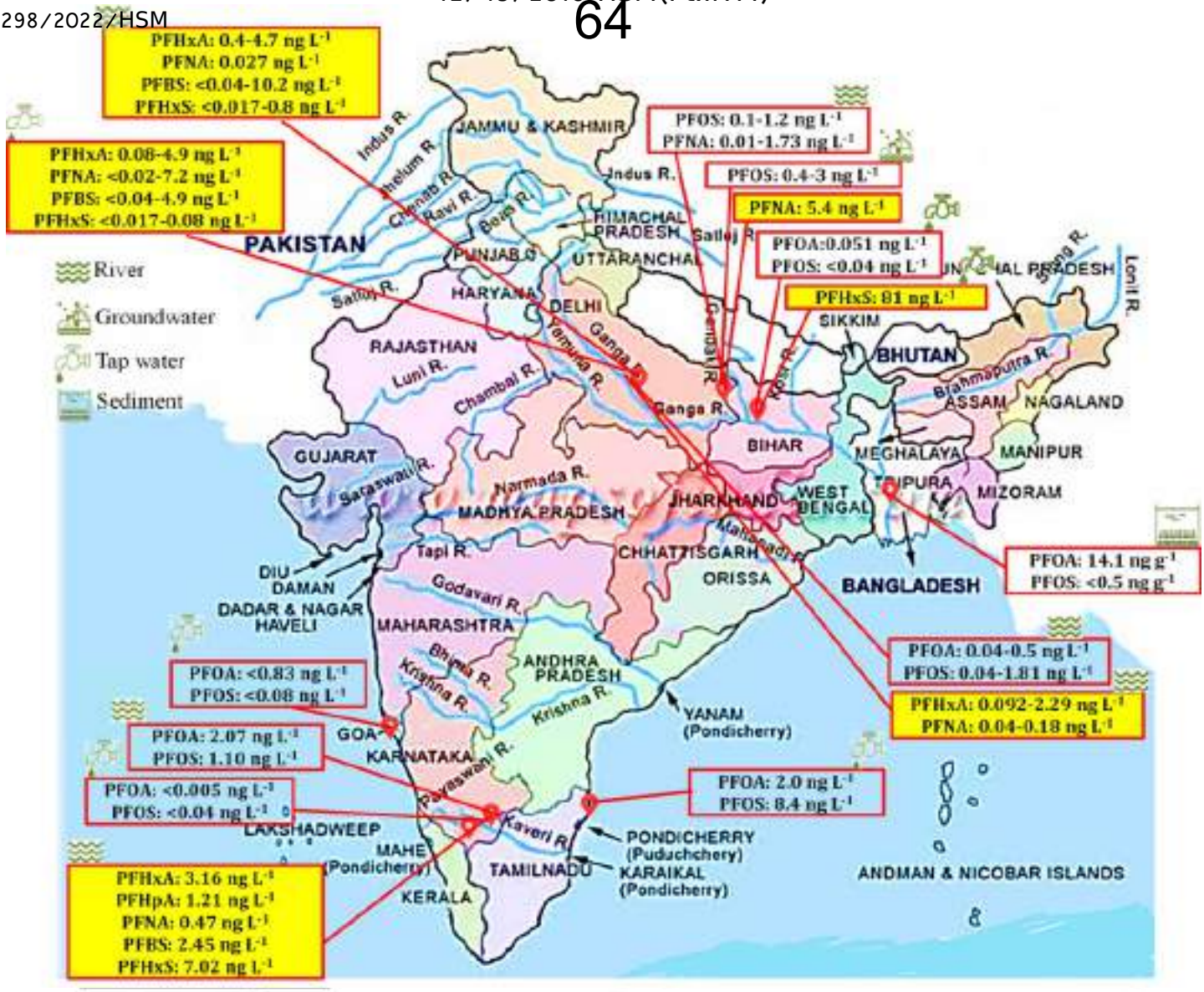


Figure 8. Occurrences of PFAS in India

(Map source: Compare InfoBase, Input value from Ref.: Mak et al. (2009), Yeung et al. (2009), Corsolini et al. (2012), Sharma et al. (2016), Sunantha and Vasudevan (2016), Lapworth et al. (2018), and Selvaraj et al. (2021)).

India were observed in Goa (<0.083-<1 ng L^{-1}), and the highest was detected in Chennai. The detected concentrations of PFOA, PFOS, and PFHxA in seawater and lake water samples are below the method detection limits.

The investigation by Corsolini et al. (2012) on the sediment samples of the Ganga River showed that PFOA (<0.5-14.1 ng g^{-1} dw) is the dominant and persistent compound in sediment samples than PFOS (below detection level (<0.05 ng g^{-1} dw). The high-level deposition of PFOA on the sediments increases their bioaccumulation. Meanwhile, it increases the bioavailability of benthic and tropic biota. The release of untreated WW and surface runoff from Hugli River to the lower stretch of the Ganga was identified as the crucial source of PFAS pollution in the Hugli estuary and Sundarban wetland. Early studies have indicated that Indian water systems predominantly contain short-chain PFAS and provided information on different pathways of PFAS distribution in India. Recently, Sharma et al. (2016) conducted a comprehensive study on the changes in the concentration pattern of PFAS in the River Ganga. They

found that short-chain PFAS (C_5-C_8) are detected more frequently than PFOA. PFBS, PFHxA, PFOA, and PFOS concentrations are in the ranges of <0.04-10.2, 0.4-4.7, 0.1-1.18, and <0.01-1.73 ng L^{-1}, respectively. The source of PFAS contamination was industrial and domestic WW discharge. Interestingly, the measured concentrations of PFHxA and PFBS in the Himalayan Reach (0.37-3.15 and 0.52-9.40 ng L^{-1}) are comparable with the concentrations measured in the middle (0.86-4.36 and <0.04-8.61 ng L^{-1}) and lower reaches (0.64-4.73 and <0.04-10.2 ng L^{-1}). It should be noted that the Himalayan Reach is a catchment area with lower anthropogenic emissions, whereas the middle and lower reaches have highly populated industrial hubs and estuaries. From this, it is clear that, atmospheric transport of PFAS and their precursors (e.g., FTOHs and Perfluorooctanesulfonamide (PFOSAs)) is the source of surface water contamination in remote areas like Himalayan Reach. Also, the detected short-chain concentrations of PFAS (e.g., PFHxA, PFHpA, and PFBS) were relatively higher than concentrations of PFOA and PFOS.

The mobility of short-chain PFAS is faster than long-chain PFAS; subsequently, it results in atmospheric distribution. The mean cumulative discharges of PFOA and PFOS in Ganga catchment areas were estimated as 240 and 210 g day^{-1}, respectively. The persistent nature and frequent detection in surface water indicate their possible intrusion into GW. The measured concentrations of PFHxA, PFHpA, PFNA, PFBS, PFHxS, and PFOS in GW were 0.8-4.9, 0.5-3.5, <0.027-0.2, <0.04-4.9, 0.017-0.08, 0.01-1.1 ng L^{-1}, respectively. A similar pattern of PFAS in GW and surface water indicates their origin from a common source (e.g., industrial discharge). Samples collected 100 meters deep from the ground near Ganga were detected with PFAS and pesticides. In these sites, PFOS levels were in the range of <0.1-33 ng L^{-1}.

Another series of measurements around Ganga (Varanasi) was conducted by Lapworth et al. (2018). Interestingly, the concentrations of PFOS have increased gradually within two years. The maximum concentration of PFOS in the GW of Varanasi in 2014 was 1.1 ng L^{-1} (Sharma et al., 2016), and it increased to 3 ng L^{-1} in 2016 (Lapworth et al., 2018). Similarly, the PFOS concentrations in Ganga increased from 1.73 to 25 ng L^{-1}. The downstream region of the study area showed the maximum PFOS distribution (e.g., a deep municipal source in Varanasi). This municipal source was also identified as a sink for PFNA (5.4 ng L^{-1}) and PFDA (5.7 ng L^{-1}). These results indicated the rapid transport pathway of the PFOS into deep (200 m) aquifers and less sorption onto sediments.

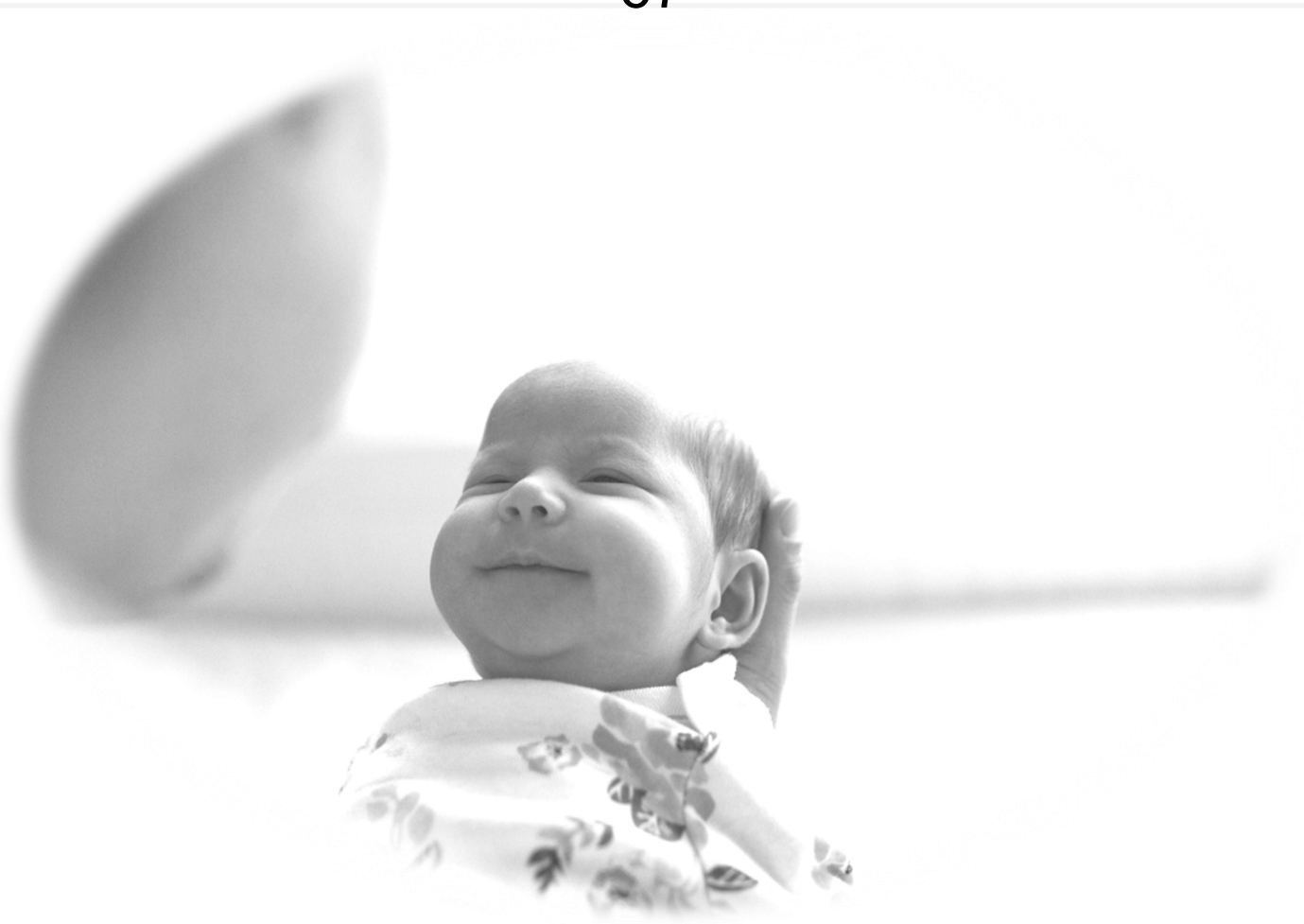
The southern part of India was found to have high concentrations of PFAS because of the presence of established industrial hubs. Sunantha and Vasudevan (2016) researched the occurrence pattern of PFAS in two important rivers of south India: Cauvery and Noyyal. Concentrations of the PFOA and PFOS in the Cauvery River sample were in the ranges of 4-93 and 3-29 ng L^{-1}, respectively. Noyyal was highly polluted, with the highest concentration of PFOA (93 ng L^{-1}) and PFOS (29 ng L^{-1}). A much detailed investigation of PFAS in three important Rivers, namely Kaveri, Vellar, and Tamiraparani, was conducted recently by Selvaraj et al. (2021). The maximum detected concentrations of PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, PFOS were 3.16, 1.21, 2.07, 0.47, 2.45, 7.02, 1.10 ng L^{-1}, respectively. The temporal variation analysis indicated that in 2014, PFOA was the predominantly detected compound (>40%), but in 2015 more than six PFAS (e.g., PFHpA, PFBS, PFHxS, and PFDS, PFOA, and PFOS) were detected with >40% of the detection frequency. The concentrations of PFAS increased. The major occurrence pattern of short-chain PFAS indicates different sources or usage patterns. Particularly, industrial emissions increased the bioaccumulation of PFAS. However, river flow dynamics and natural environmental degradation might

be the possible reason for this occurrence enhancement. The concentrations of PFAS in the summer season (e.g., Kavery: 1.92 ng L^{-1} in 2014) were higher than concentrations during the post-monsoon season (e.g., Kavery: 0.32 ng L^{-1} in 2014), which is due to the reduced flow in the summer. The statistical evaluation indicated a correlation between the PFCA and PFSA, suggesting a common pollution source. The industrial areas (e.g., Erode and Kulithalai) showed lesser concentrations of PFAS compared to non-industrial areas (e.g., catchment of Vellar River). Therefore, domestic and commercial WW is the primary contributor to polluting the selected Rivers. Although the flux, human health, and environmental risk assessments confirmed that the occurrences of PFAS are within the safer threshold, the biomagnification property of PFAS needs to be considered.

The long-range transport nature of PFAS is verified from their occurrence in the precipitation samples (Kwok et al., 2010). A total concentration of 1.40 ng L^{-1} of PFAS was found in the precipitation samples of Patna, India. PFCA and PFSA contributed 97 and 3%, respectively. Precipitation samples collected from an agricultural area of Patna indicated the predominant presence of short-chain PFAS (PFPeA (25%), PFPrA (15%), and PFHxA (12%)) and PFOA (13%). The existence of PFCAs in the precipitation samples was due to their higher water solubility (e.g., PFOA: 3.3×10^3 and PFOS: 680 mg L^{-1}) (Table 5). The relatively lesser PFOS (<5%) concentrations in precipitation samples are due to their non-preferential usage and physio-chemical properties (lower water and air partitioning).

PFAS was detected in the air samples from Chennai, Tamil Nādu. High concentrations of PFAS in $PM_{0.1}$ indicate their possible exposure to humans and others.

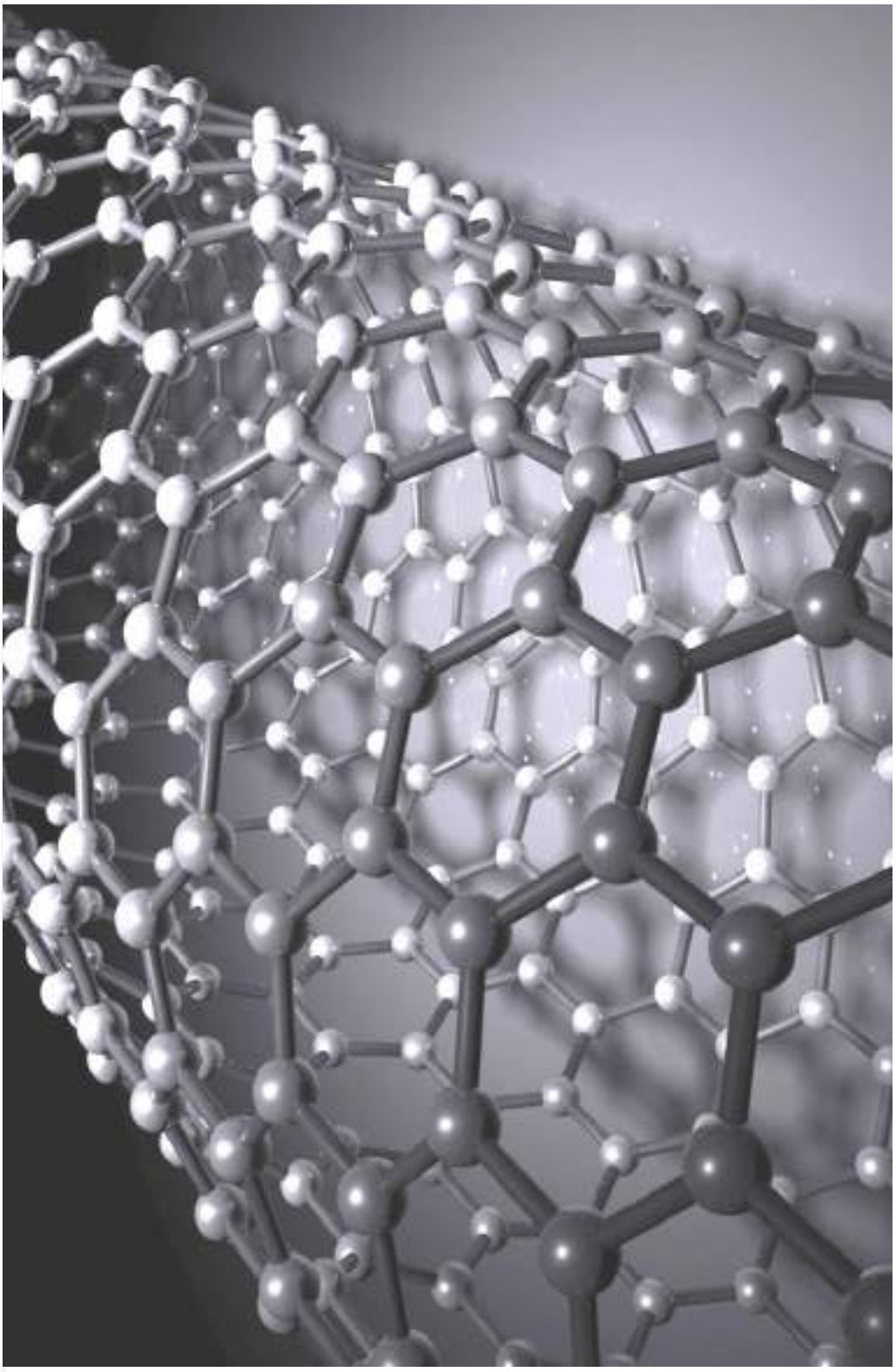
PFAS was detected in the air samples from Chennai, Tamil Nādu (Ge et al., 2017). High concentrations of PFAS in $PM_{0.1}$ indicate their possible exposure to humans and others. The ratio between $PM_{2.5}/PM_{10}$ was 0.38 and in all samples. The concentration of PFOA's was higher than the concentration of PFOS. Wang et al. (2014b) found the presence of PFAS in the snow cores of Mt. Muztagata and Mt. Zuoqiupu. It has been postulated that these mountains received PFAS contamination from wind, and according to the wind direction, the concentration varied. PFOA was identified as the dominant PFAS in Mt. Muztagata (61.4-349 pg L^{-1}) and Mt. Zuoqiupu (27.8-183 pg L^{-1}). The lesser concentrations of PFOS (40.9-243 pg L^{-1}) indicate their minimized exposure to the surrounding areas. Wang et al. (2019a) also observed that long-chain PFAS were common pollutants in the north and west regions, whereas short-chain PFAS were predominant in the south and east. Generally, the Indian monsoon results in atmospheric changes in PFAS distribution patterns in the atmosphere, accounting for 70% of PFAS transport to Western China.



6.3.2. Occurrences of PFAS in biota

Breast milk samples collected from Tamil Nādu and Kolkata showed PFAS in the range of 1.11–335 ngL^{-1} (Tao et al., 2008). In contrast with the reported occurrence pattern in different water sources, PFOA (<40-335 ng L^{-1}) and PFOS (<10-120 ng L^{-1}) were found to be predominant. This might be related to the increased use of PFAS coated products in food preparation (e.g., non-stick pans and food wrappers). Landfill causes the distribution of a wide variety of pollutants to the environment. It has been found that the landfill acted as a source to spread the PFAS in the pigs that strayed near the landfill sites (Watanabe et al., 2010). Female pigs had higher levels of PFAS (71 ng g^{-1} ww) than male pigs (9 ng g^{-1} ww). The measured concentrations of PFOS in fish (0.5-1 ng g^{-1} ww) from India were relatively lower than in Vietnam (Murakami et al., 2011). The observed increase in the detection frequency of PFOA and PFOS in surface water samples after 2010 is similar to the increased levels of these contaminants in human blood samples in different Indian cities (Ganesan and Vasudevan, 2015). Recently, high concentrations (<0.02-3.78 ng g^{-1}) of PFAS in human hair samples were also reported (Ruan et al., 2019). Among those, PFHxS (1.32 ng g^{-1}), PFOS (2.13 ng g^{-1}), PFDS (1.19 ng g^{-1}), and PFOA (0.25 ng g^{-1}) are the predominantly detected compounds. In terms of region, the Southern part of India contributed more than other parts of India.

7. TREATMENT METHODS FOR PFAS- AN OVERVIEW



The hazardous nature of the PFAS necessitates removal techniques to eliminate or reduce their concentrations in various environmental matrices. The stable physicochemical properties (e.g., nonvolatile) of PFAS reduce the efficiency of some conventional treatment technologies (e.g., air stripping, soil vapor extraction, and biodegradation). Adsorption using Granular Activated Carbon (GAC) and ion-exchange resin is the commonly employed full-scale treatment method for PFAS (Table 6). Also, thermal destruction (incineration), chemical oxidation/ reduction, and bioremediation-as treatment options are still in the pilot-scale stage. In this section, different treatment methods employed to treat PFAS have been discussed.

Table 6. Implemented treatment methods for the removal of PFAS from real field water samples

S. No	Influent type	Influent concentration (μg L^{-1})	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
Sorption-based techniques						
a) Activated carbon						
1	GW	PFOS: 0.6, PFHxS: 0.4, PFCA: <0.1 (DOC < 1 mg L^{-1})	1,240 EN GAC (Norit), FV: 12 L, pilot-scale, Flux: 4 or 7 m^3 h^{-1}, EBCT: 15 or 9 min, VF: 6 or 10 m h^{-1}	PFOS: 40,000 m^3 PFHxS: 20,000 m^3 PFCA: 4,000-12,000 m^3	Lesser efficiency for short-chain	(TZW, 2011; Janda et al., 2017)
2	GW	PFHxA: 0.6, PFPA: 0.6, PFOA, PFHpA, PFBA: 0.1-0.5, (EC: 600 μs cm^{-1}, sulphate: 37 mg L^{-1}, DOC: 0.7 mg L^{-1})	Hydraffin CC 8 ×30 GAC (coconut shell) (Donau carbon), VF: 140 L^{-1}, pilot-scale, Flux: 5 m^3 h^{-1}, EBCT: 15 min, VF: 8 m h^{-1}	PFOA: 35,000 m^3 PFHxA: 24,000 m^3 PFPeA: 20,000 m^3 PFBA: 8,000 m^3		(TZW, 2015)
3	FFWT	PFOS: 9,000, PFHxS: 1,000, PFHpS: 200, PFBS: 20, PFOA: 80, PFHxA: 70, PFHpA: 20, PFBA: 20, PFPeA: 20 (TOC: 80 mg L^{-1}, COD: 270 mg L^{-1}, Turbidity: 1.5 NTU)	Filtrasorb 600, VF: 400 L^{-1}, pilot-scale, two filter in series, Flux: 3 m^3 h^{-1}, EBCT: 20 min	For all PFAS >99.9% removal at 40 m^3	PFOS removal was not below guideline value	(USEPA, 2017b)
b) MatCare						
1	FFWT	PFOS: 75.8, H4PFOS: 508, 8:2 FTSA: 15.2, PFOA: 2.8, PFSOA: <0.2	4 filters in series, lab-scale	Removal: 90 to 99.9% PFOS: <0.02, H4PFOS: <0.1, 8:2 FTSA: <0.1, PFOA: <0.02, PFSOA: <0.02	Low throughput (40 m^3)	(CRCCare and R., 2015)
2	FFWT	PFOS: 35,500, PFOA: 6,859	Pilot-scale and lab-scale: fixed bed filter column (PFOS: 2077 and PFOA: 9001)	PFOS: 680 m^3, PFOA: 680 m^3	Sorbent exhaustion was not studied	(Kambala, 2016)
3	FFWT	PFOS: 6,000-14,000, PFOA: 1,000-2,000	Large-scale (Australian air force base)	PFOS: <2, PFOA: <2	-	(CRCCare and R., 2015)

S. No	Influent type	Influent concentration (μg L^{-1})	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
c) RemBind						
1	GW	PFOS: 2.9-3.4, PFOA: 0.58-1.3, PFBS: 0.016-0.022, PFHxA: 0.36-1.2, H4PFOS: 0.84-1.3 (TOC: 1.9 mg L^{-1})	Batch, **pilot-scale**, contact time: 1 h, Pre-treatment: sedimentation followed by overnight decantation	PFOS: 0.15-0.17, PFOA: 0.058-0.13, PFBS: ND, PFHxA: 0.0036-0.012, H4PFOS:0.026-0.039	-	(Chiang et al., 2017a)
2	FFWT	PFAS: 18,50,000	Lab-scale (filter column)	100% at 40 m^3	-	(Ziltek, 2017)
3	FFWT	PFOS: 9,000, PFHxS: 1,000, PFHpS: 200, PFBS: 20, PFOA: 80, PFHxA: 70, PFHpA: 20, PFBA: 20, PFPeA: 20 (TOC: 80 mg L^{-1}, COD: 270 mg L^{-1}, Turbidity: 1.5 NTU)	50:50 Rembind and silica sand, Flux: 0.9 BV h^{-1}, EBCT: 60 min	PFOS: 0.5, PFHxS: <0.01, PFHpS: <0.01, PFBS: <0.0004, PFOA: 0.001, PFHxA: <0.005, PFHpA: 0.001, PFBA: <0.0001, PFPeA: <0.0001	Low throughput and high-pressure loss	(USEPA, 2017b)
d) Ion exchange						
1	GW	PFOS: 0.027, PFOA: 0.43, PFBS: 0.17, PFBA: 0.21 (Sulphate: 44 mg L^{-1})	Purolite A532E, VF: 0.35 L, **lab-scale**, Flux: 6 m^3 h^{-1}, EBCT: 10 min, VF: 3 m h^{-1}	PFOS: 1,50,000 m^3, PFOA: 80,000 m^3, PFBS: 30,000 m^3, PFBA: 10,000 m^3	NH4Cl and NH4OH - 0.5%, KCl and KOH: 0.5%	(Zaggia et al., 2016)
e) Cyclodextrin (CD)-based adsorbents						
1	GW	PFOA: 50, pH 6.1-6.8	β-CD coated silica (1.5 g L^{-1}), lab-scale, contact time: 48 h	Removal: 95% PFOA: 2.5	-	(Bhattarai et al., 2014)
2	GW and FFWT	PFOA: 25 (pH 5.5) and PFAS: 200 (pH: 3)	β-CD coated magnetite (0.2-1.5 g L^{-1}), **lab-scale**, contact time: 24 h	Removal: 97% and 90%	-	(Badruddoza et al., 2017)
3	GW	PFOS: 50, PFOA: 50	β-CD coated magnetite (0.6 g L^{-1}), pH 5.5, **lab-scale**, contact time: 24 h	PFOS: 99% and PFOA: 96%	PFOS removal insensitive to pH change PFOA removal higher in acidic condition	(Badruddoza et al., 2017)
4	GW	Column exp: PFOS and PFOA: 20-30	β-CD coated magnetite (0.8 g L^{-1}), **lab-scale**, DOC: 4 mg L^{-1} in shaking exp, column exp: Flux: 0.3 BV h^{-1}, EBCT: 3 min	Column: PFOS: 6,300 m^3 and PFOA: 6,300 m^3, shaking: PFOS: 1.5 and PFOA: 5	Increase in DOC from 4 to 12 mg L^{-1} reduced removal by 49%	(Bhattarai, 2015)
		Shaking exp: PFOS and PFOA: 50	Shaking exp: contact time: 24 h			
5	GW	PFOA: 1	Porous CD polymer (0.01 g L^{-1}), **lab-scale**, contact time: 24 h	PFOA: <0.01	No effect of DOC was observed	(Xiao et al., 2017a)

S. No	Influent type	Influent concentration ($\mu g\ L^{-1}$)	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
5	GW	PFOA: 1	Porous CD polymer (0.01 g L^{-1}), lab-scale, contact time: 24 h	PFOA: <0.01	No effect of DOC was observed	(Xiao et al., 2017a)
6	GW	PFBA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, and PFOS: 1 (each)	Dexsorb+ (0.01 g L^{-1}), lab-scale, contact time: 30 min	PFBA: 0.01, PFHxA: 0.05, PFHpA: 0.03, PFOA: 0.03, PFNA: 0.05, PFBS: 0.03, PFHxS: 0.03, and PFOS: 0.08	-	(Dexsorb+)
7	FFWT	PFOA: 6.6, PFHpA: 1.61, PFOS: 519, PFBS: 11.4	Dexsorb+, lab-scale,	PFOA: ND, PFHpA: ND, PFOS: 0.022, PFHxS: 0.001, PFBS: ND	-	(Dexsorb+)
Coagulation/Flocculation						
1	FFWT	PFOS+PFHxS: 180 PFOA:1.22	InSite (1 g L^{-1}), **lab-scale**, contact time: 60 s (per reactor)	>99.9% (without polishing)		(CARE, 2017)
2	FFWT	PFOS: 40-380	Insite, **lab-scale**, contact time: 60 s (per reactor)	PFOS <0.02-0.11	US EPA threshold value (0.07 $\mu g\ L^{-1}$) was not observed PFOS >300 $\mu g\ L^{-1}$	(InTreat Water)
Foam fractionation and ozofractination						
1	FFWT	PFOS: 21, PFHxS: 18, and PFOA: 2.9	Two-stage fractionation in presence of air, contact time: 15 min (per reactor)	PFOS, PFHxS, and PFOA: 0.01 (without polishing)		(OPEC)
2	SW	PFOS: 0.5, PFHxS: 0.5, PFOA: 0.3, PFHxA: 1.1, PFPeA: 1.0, H4PFOS: 18.4	Three-stage fractionation with ozone, Polishing treatment: NF, **large-scale**	PFOS: 0.01, PFHxS: 0.01, PFOA: 0.009, PFHxA: 0.01, PFPeA: 0.18, H4PFOS: 0.18 (without polishing) NF: 99.99%	Long-chain PFAS are fractionated into small-chain PFCA-based substances	(Ross et al., 2018)
3	WW	PFAS: 100-5,400	Large scale: Airport in Australia, 40,000 m^3 WW treated with a recovery of 98%	Removal 99.9% (PFAS: 0.04)		
4	GW	PFOS: 52.2, PFHxS: 12.5, PFPeS: 1.27, PFBS: 1.9, PFOA: 1.2, PFHxA: 5.2, PFPeA: 2.0, PFBA: 1.0, H4PFOS: 1.89	Three-stage fractionation with ozone, Polishing treatment: Adsorption GAC, **large-scale**	PFOS: <0.012, PFHxS: 0.001, PFPeS: 0.062, PFBS: 0.7, PFOA: 0.0005, PFHxA: 1.3, PFPeA: 2, PFBA: 0.002, H4PFOS: 0.001 (without polishing) Adsorption (GAC): 99.99%	Removal of short-chain PFAS e.g., PFPeA, PFBS, and PFBA is lower 35-63%	(EVOCRA, 2017)
Electrochemical oxidation						
1	GW	PFOS: 70, PFHxS: 120, PFBS: 16, DOC: 13; (pH: 6.1)	1 L, **Lab-scale**, Current density: 2.3 mA cm^{-2}, Power input: 1.4 W, Electrolysis time: 120 h	Removal 96%, DOC removal 96%	i) PFBS not treated ii) High concentrations of bromate: 124 $\mu g\ L^{-1}$ and perchlorate: 65 mg L^{-1} iii) AOX: 2.7 mg L^{-1}	(Schmidt et al., 2014; Trautmann et al., 2015)
2	Membrane elute treatment (polishing system)	PFOS:19,000, PFHxS: 10,000, PFBS: 4,000, DOC: 38; (pH: 8.4)	1 L, **Lab-scale**, Power input: 0.9 W, Electrolysis time: 18 h	Removal 80-90%, DOC removal: 48%	Energy consumption: 50 kWh m^{-3}	

S .No	Influent type	Influent concentration (µg L-1)	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
6	GW	PFBA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, and PFOS: 1 (each)	Dexsorb+ (0.01 g L^{-1}), lab-scale, contact time: 30 min	PFBA: 0.01, PFHxA: 0.05, PFHpA: 0.03, PFOA: 0.03, PFNA: 0.05, PFBS: 0.03, PFHxS: 0.03, and PFOS: 0.08	-	(Dexsorb+)
7	FFWT	PFOA: 6.6, PFHpA: 1.61, PFOS: 519, PFBS: 11.4	Dexsorb+, lab-scale,	PFOA: ND, PFHpA: ND, PFOS: 0.022, PFHxS: 0.001, PFBS: ND	-	(Dexsorb+)
c) RemBind						
1	GW	PFOS: 2.9-3.4, PFOA: 0.58-1.3, PFBS: 0.016-0.022, PFHxA: 0.36-1.2, H4PFOS: 0.84-1.3 (TOC: 1.9 mg L^{-1})	Batch, **pilot-scale**, contact time: 1 h, Pre-treatment: sedimentation followed by overnight decantation	PFOS: 0.15-0.17, PFOA: 0.058-0.13, PFBS: ND, PFHxA: 0.0036-0.012, H4PFOS:0.026-0.039	-	(Chiang et al., 2017a)
2	FFWT	PFAS:18,50,000	Lab-scale (filter column)	100% at 40 m^3		(Ziltek, 2017)
3	FFWT	PFOS: 9,000, PFHxS: 1,000, PFHpS: 200, PFBS: 20, PFOA: 80, PFHxA: 70, PFHpA: 20, PFBA: 20, PFPeA: 20 (TOC: 80 mg L^{-1}, COD: 270 mg L^{-1}, Turbidity: 1.5 NTU)	50:50 Rembind and silica sand, Flux: 0.9 BV h^{-1}, EBCT: 60 min	PFOS: 0.5, PFHxS: <0.01, PFHpS: <0.01, PFBS: <0.0004, PFOA: 0.001, PFHxA: <0.005, PFHpA: 0.001, PFBA: <0.0001, PFPeA: <0.0001	Low throughput and high-pressure loss	(USEPA, 2017b)
d) Ion exchange						
1	GW	PFOS: 0.027, PFOA: 0.43, PFBS: 0.17, PFBA: 0.21 (Sulphate: 44 mg L^{-1})	Purolite A532E, VF: 0.35 L, **lab-scale**, Flux: 6 m^3 h^{-1}, EBCT: 10 min, VF: 3 m h^{-1}	PFOS: 1,50,000 m^3, PFOA: 80,000 m^3, PFBS: 30,000 m^3, PFBA: 10,000 m^3	NH4Cl and NH4OH - 0.5%, KCl and KOH: 0.5%	(Zaggia et al., 2016)
e) Cyclodextrin (CD)-based adsorbents						
1	GW	PFOA: 50, pH 6.1-6.8	β-CD coated silica (1.5 g L^{-1}), lab-scale, contact time: 48 h	Removal: 95% PFOA: 2.5	-	(Bhattarai et al., 2014)
2	GW and FFWT	PFOA: 25 (pH 5.5) and PFAS: 200 (pH: 3)	β-CD coated magnetite (0.2-1.5 g L^{-1}), **lab-scale**, contact time: 24 h	Removal: 97% and 90%	-	(Badruddoza et al., 2017)
3	GW	PFOS: 50, PFOA: 50	β-CD coated magnetite (0.6 g L^{-1}), pH 5.5, **lab-scale**, contact time: 24 h	PFOS: 99% and PFOA: 96%	PFOS removal insensitive to pH change PFOA removal higher in acidic condition	(Badruddoza et al., 2017)

S. No	Influent type	Influent concentration ($\mu g\ L^{-1}$)	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
4	GW	Column exp: PFOS and PFOA: 20-30 Shaking exp: PFOS and PFOA: 50	β-CD coated magnetite (0.8 g L^{-1}), **lab-scale**, DOC: 4 mg L^{-1} in shaking exp, column exp: Flux: 0.3 BV h^{-1}, EBCT: 3 min Shaking exp: contact time: 24 h	Column: PFOS: 6,300 m^3 and PFOA: 6,300 m^3, shaking: PFOS: 1.5 and PFOA: 5	Increase in DOC from 4 to 12 mg L^{-1} reduced removal by 49%	(Bhattarai, 2015)
5	GW	PFOA: 1	Porous CD polymer (0.01 g L^{-1}), **lab-scale**, contact time: 24 h	PFOA: <0.01	No effect of DOC was observed	(Xiao et al., 2017a)
6	GW	PFBA, PFHxA, PFHpA, PFOA, PFNA, PFBS, PFHxS, and PFOS: 1 (each)	Dexsorb+ (0.01 g L^{-1}), **lab-scale**, contact time: 30 min	PFBA: 0.01, PFHxA: 0.05, PFHpA: 0.03, PFOA: 0.03, PFNA: 0.05, PFBS: 0.03, PFHxS: 0.03, and PFOS: 0.08	-	(Dexsorb+)
7	FFWT	PFOA: 6.6, PFHpA: 1.61, PFOS: 519, PFBS: 11.4	Dexsorb+, **lab-scale,**	PFOA: ND, PFHpA: ND, PFOS: 0.022, PFHxS: 0.001, PFBS: ND	-	(Dexsorb+)
Coagulation/Flocculation						
1	FFWT	PFOS+PFHxS: 180 PFOA:1.22	InSite (1 g L^{-1}), **lab-scale**, contact time: 60 s (per reactor)	>99.9% (without polishing)		(CARE, 2017)
2	FFWT	PFOS: 40-380	Insite, **lab-scale**, contact time: 60 s (per reactor)	PFOS <0.02-0.11	US EPA threshold value (0.07 $\mu g\ L^{-1}$) was not observed PFOS >300 $\mu g\ L^{-1}$	(InTreat Water)
Foam fractionation and ozofractination						
1	FFWT	PFOS: 21, PFHxS: 18, and PFOA: 2.9	Two-stage fractionation in presence of air, contact time: 15 min (per reactor)	PFOS, PFHxS, and PFOA: 0.01 (without polishing)		(OPEC)
2	SW	PFOS: 0.5, PFHxS: 0.5, PFOA: 0.3, PFHxA: 1.1, PFPeA: 1.0, H4PFOS: 18.4	Three-stage fractionation with ozone, Polishing treatment: NF, **large-scale**	PFOS: 0.01, PFHxS: 0.01, PFOA: 0.009, PFHxA: 0.01, PFPeA: 0.18, H4PFOS: 0.18 (without polishing) NF: 99.99%	Long-chain PFAS are fractionated into small-chain PFCA-based substances	(Ross et al., 2018)
	WW	PFAS: 100-5,400	Large scale: Airport in Australia, 40,000 m^3 WW treated with a recovery of 98%	Removal 99.9% (PFAS: 0.04)		

S. No	Influent type	Influent concentration (μg L^{-1})	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
3	GW	PFOS: 52.2, PFHxS: 12.5, PFPeS: 1.27, PFBS: 1.9, PFOA: 1.2, PFHxA: 5.2, PFPeA: 2.0, PFBA: 1.0, H4PFOS: 1.89	Three-stage fractionation with ozone, Polishing treatment: Adsorption GAC, **large-scale**	PFOS: <0.012, PFHxS: 0.001, PFPeS: 0.062, PFBS: 0.7, PFOA: 0.0005, PFHxA: 1.3, PFPeA: 2, PFBA: 0.002, H4PFOS: 0.001 (without polishing) Adsorption (GAC): 99.99%	Removal of short-chain PFAS e.g., PFPeA, PFBS, and PFBA is lower 35-63%	(EVOCRA, 2017)
Electrochemical oxidation						
1	GW	PFOS: 70, PFHxS: 120, PFBS: 16, DOC: 13; (pH: 6.1)	1 L, **Lab-scale**, Current density: 2.3 mA cm^{-2}, Power input: 1.4 W, Electrolysis time: 120 h	Removal 96%, DOC removal 96%	i) PFBS not treated ii) High concentrations of bromate: 124 μg L^{-1} and perchlorate: 65 mg L^{-1} iii) AOX: 2.7 mg L^{-1}	(Schmidt et al., 2014; Trautmann et al., 2015)
2	Membrane elute treatment (polishing system)	PFOS:19,000, PFHxS: 10,000, PFBS: 4,000, DOC: 38; (pH: 8.4)	1 L, **Lab-scale**, Power input: 0.9 W, Electrolysis time: 18 h	Removal 80-90%, DOC removal:48%	Energy consumption: 50 kWh m^{-3}	
3	IWW	PFOA: 2, PFHpA: 38, PFHxA: 25, PFPeA: 53, PFBA: 8, H4PFOS: 382, 6:2 FTAB: 1143; (TOC: 99 mg L^{-1}, pH: 7.6, conductivity: 6.9 mS cm^{-1}, chloride: 1,330 mg L^{-1})	2 L, **Lab-scale**, Current density: 50 mA cm^{-2}, Power input: 0.9 W, Electrolysis time: 10 h	Removal 99.7%	Energy consumption: 256 kWh m^{-3}	(Gomez-Ruiz et al., 2017)
Sonochemistry						
1	GW	PFOS: 100, PFOA: 100; (TOC: 20 mg L^{-1}, pH: 6.9-7.9)	0.6 L and **Lab-scale**, Power density: 250 W L^{-1}, ultrasonic frequency: 350 kHz, reaction time: 2h	PFOS: 68% and PFOA: 90%		(Vecitis et al., 2008)
2	FFWT	(pH 4)	91 L, **pilot scale**, Power density: 130 W L^{-1}, ultrasonic frequency: 350 kHz, reaction time: 13 h	PFSA: 90% and PFCA: 27%	At neutral pH efficiency decreased to 30%; Energy consumption: 100-300 kWh m-3	(Gole et al., 2018)
Vacuum distillation with plasma destruction						
1	GW	PFOS: 1.2, PFOA: 0.21, PFHxS: 0.4 (T: 15ºC, conductivity: 1150 μs cm^{-1}, TOC: 0.67 mg L^{-1}, Trichloroethane: 3.6, Tetrachloroethane: 0.33	3.8 L, **Lab-scale**, non-thermal plasma with Ar bubbling, Discharge frequency: 20-120 Hz, Discharge voltage: 16.5-25 kV, Reaction time: 30 min	PFOA: 99%, PFOS: 85%, and PFHxS: 60%	Complete removal not achieved	(Stratton et al., 2017)

S. No	Influent type	Influent concentration ($\mu g\ L^{-1}$)	Experimental parameters	Breakthrough/removal/degradation efficiency	Limitations	References
2	FFWT	PFOA: 45,000	0.2 L, **Lab-scale**, non-thermal plasma with Ar bubbling, Power consumption: 60 W, reaction time: 2.5 h	PFOA: 89%	-	(Obo et al., 2013)
3	FFWT	PFOA: 42,000, PFOS: 1,20,000	0.02 L, **Lab-scale**, non-thermal plasma with Ar bubbling, Power consumption: 95 W (3 h) and 250 W (8 h), reaction time: 3-8 h	PFOA: 98% in 3 h and 100% in 8 h	Toxicity of by-product not investigated	(Hayashi et al., 2015)
Fenton's reagent						
1	GW	PFAS: 200	0.1 L, **Lab-scale**, Magnesium-aminoclay coated zerovalent nanoscale iron, Iron dosage: 1 $g\ L^{-1}$	Removal: 70% (degradation: 40% and adsorption: 30%)	-	(Arvaniti et al., 2014)
2	GW	PFAS: 200 (pH:3)	0.1 L, **Lab-scale**, Magnesium-aminoclay coated zerovalent nanoscale iron, Iron dosage: 0.1-1 $g\ L^{-1}$, Reaction time: 1 h	PFOA: 38%, PFOS: 94%	No evidence of complete degradation	(Arvaniti et al., 2015)

7.1. Aqueous PFAS treatment technologies

Sorptive separation-based techniques

7.1.1. Sorption

Adsorption and ion exchange are the commonly used sorption technologies to remove PFAS from water and WW. Adsorption is a phase separation process where the adsorbate binds on the adsorbent surface via weak interactions (e.g., van der Waals and ionic interactions). In the case of the ion exchange process, the hydrophilic functional groups of PFAS are exchanged with the equivalent mass of innocuous ions of the ion exchange medium. So, these processes' efficiency depends on pH and concentrations of co-existing pollutants (e.g., natural organic matter).

GAC is widely used as an adsorbent to remove PFAS from GW and Fire-Fighting Wastewater (FFWT). The removal efficiency of GAC for both long and short-chain PFAS was in the range of 10-60 mg kg^{-1}, and it is effective (99.9%) in the removal of PFAS in the concentration range of 0.6-35,500 µg L^{-1}. Diffusion through the pores is the fundamental sorption mechanism. Flow-through fixed-bed filtration column is the widely used configuration at pilot-scale and large-scale, whereas stirred batch reactor is preferred set up at the lab scale (Rahman et al., 2014; Ross et al., 2018) (Figure 9). In the former case, contaminated water enters the adsorbent column, and PFAS are adsorbed on the adsorbent surface resulting in the purified effluent. Generally, the GW contaminated with PFAS has low concentrations of PFAS (0.6-10 µg L^{-1}). The maximum removal efficiency is obtained using a single adsorption column in this case. FFWT contains higher concentrations of PFAS (10-40,000 µg L^{-1}), and a series of sorbent columns are required to achieve a higher removal efficiency (>99%). The adsorption column needs replacement after reaching saturation point. The saturated adsorbent requires a secondary high temperature (800-1,000ºC) to de-

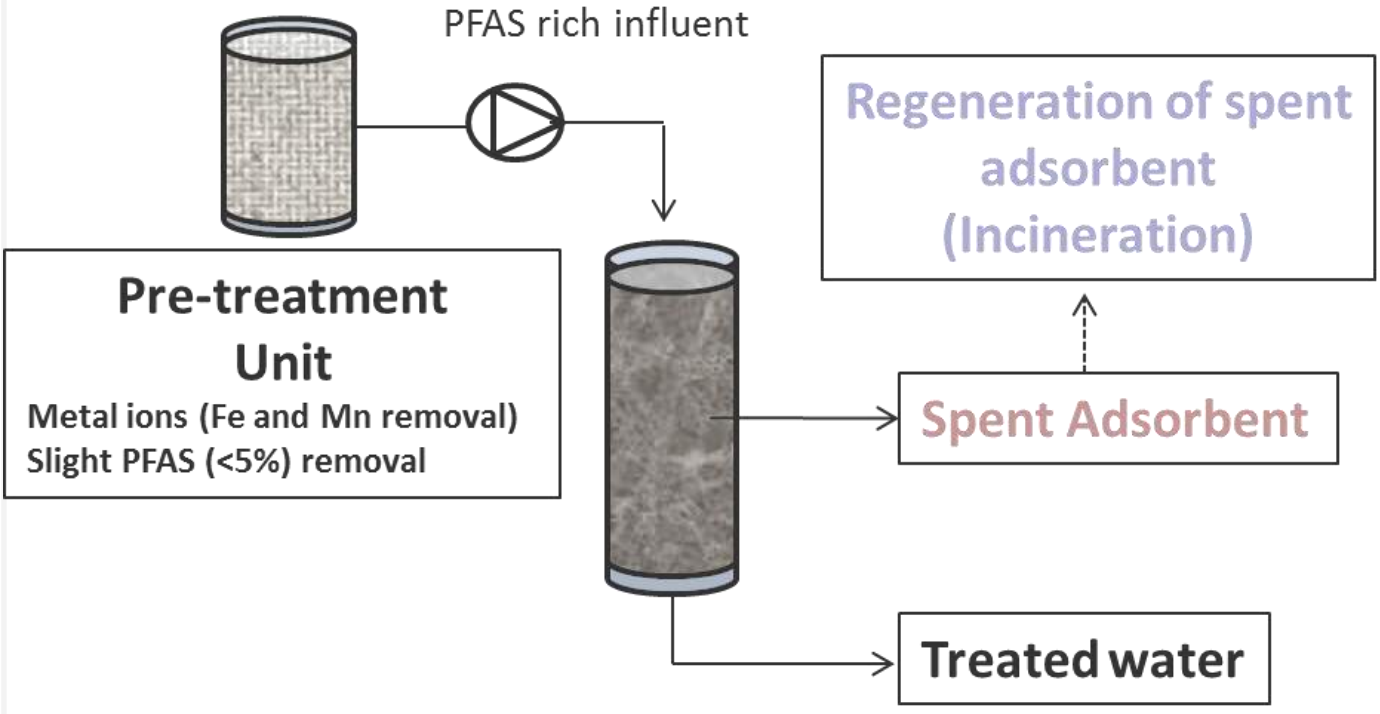


Figure 9. Schematic representing the adsorption techniques used to remove PFAS

stroy the adsorbed PFAS (Ross et al., 2018). Also, depending on the composition of the influent, pre-treatment using sand filtration is needed to remove other organic and inorganic impurities.

Treatment of PFAS contaminated GW using commercial (Norit) and coconut-shell-derived GACs could achieve a treated water volume of 8,000 to 40,000 m^3 with the flux and effective bed contact time (EBCT) of <7 m^3 h^{-1} and 15 min (TZW, 2011, 2015). The system exhibited >99.9% removal even in the presence of a higher concentration of sulfate (37 mg L^{-1}) and organic matter (DOC <1 mg L^{-1}). Commercial Filtrasorb 600 also removed >99.9% of PFAS (20 (PFBA)-9,000 (PFOA) µg L^{-1}) along with the high Total Organic Carbon (TOC) (80 mg L^{-1}) and Chemical Oxygen Demand (COD) (270 mg L^{-1}) (Szabo et al., 2017). Though Filtrasorb 600 showed commendable performance, the achieved effluent concentration of PFOS was not meeting the regulatory guideline value set by WHO. The Cooperative Research Centre for Contamination Assessment and Remediation of the Environment in Australia prepared an adsorbent called MatCare to treat FFWT in Australian Air Force Base. MatCare is made up of amine-treated palygorskite and can remove PFAS and its precursors (e.g., 1H, 1H, 2H, 2H-Perfluorooctan-Sulfonate (H4PFOS) and 8:2 Fluorotelomer Sulfonate (FTSA)) in pilot and large-scale studies (CRCCare and R., 2015; Kambala, 2016). Field-scale treatment (10 m^3) of actual Aqueous Film Forming Foam (AFFF) WW containing 35 and 6.85 mg L^{-1} of PFOS and PFOA using 10 kg of MatCare resulted in greater than 99% removal (Kambala, 2016). The adsorption capacity of MatCare was 1,112 mg g^{-1} (Kambala, 2016). Low throughput was observed in the case of high concentrations of PFAS (>1,000 µg L^{-1}). As a result, frequent replacement of the column was required to treat higher concentrations of PFAS contaminated WW. Ziltek, Australia also developed a composite of activated carbon (amorphous aluminum hydroxide, activated carbon, organic matter, and the mineral kaolinite) called RemBind to achieve higher adsorption capacity in PFAS removal from GW and FFWT. RemBind combined with sand filtration unit showed complete removal of PFAS up to <9,000 µg L^{-1} in 60 min. However, high-pressure loss and low throughout were the main hurdles in their continuous operation (Chiang et al., 2017b). Injectable particulate carbon is used as a 'trap and treats' in-situ technology to treat PFAS-contaminated GW (Regenesis, 2017). The dispersion of particulate carbon throughout the aquifer restricts the mass flux and offers the adsorption of PFAS on the carbon surface. However, lab-scale studies demonstrated that particulate carbon is effective for long-chain PFAS removal while it is ineffective for removing short-chain and PFAS precursors (Xiao et al., 2017b). The saturated particulate carbon acts as a source zone for the subsequent release of the PFAS over time (Ross et al., 2018). So, the injectable particulate carbon technology is not recommended in pilot or large-scale in-situ removal of PFAS.

CD-based adsorbents were prepared by considering the electrostatic interactions between the cup-shaped CD molecules and carbon atoms of the PFAS. β-CD coated silica removed 50 µg L^{-1} of PFOA at neutral pH within 48 h (Bhattarai et al., 2014). Replacing silica with magnetite improved the removal efficiency (97%) and treatment time (24 h), but it required acidic pH (3-5.5). The PFOS removal using β-CD coated magnetite is independent of pH variation and depends on DOC concentration (Bhattarai et al., 2014; Badruddoza et al., 2017). Porous CD polymer showed >99.9% removal for PFOA from contaminated GW, and the adsorption capacity was not influenced by DOC or pH (Xiao et al., 2017a). In terms of commercialization of CD-based adsorbents, Cyclopure (US) prepared a cross-linked CD from cornstarch called Dexsorb+ with the adsorption capacity of 90 (short-chain) -180 (long-chain) mg g^{-1}. The material showed 90% removal of long-chain PFAS and >99% short-chain PFAS from FFWT within 30 min

(Dexsorb+). Among the listed adsorbents, Dexsorb+ showed faster kinetics and higher regeneration ability. However, their efficacy in treating PFAS concentrations greater than 1000 ng L^{-1} needs to be verified. ABS materials developed cross-linked silica-based alkoxysilanes with the trade name of Osorb (Materials, 2013). Osorb tends to adsorb organic compounds with the log K_{OW} value of 2.5, and it tends to float on the surface of the water and starts swelling after absorbing the PFAS. It shows a cumulative removal efficiency of ~95% for removing PFOS, PFOA, and PFBA. Pressure swing desorption in the presence of butane gas provided ~98% of the regeneration of Osorb. However, pH and organic matter reduced the efficiency in the pilot-scale, and so far, large-scale testing of Osorb to treat a broad spectrum of PFAS is not available.

Conventionally, ion-exchangers are used to treat PFAS contaminated GW. Purolite A532E was tested to remove PFAS in the presence of higher sulfate concentrations (44 mg L^{-1}) and offered a bed volume of 1,50,000 m^3 for PFOS (Zaggia et al., 2016). Additionally, it offered simple regeneration of spent adsorbent using a mixture of NH_4Cl and NH_4OH. Overall, pilot and lab-scale investigations indicated the possibility of extending the adsorption to large-scale treatment units for removing PFAS in GW and FFWT. However, more research is needed to verify the influence of multiple co-contaminants (e.g., organic and inorganic impurities). Also, the regeneration success and shelf-life of the adsorbents need to be analyzed.

GAC has been used for over a decade to treat PFAS (specifically PFOA, PFOS, and PFNA). GAC-based adsorption columns have been used to treat PFAS-contaminated water in 45 military sites in the US (ITRC, 2018). The total cost to treat 1.89 m^3 d^{-1} requires USD 50,000, the bed volume of PFOA and PFOS is USD 31,000 and 45,000, and EBCT was 20 min (USEPA, 2020a). Additionally, this treatment option is effective in point-of-use-based applications to treat about 225 gallons per day with the cost of $280 (USEPA, 2020a). The installation of Point-Of-Use (POU)-based GAC treatment requires 4'×4' room; however, the frequent GAC replacement is the main drawback. Another limitation is the regeneration of spent adsorbent. Currently, thermal-swing-desorption is commonly used to regenerate the GAC. Nonetheless, the fate of desorbed PFAS in the regeneration process is unknown. Apart from these limitations, GAC is the best and recommended technology to remove PFAS from water and WW.

7.1.2. Coagulation/Flocculation

The principle of coagulation/flocculation is the addition of a chemical compound known as coagulant or flocculants into WW to remove suspended solids and dispersed particles in the form of precipitate or sludge (Figure 10) (Riegel et al., 2020). Iron chloride or aluminum chloride have been commonly used as coagulants, and the normal sedimentation and filtration processes remove suspended particles up to 85 -90% (ITRC, 2018). However, this type of conventional coagulation process is ineffective for PFAS removal. For example, aluminum sulfate and polyaluminium chloride combined with air flotation removed only 49% of PFOS. Likewise, ferric chloride showed 3 to 12% removal for short and long-chain PFAS. From these preliminary investigations, a specific coagulant that separates the PFAS from the influent is essential. Insite remediation services, Australia developed a flocculent called InTreat, which showed higher efficiency (>99.9%) in treating PFAS in the concentration ranges of 1,000-12,000 ng L^{-1} within a short reaction time of 60 s (Insite, 2017).

However, multistage flocculation units are required to treat PFAS concentrations greater than 10,000 ng L^{-1}. Treatment of FFWT with relatively higher concentrations (e.g., 40,000 ng L^{-1}) of PFOS results in poor removal efficiency. In most cases, the effluent PFOS concentrations are higher than the regulatory guideline values. A German company, Cornelsen Umwelttechnologie, prepared a specific coagulant called PerfluorAd to treat PFAS concentrations greater than 300 ng L^{-1} (TRS, 2020). The treatment efficiency of PerfluorAd (100 mg L^{-1}) for PFOS, PFOA, and PFHxS was 90%, and a polishing treatment unit was necessary to achieve the threshold value. PerfluorAd also removed turbidity. But it showed poor removal of manganese (TRS, 2020). Coagulation/flocculation is a cost-effective and easy-to-operate method to remove long-chain PFAS. However, not many pilot-scale studies have been conducted, and its effectiveness for treating short-chain PFAS is unknown. The influence of co-contaminants and pH are not verified, and vendors have not suggested proper methodology for PerfluorAd-based systems. None of the vendors revealed the exact dosage of PFAS to remove PFAS from the FFWT at the pilot-scale level. More transparent research results are needed to identify the large-scale application of coagulation/flocculation.

Although coagulation is considered as a conventional treatment to remove PFAS, the efficiency in pilot and large-scale is ineffective (less than 10%). The pH-dependent efficiency of coagulation restricts its application in field-scale because it requires special chemicals/buffers to maintain the pH. Even so, the treatment is ineffective (<30%) in treating PFAS contaminated water at neutral pH. Also, coagulation is unsuccessful in treating AFFF contaminated WW and its necessary adsorption as an adjunct unit. Finally, incineration is required to destroy the PFAS loaded sludge. This increases the operation cost and sec-

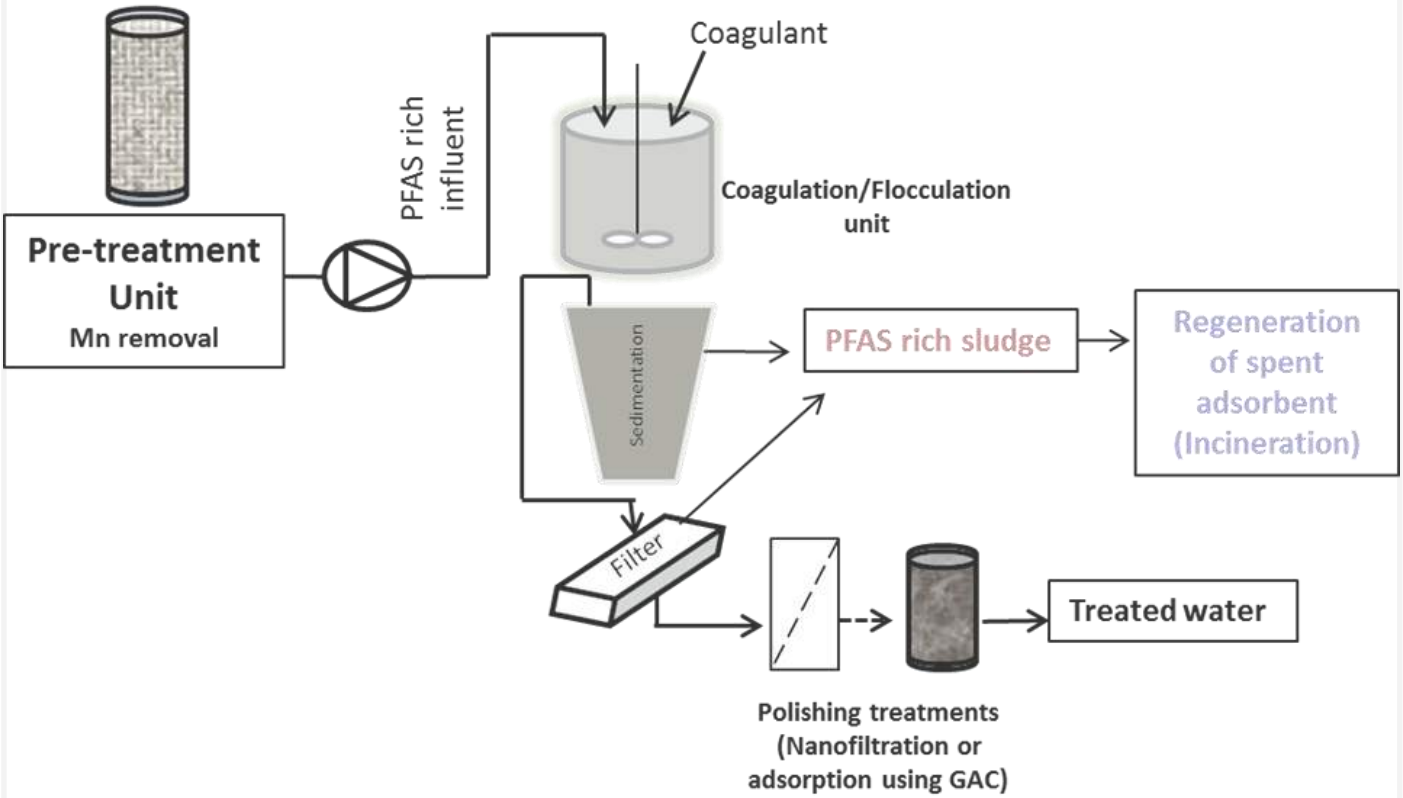


Figure 10. Schematic representing the coagulation techniques used to remove PFAS

ondary pollution (e.g., emission of PFAS intermediates). Therefore, coagulation is not a recommended technology to treat PFAS.

7.1.3. Foam fractionation

PFAS possess a surfactant nature (i.e., hydrophilic and hydrophobic functional ends), and in the presence of foam, PFAS adhere to the bubble walls via the gas-liquid partition phenomenon (Figure 11). The collected PFAS-loaded foam is transferred to the water surface, and the collected concentrate requires additional post-treatment. OPEC systems, Australia developed Surface Active Foam Fractionation (SAFF) as an on-site continuous treatment unit to remove PFAS in the concentration range of 0.1 to 10^8 µg L^{-1} (OPEC, 2020). The large-scale two-stage foam fractionation in compressed air results in 99.9% removal of PFOS, PFHxS, and PFOA from FFWT. The effluent PFAS concentrations were below the regulatory guideline values, so the polishing treatment was unnecessary. Similarly, another Australian company EVOCRA has used ozone instead of air to produce foam, and the technology showed higher removal efficiencies also for PFAS precursors (e.g., H4PFOS). Three-stage ozofractionation of sewage water loaded with 0.3 (PFOA) to 18.4 (H4PFOS) µg L^{-1} resulted in 99% of removal efficiency (EVOCRA, 2016). Additionally, using nanofiltration as a polishing step showed the complete removal of PFAS. The large-scale treatment unit of ozofractionation is capable of treating 40,000 m^3 of WW per day in an Australian airport (Ross et al., 2018). The removal efficiency was 98%, and the effluent PFAS concentrations were 0.04 µg L^{-1}. The three-stage ozofractionation unit was also used to treat long and short-chain PFAS in the GW. Without polishing treatment, 98% removal efficiency was observed for long-chain PFAS, whereas 35-63% was observed for short-chain PFAS (EVOCRA, 2016). 99.99% of long-chain PFAS was removed using adsorption as a polishing treatment. The estimated volume of concentrated foam from a two-stage ozofractionation was ~0.5 to 2% of the influent volume (Ross et al., 2018). On the other hand, SAFF with air develops a foam-concentrate of 0.0025% of the influent volume (EVOCRA, 2016). So, SAFF is found to be a better option to remove PFAS than ozofractionation. The estimated cost of the foam fractionation technique is 3 to 7 kWh m^{-3}, and polishing treatments such as adsorption cost an additional 2 to 3

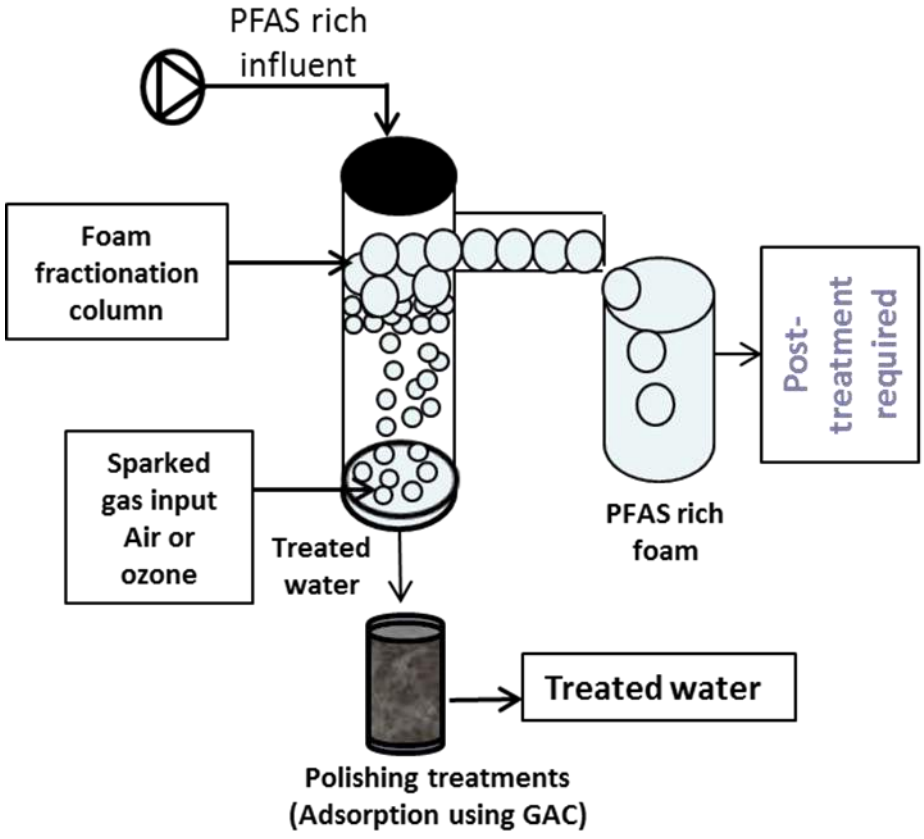


Figure 11. Schematic representing the foam fractionation techniques used to remove PFAS

kWh m^{-3} (OPEC, 2020). The presence of ozone is also beneficial because it minimizes the interferences developed by co-contaminants such as natural organic matter, salt content, and petroleum hydrocarbons (Horst et al., 2018).

Overall foam fractionation is considered another possible treatment to treat PFAS with concentrations less than 6,000 µg L^{-1}. For treating low concentrations of PFAS, multi-stage foam fractionation is recommended. The generation of short-chain PFAS from the long-chain is essential because they are more persistent and mobile. It is noteworthy that foam fractionation or ozofractionation is ineffective for short-chain PFAS treatment (35-63%) (EVOCRA, 2016). So, an additional polishing unit such as adsorption and nanofiltration is necessary to increase the treatment quality, which elevates the operational cost. Therefore, foam fractionation is not recommended primarily, as adsorption using GAC yields a similar result.

Destruction-based techniques

7.1.4. Electrochemical oxidation

Electrochemical oxidation is a process in which electrons from the electrodes are used to degrade pollutants in the bulk liquid (Figure 12). It is an electron transfer process, and the efficiency of the electron generation is directly proportional to the applied potential. The Boron-Doped Diamond (BDD) electrode is commonly used to destroy PFAS from GW and industrial WW (Gomez-Ruiz et al., 2017). Electrochemical oxidation is also used to treat the concentrate of PFAS resulting from the sorptive separation processes (e.g., membrane elute). Electrochemical oxidation is a pH-dependent process and shows maximum efficiency at alkaline pH. For example, lab studies to remove low concentrations (13-120 µg L^{-1}) of PFOS, PFHxS, and PFBS at the density of 2.3 mA cm^{-2} showed 96% degradation efficiency with the electrolysis time of 120 h (Michael and Kathrin, 2014). High concentrations of PFAS (4,000 to 19,000 µg L^{-1}) were treated via the BDD system with the power input of 0.9 W, resulting in 80 to 90% degradation, while it showed only 48% removal of DOC. This result shows the necessity of a polishing treatment unit to achieve greater than 99.9% of PFAS degradation. The treatment of industrial WW containing moderate concentrations of PFAS (e.g., PFOA: 2 µg L^{-1} and PFPeA: 53 µg L^{-1}) and its precursors (e.g., H4PFOS: 382 µg L^{-1} and 6:2 FTAB: 1,143 µg L^{-1}) with high concentrations of chloride (1,330 mg L^{-1})

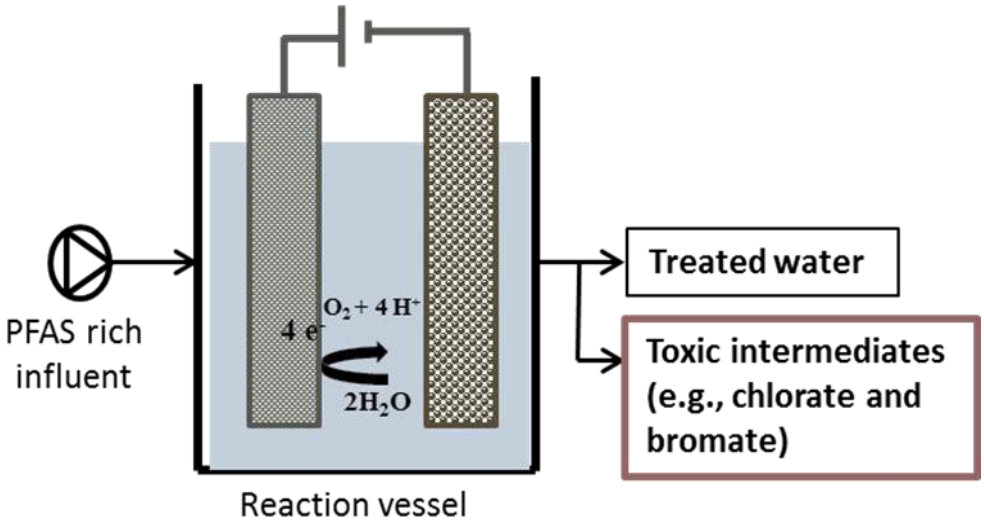


Figure 12. Schematic representing the electrochemical oxidation to remove PFAS

showed removal efficiency of 99.7% with the power input and electrolysis time of 0.9 W and 10 h, respectively (Gomez-Ruiz et al., 2017).

Electrochemical oxidation is a non-selective oxidation process. Therefore, the formation of chlorinated (e.g., ClO_3^- and ClO_4^-) and brominated (BrO_3^-) by-products is inevitable, and so far, none of the studies have investigated the toxicity of the treated solution. However, the generation of these by-products (e.g., short-chain and fluorinated derivatives) leads to ecotoxicological and human toxicological impacts. The estimated energy consumption of this process was 50 to 256 kWh m^{-3}, which is far higher than sorption technologies (Trautmann et al., 2015; Gomez-Ruiz et al., 2017). The efficiency of the electrochemical unit is unknown at pilot and large-scale. Considering the toxicity of end-products, specific experimental setup, and expenses, electrochemical degradation is ineffective in treating PFAS from water and wastewater.

7.1.5. Ultrasound technology

The principle of ultrasound technology is like foam fractionation. The application of ultrasound frequencies in the ranges of 20 to 1,100 kHz, creates cavitation by collapsing the microbubbles produced in the reaction medium (Figure. 13) (Cao et al., 2020). Subsequently, the hydrophobic end of the PFAS accumulates in the bubble-water interfaces and results in pyrolytic decomposition (Vecitis et al., 2008). The increase in the PFAS chain length increases the degradation rates via increasing the hydrophobic properties. The method is effective for PFOA and PFOS up to the concentration ranges of 10,000 µg L^{-1}. Typically 200 kHz is required to degrade PFOA and PFOS in GW, and the estimated power ranges from 100 to 300 kWh m^{-3} (Vecitis et al., 2009). The presence of lower concentrations (20 mg L^{-1}) of dissolved organic matter did not affect the degradation. In contrast, volatile organic compounds (VOCs) negatively impacted the degradation of PFAS (Gole et al., 2018). VOCs reduced the bubble vapor rate and microbubble collapsing speed. So, VOCs act as a competing co-existing pollutant for PFAS and reduce the cavitation yield. Therefore, the treatment of PFAS in petroleum-contaminated sites or water needs a proper pre-treatment system before the ultrasound destruction. Pre-treatment is also necessary for the influent containing high concentrations of metal ions (e.g., Fe^{2+}/Fe^{3+} and Mn^{2+}) (Cheng et al., 2008). The treatment of PFOS and PFOA (100 µg L^{-1}) under the ultrasonic frequency of 350 kHz for 2 h resulted in 68 and 90% removal of PFOS and PFOA, respectively. A similar reactor step showed greater (90%) removal efficiencies for PFSA than PFCA (27%). Removal of PFAS was prominent

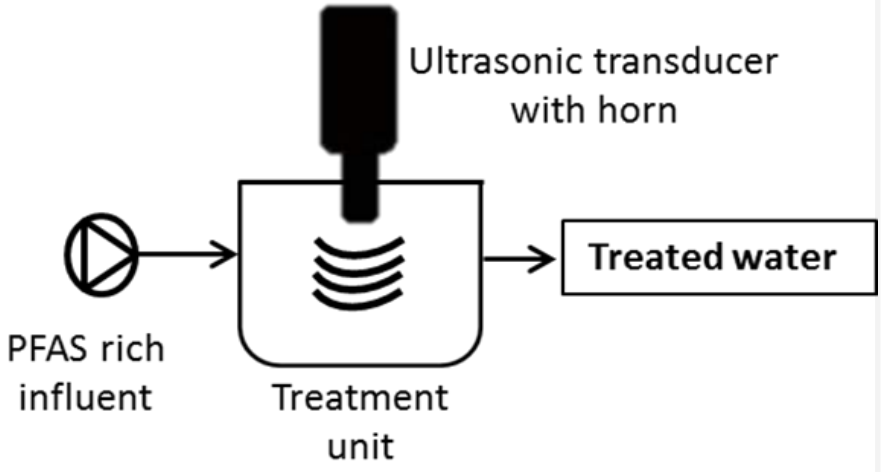


Figure 13. Schematic representing the sonochemical techniques to remove PFAS

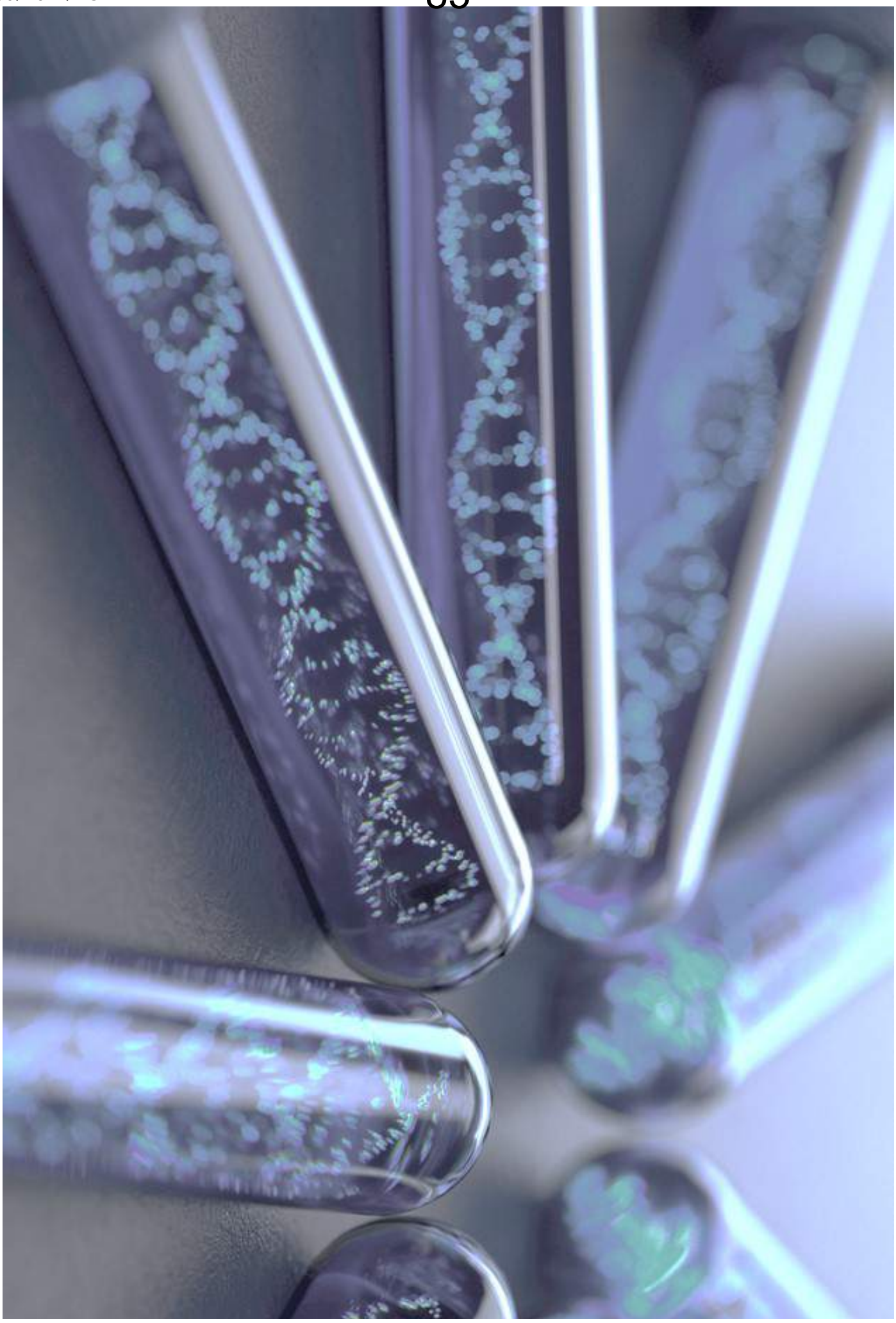
at acidic pH (4), and an increase in the pH to neutral decreased the degradation efficiency by 30% (Gole et al., 2018). Sonochemical technologies completely mineralize the PFAS, so no post-treatment or polishing step is required.

Sonochemical destruction was performed at the lab and pilot-scale levels using a lower volume of influent (maximum 91 L). Increasing the influent volume might decrease the cavitation effect and using more transducers and improper supply of stirring speed may reduce the bubble formation. Also, the large-scale operation requires a cooling jacket as the temperature of the reaction medium increases up to 5,000 K. Low pH is favorable for PFAS destruction, as it necessitates additional chemicals to neutralize the pH of the solution before and after treatment (Gole et al., 2018). Sonochemical technology is ineffective in treating PFAS, which is co-contaminated with petroleum compounds because VOCs are known to destroy the bubble vapor pressure and collapse (Cheng et al., 2008). The energy cost of ultrasound technology is 100-350 kWh m^{-3} which is far higher than adsorption and electrochemical degradation (Ross et al., 2018). Therefore, ultrasound technology is not a feasible treatment method for treating PFAS.

7.1.6. Vacuum distillation with plasma destruction

Vacuum distillation is used to concentrate the PFAS in the influent and applying plasma to the PFAS concentrate results in complete mineralization. The plasma destruction unit consists of a high voltage pulse generator with an oscilloscope. It produces plasma with a core temperature of 10,000ºC, and the PFAS is degraded via the thermal mineralization phenomenon. Plasma destruction is more cost-intensive than the normal incineration used to treat PFAS concentrate. On the other hand, incineration may develop toxic PFAS vapor, requiring a stripping unit to capture the escaping pollutants. ECON industries, Germany have developed a vacuum distillation aided plasma destruction unit called 'VacuPlas,' and it is a mobile unit with the size of two shipping containers (ECON). Perhaps, the high-temperature of plasma requires special production measures to successfully achieve the desired degradation of PFAS. Non-thermal plasma technology offers destruction of PFAS in the presence of reactive radicals produced from the electrical discharge (Stratton et al., 2017). The mineralization efficiency of this process is similar to thermal destruction. In both cases, the PFAS concentrate sprayed near the plasma discharge unit showed maximum removal (Obo et al., 2013; ECON, 2019). The lab-scale studies for the treatment of low concentrations of PFAS (e.g.,0.42-1.2 µg L^{-1}) in GW using a non-thermal plasma process resulted in the 99% removal of PFOA within 30 min. Similarly, treatment of FFWT with relatively higher concentrations of PFAS (PFOA: ~40,000 and PFOS: 1,20,000 µg L^{-1}) showed 80-90% removal in 3 to 8 h.

The vacuum distillation with non-thermal plasma resulted in higher removal and mineralization efficiency, and this treatment option was inefficient (60% removal) for treating PFSAs (e.g., PFOS and PFHXs). In some cases, the presence of co-existing pollutants such as trichloroethane developed chlorinated by-products. It increases the toxicity of the effluent. On the other hand, organic matter, pH, and salt concentration did not affect the efficiency of the process. The estimated energy requirement of vacuum distillation with plasma treatment is 4,500 kWh m^{-3}. The energy demand and cost largely hinder their application. Lack of information on the reactor design and degradation mechanism and higher energy cost informs the inefficiency of the pulse power technology to treat PFAS.



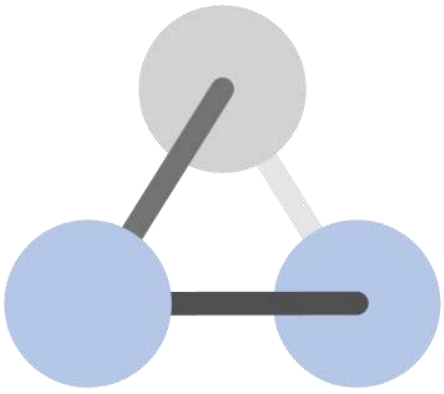
7.1.7. Reduction technologies

Nanoscale Zerovalent Iron (nZVI) is a commonly used PFAS reducing agent for in-situ treatment. The electrons produced by the zerovalent iron decompose the PFAS into small molecular weight, non-toxic byproducts (Crane and Scott, 2012). The fundamental mechanism is the complex formation between PFAS and the surface charges of the nZVI. Lab-scale studies for the removal of PFAS (200 µg L^{-1}) in GW indicated 38-94% removal at pH 3 (Arvaniti et al., 2014; Arvaniti et al., 2015). Generally, the adsorbent dosage of 0.1-1 g L^{-1} of nZVI was used to remove PFAS concentrations less than 1,000 µg L^{-1}. However, there is no evidence for the complete degradation of PFAS via reduction technologies. Additionally, the oxidation of nZVI is a crucial limitation that reduces its adsorption capacity by about 30% within three days (Crane and Scott, 2012). Therefore, special storage in the presence of non-reactive gases (e.g., Ar or N_2) is required.

Overall, lab-scale studies indicated that achieving degradation efficiency for PFAS up to the regulatory guideline values is not feasible with nZVI-based technology and is not a recommended treatment option. The cost of nZVI is relatively higher (USD 200 kg^{-1}) than GAC and ion-exchange membranes. Reduction technologies showed no removal of PFAS at neutral pH. It is not beneficial in terms of the actual pH of the influent (e.g., 5-7) (Arvaniti et al., 2014; Arvaniti et al., 2015). Also, the efficiency of reduction technologies to treat PFAS in the presence of co-contaminants is unavailable. From these limitations, reduction technology is at the toddler level. So, it is not a preferred method to treat PFAS.

7.1.8. Chemical oxidation

Chemical oxidation using ozone (O_3), hydrogen peroxide (H_2O_2), O_3/UV, persulfate, and Fenton's reagent demonstrated considerable removal efficiencies for PFOA, yet they were ineffective in treating PFOS (Qiu et al., 2006). Other strong oxidants like potassium dichromate ($Cr_2O_7^-$) and potassium permanganate (MnO_4^-) were completely inefficient against PFAS (Qiu et al., 2006). UV/persulfate and heat-activated persulfate-based oxidations showed greater than 95% removal for PFOA and achieved complete defluorination (Hori et al., 2005). Lab studies showed the maximum efficiency of persulfate-based oxidation at acidic pH and drastic degradation of efficiency at neutral and alkaline pH. Likewise, H_2O_2 with Fenton's reagent showed >85% of PFOA removal within 150 min (Mitchell et al., 2014). Further investigation of the treated solution showed poor defluorination efficiency. Therefore, considering the strong binding ability of the C-F bond, the advanced oxidation processes are not effective for treating PFAS in field-scale applications. The production of short-chain PFAS, low defluorination efficiency, and unfavorable



experimental parameters (e.g., acidic pH) limit the extension of lab-scale chemical oxidation-based technologies to pilot-scale.

Ozone-based chemical oxidation is not yet found to be suitable for treating PFAS. Most importantly, hydroxyl radical ($OH^{\bullet}$) attacks only the alkyl groups of the PFAS, and it does not react with the perfluoroalkyl chain. It results in the transformation of PFAAs to PFCAs (Bruton and Sedlak, 2017). Further destruction of PFCAs is not feasible due to the inefficacy of $OH^{\bullet}$ to break the C-F bond. Therefore, the chemical oxidation technology (e.g., H_2O_2, UV, and photocatalysis) works under the generation of $OH^{\bullet}$ is not beneficial to degrade PFAS. The persulfate-based system mineralizes PFCA into their short-chain compounds. Further decarboxylation results in defluorination. However, sulfate radical is unreactive with PFSAs and transforms them into PFCAs (Bruton and Sedlak, 2017). Therefore, the production of short-chain PFCAs is evident and complete mineralization of these short-chain PFAS by chemical oxidation is not yet achieved. Therefore, chemical oxidation is not recommended to treat PFAS.

To this end, in comparison with destruction technologies, adsorption using activated carbon-based adsorbent is an economically viable and technically sound treatment method for PFAS. However, care should be taken for storing and treating the spent adsorbent via incineration. The improper design of spent adsorbent disposal may lead to the secondary contamination of PFAS.

7.1.9. Biodegradation

Most PFAS are resistant to biological degradation, and some polyfluorinated compounds are known to biotransform into their precursors. For example, conversion of fluorotelomer thioether amido sulfonate into 4:2, 6:2, and 8:2 fluorotelomer sulfonate were observed by Harding-Marjanovic et al. (2015). The defluorination of PFOS and PFOA using A6 culture over a 100-day incubation time resulted in 60% efficiency (Huang and Jaffé, 2019), and no information related to degradation mechanism and by-product formation is provided. Therefore, relatively limited research informs the inefficiency of biodegradation for treating PFAS.

7.1.10. Summary of the treatment technologies for PFAS removal

Detected concentrations of PFAS in GW were ng L^{-1} to µg L^{-1}, and the ex-situ treatment of PFAS generally possesses lesser complexity than the in-situ treatment. GAC is the preferred adsorbent for large-scale treatment of GW contaminated with long-chain PFAS, whereas ion-exchangers are effective for short and long-chain PFAS (Table 7). In both cases, the organic matter, pH, and salt concentration need to be maintained. Pre-treatment options to remove iron and manganese increase the life of the GAC-based adsorption column. In removing high concentrations of PFAS, hybrid technologies such as flocculation coupled with adsorption offer higher removal efficiency. The flocculation efficiency of PerfluorAd or Insite at 0.1-1 g L^{-1} is effective for treating PFAS greater than 10,000 µg L^{-1} and adding adsorption as a polishing unit result in 99% PFAS removal. The estimated annual cost to treat PFAS is around $50,000 for 0.5 MGD. Sludge management and regeneration of adsorbent demand 8-17% of the annual cost (Water Environment Federation, 2020). So, regeneration and frequent saturation of adsorbent columns at high concentrations of PFAS are the two notable drawbacks of this technology. Apart from this, adsorption is the best way to economically and effectively treat PFAS.

Concentrating the PFAS from the bulk solution is another way to achieve maximum removal of PFAS. In this case, liquid-liquid separation technologies such as nanofiltration, Reverse Osmosis (RO), and foam fractionation are apt to concentrate the PFAS in the presence of co-existing pollutants. A membrane process is viable for removing low to moderate concentrations (ng L^{-1} to µg L^{-1}) of PFAS and is ineffective for treating PFAS containing high salt concentrations. Membrane technology favors pre-treated influents to offer maximum removal. Also, post-separation of PFAS from the membrane is an energy-intensive pressure-swing desorption process. Therefore, it is not recommendable as a standalone unit. An ion exchanger is an economical separation option to concentrate PFAS. CD-based cross-linked materials offer excellent regeneration features. So, most of the time, it is used as a polishing treatment. Ozofractionation is a suitable method for treating very high concentrations of PFAS (e.g., FFWT), and producing high-quality water, but sludge management is expensive and complex. So, it is not recommended. Destruction technologies create secondary pollution by converting long-chain PFAS into short-chain precursors. So, it is not a feasible option. Overall, considering the technical maturity and practical application, adsorption is a feasible treatment option to remove PFAS on a field scale.

Table 7. Comparison of treatment technologies used to treat PFOA and PFOS.

(The pros and cons of the treatment options summarized based on the discussion from (USEPA, 2021c) and (ITRC, 2020)

Treatment	PFOA			PFOS		
	Advantages	Limitations	Possibility in practical scale	Advantages	Limitations	Possibility in practical scale
Aeration and air stripping	Not applicable	Lab-scale, pilot-scale, and full-scale is ineffective	×	Not applicable	Lab-scale, pilot-scale, and full-scale is ineffective	×
Biological filtration	Not applicable	Lab-scale, pilot-scale, and full-scale is ineffective	×	Use of GAC as a filter material increased PFOS removal	Ineffective for treating high concentrations of PFOS and other PFCA or PFSA	×
Biological treatment	Not applicable	Aerobic and anaerobic condition is ineffective (0-8% removal)	×	Not applicable	Aerobic and anaerobic condition is ineffective (0-10%)	×
Chlorine/Chlorine dioxide	Not applicable	Ineffective- seven full-scale studies [2441, 2509, 2619]	×	Not applicable	Ineffective- seven full-scale studies [2441, 2509, 2619]	×
Coagulation	High-affinity coagulants (e.g., CD) superior removal	1] Partially developed technology 2] pH dependent removal [2518] 3] Limited information on the treatment of PFAS rich sludge	×	High-affinity coagulants (e.g., CD) superior removal	Neutral pH: no removal, Pilot scale no removal [2518]	×
GAC-based adsorption	1] High removal and feasible technology 2] longer service time 3] Regeneration option 4] Effective to remove short-chain PFAS 5] Cost-effective 6] Point of use and point of entry-based applications	Disposal and reactivation of spent adsorbent	*	1] High removal and feasible technology 2] Longer service time 3] Regeneration option 4] Effective to remove short-chain PFAS 5] Cost-effective 6] Point of use and point of entry-based applications	1] Poor adsorption on short-chain PFOS 2] Disposal and reactivation of spent adsorbent	*
Ion Exchange	1] High removal and feasible technology 2] longer service time 3] Cost-effective 4] Point of use and point of entry-based applications	1] Poor removal for short-chain PFCA 2] Liability issues related to incineration of resin 3] Unknown regeneration capacity	*	>99% removal [2424, 2441, 2504, 2568]	1] Poor removal for short-chain PFCA 2] Liability issues related to incineration of resin 3] Unknown regeneration capacity	*

Treatment	PFOA			PFOS		
	Advantages	Limitations	Possibility in practical scale	Advantages	Limitations	Possibility in practical scale
Membrane filtration/ separation	1] Good for PFAS with the molecular weight <700 g mol^{-1} 2] Suitable for all types of PFCA 3] Effective treatment in presence of salt concentration (<1%)	1] High capital cost 2] Mandatory corrosion control 3] Unclear solutions to treat concentrated stream 4] Membrane fouling at high concentrations of PFAS	×	Effective Removal at low concentrations of PFOS	1] Negative removal 2] Back flush requires higher pressure 3] Treatment efficiency depend on pore size 4] Mandatory polishing unit	×
Electrochemical treatment	1] High mineralization for PFCAs 2] Suitable to treat high concentrations of PFAS	1] No pilot or large-scale treatment 2] Increase in current density increases the removal and cost 3] In effective to treat low concentrations of PFAS	×	Effective at bench scale	1] No pilot or large-scale treatment 2] Increase in current density increases the removal and cost 3] In effective to treat low concentrations of PFAS	×
Plasma treatment	1] Diverse reactive radicals increase the mineralization of PFAS 2] Higher efficiency at shorter reaction time	1] Lower efficiency in presence of co-contaminant 2] No pilot or large-scale treatment 3] Unclear degradation mechanism and fate of by-products	×	1] Diverse reactive radicals increase the mineralization of PFAS 2] Higher efficiency at shorter reaction time	1] Lower efficiency in presence of co-contaminant 2] No pilot or large-scale treatment 3] Unclear degradation mechanism and fate of by-products	×
Persulfate based oxidation	1] Capable of destructing alkyl chain of PFAS	1] Requires alkaline pH and end solution pH is acidic 2] Presence of Fe^{2+}/Fe^{3+} reduced performance 3] PFHpA, PFHxA, PFPeA, and PFBA found as intermediates (Liu et al., 2012)	×	Not applicable	1] Ineffective to treat PFAS 2] In some cases, it converts PFSA into PFCA 3] Generates untreatable intermediates	×

Treatment	PFOA			PFOS		
	Advantages	Limitations	Possibility in practical scale	Advantages	Limitations	Possibility in practical scale
Ozone	1] Generation of superoxide radical help increase mineralization efficiency	1] Many full-scale studies found ozonation is ineffective 2] High ozone production cost and instability of ozone 3] Generation of short-chain intermediates	×	1] Generation of superoxide radical help increase mineralization efficiency	1] Many full-scale studies found ozonation is ineffective 2] High ozone production cost and instability of ozone 3] Generation of short-chain intermediates	×
Ozone and H_2O_2	Hydroperoxide radical increase the defluorination efficiency	1] No practical application testified 2] Operation under neutral pH is not known 3] By-products and mechanism is unclear	×	Hydroperoxide radical increase the defluorination efficiency at alkaline pH	1] No practical application testified 2] Operation under neutral pH is not known 3] By-products and mechanism is unclear 4] PFOS converted into short-chain PFCAs	×
UV	Generation of $OH^{\bullet}$ help increase destruction of alkyl chain in PFOA	1] Wavelength dependent degradation efficiency 2] Longer contact time (24-72 h) and addition of H_2O_2 results lower mineralization [1701] 3] $OH^{\bullet}$ radical is ineffective to break C-F bond	×	Generation of $OH^{\bullet}$ help increase destruction of alkyl chain in PFOA	1] Wavelength dependent degradation efficiency 2] Lower mineralization 3] $OH^{\bullet}$ radical is ineffective to break C-F bond	×

×: Ineffective treatment method, *: Effective treatment, UF: ultrafiltration, w/o: without, POU: point of use, UV: ultraviolet

7.2. Treatment of PFAS from landfill leachate

Landfill leachate comprises dissolved organic matter, xenobiotic organic compounds, heavy metals, and inorganic substances. The concentration of these components depends on waste sources, waste age, and weather conditions (Wei et al., 2019). For instance, humid regions having precipitation higher than 75 cm y^{-1} showed a gradual decline in the PFOA and PFOS concentration in the leachate. Quantified concentrations of these contaminants decide the nature of the treatment process. The prominent sources of PFAS in landfills are the disposal of consumer products (e.g., PFAS coated non-stick vessels, food contact materials, carpets, popcorn bags, water repellent clothing) and industrial wastes (Busch et al., 2010; Benskin et al., 2012a). PFAS commonly detected in the concentration ranging from ng L^{-1} to µg L^{-1}. Also, the presence of a complex matrix develops an analytical challenge (Wei et al., 2019). PFCAs are identified as a more prevalent compound than PFSAs. The hydrophilic and mobile nature of short-chain PFAS made their abundance in landfill leachate. The short-chain PFAS are usually intermediate compounds of long-chain PFAS (Busch et al., 2010).

The standard method for treating PFAS from the leachate is not yet established. On the other hand, the recycling industry of developed countries like the US (e.g., Michigan Waste and Recycling Association) continuously monitors the concentrations of PFAS in the leachate (Michigan Recycling Association, 2019). The effect of different leachate disposal methods (e.g., direct sanitary sewer discharge, pump-and-haul for discharge, and deep well injection) towards the distribution of PFAS into the different environments has been studied. Reverse osmosis has been used as a treatment method for removing PFAS from the leachate, and the treated water was discharged into surface water. From the report of the Michigan Waste and recycling association, direct sanitary discharge is the major disposal method (47%), followed by pump and haul to centralized water treatment (26%) (Michigan Recycling Association, 2019).

Additionally, they found that the influent concentration of leachate entering the leachate treatment facility is lower than Michigan's conservative surface water criteria. Therefore, it has been concluded that the contribution of leachate was lesser than the other sources, such as industrial and domestic discharges. It has been envisioned that, for lower PFAS concentrations, RO is an effective treatment method. US EPA has initiated a project entitled "A system-based approach to understanding the role of management strategies and treatment methods on the occurrence, source, and fate of PFAS in landfills"(EPA, 2021). Accordingly, foam fractionation has been identified as an efficient pre-concentration for PFAS leached from municipal solid waste landfills (Robey et al., 2020). It is 69% effective in removing PFOA and PFOS, while it was less effective for removing short-chain PFAS (i.e., PFAS alternatives). However, further treatment of concentrated foam via separation or destruction technologies was not carried out. It has been leftover as a future research scope.

Meanwhile, the State of Florida Department of Environmental Protection (FDEP) and GW Cleanup Target Levels (GCTLs) set up the guideline value for the discharge of WW/leachate into surface water as 70 ng L^{-1} (combined concentration of PFOA and PFOS) (Stuchal and Roberts, 2019). From this aspect, energy incentive destruction technologies (e.g., electron beam, plasma, incineration, oxidation, and sonication) are suggested as an optional method to treat the concentrate of foam fractionation. Nonetheless, the destruction of PFAS was affected by different parameters (e.g., C-F bond length and physico-


Landfill leachate comprises
dissolved organic matter, xenobiotic
organic compounds, heavy metals,
and inorganic substances.
The concentration of these components
depends on waste sources, waste age,
and weather conditions.

chemical properties). In Australia, the Airport soil (1,000 ton) contaminated with PFAS was treated with a specific commercial adsorbent called RemBind and yielded >98% of PFAS removal and <0.2 µg L^{-1} of PFAS in the leachate (RemBind, 2009). Developing the protocols for treating the PFAS from the leachate is still in the initial phase.

7.3. Remediation of PFAS contaminated soil

7.3.1. Stabilization

Stabilization is a fixation process where PFAS contaminated soil is mixed with stabilizing agents (e.g., activated carbon) to mitigate the environmental risk and leaching tendency of PFAS from the impacted soil to the surroundings. *In-situ and ex-situ* soil stabilization methods have been adopted to stop the spread of PFAS contamination to GW and surface water. Technically, *in-situ* soil mixing can treat vadose and saturated zone. Ex-situ soil mixing helps prevent PFAS leakage to surroundings. The main disadvantage of this technique is its inability to destruct the PFAS.

Electrostatic and hydrophobic interactions are the two fundamental mechanisms of soil stabilization. Generally, carbon materials, aluminum hydroxide, kaolin, and polymeric composites are used as stabilizing agents. The efficiency of the soil stabilization varies depending on the mixing methods, target PFAS (e.g., short-chain or long-chain or both), geology, and soil characteristics. For example, long-chain PFAS shows higher adsorption on activated carbon than short-chain PFAS (Xiao et al., 2017b). Aluminum hydroxide possesses a positively charged surface (pKa of 9.1) and exhibits a wide operational window (pH 3-11) for stabilizing the PFAS (Richard et al., 2016). Soil characteristics (e.g., high carbon content, metals, minerals, and low soil pH) affect the sorption efficiency of carbonaceous adsorbents. On the other hand, aluminum-based adsorbents are known to yield higher sorption efficiency even in the above-mentioned interfering parameters. Therefore, combining activated carbon with aluminum-based adsorbents helps to increase the removal of PFAS (NGWA, 2017; Li et al., 2018). The composite of activated carbon, aluminum hydroxide, and clay was tested on a lab scale to treat PFAS-impacted soil collected from Australia, Europe, and the US (Szabo et al., 2017). A large-scale *ex-situ* stabilization project was established in an Australian airport in 2015 to mitigate approximately 1,000 ton of PFAS-contaminated soil (Stewart, 2017). A similar lab-scale analysis was performed to test the efficacy of the carbon-based composites to immobilize 28 PFAS from the soil samples collected from 14 different PFAS-impacted sites across Australia. The study observed 95-99% of PFAS removal (Stewart and McFarland, 2017). The application of carbon-based stabilizers also reduced the accumulation of PFAS in earthworms and plants (Kempisty et al., 2018). Edinburgh management activities of the Australian Government treated 10,000 ton of PFAS-impacted soil from Royal Australian Air Force (RAAF) Base (Australian Government, 2019). The treatment unit showed 90 and 98% removal of PFAS from clayey and sandy soil, respectively.

The primary consideration of the stabilization method is establishing proper sampling, reactor design for lab-scale investigations, and treatability tests to ensure the stabilization of the PFAS-impacted soil. In all cases, PFAS-impacted soils were collected and tested in the lab using different adsorbents, and their leaching efficiency was monitored through US EPA methods 1311, 1312, 1320, and ASTM D5084. After successful lab-scale investigations, field studies were carried out. The sustainability of this method is related to the adsorbent characteristics, equipment, and transportation of PFAS-impacted soil. This technology is considered viable and cost-effective for treating military bases and government-owned PFAS-impacted sites (Hou et al., 2016).

Cementitious material (e.g., Portland cement) is a suitable material for improving the encapsulation property of stabilizing agents. In this regard, the stability of the Portland cement under harsh environmental conditions (e.g., alkaline pH) needs further investigation. Solidification technologies require regular monitoring of PFAS. Also, incorporating a composite made up of Portland cement, fly ash, and furnace base slag reduced the leaching rate of the PFAS (Sörengård et al., 2019).

7.3.2. Excavation and disposal

Excavation is a technology of transporting contaminated soil from on-site to permit off-site. The mandatory prerequisite of this approach is designing the proper leachate management procedure. In this regard, sorption, stabilization, and construction of slurry walls have been used as leachate management options. Excavation and disposal were proven technology in early 1990. However, the risk associated with the migration of PFAS from the contaminated soil to the surrounding areas limited their application in the following years (Lang et al., 2017). The thermal treatment or stability approach mandatorily was used to prevent secondary pollution of PFAS. Economically, excavation is not a feasible method to treat PFAS-impacted soil. Therefore, it is not the best treatment option.

7.3.3. Incineration

Incineration is a proven technology to treat PFAS-impacted soils, and it is the most applied methodology at the field scale (USEPA, 2020c). High temperature is used to treat PFAS-contaminated soil. The resulting vapors are collected by precipitation or wet scrubbing methods. No prerequisite experimental or reactor design is required to treat PFAS-contaminated soil. Proper management of fume hood vapors and safe disposal of treated soils has lesser sustainability impacts.

Figure 14. Portable VEG-based ex-site PFAS-contaminated soil remediation unit (Endpoint Consulting,

The Vapor Energy Generator (VEG) process uses high temperature (e.g., 1100ºC) ex-situ treatment (Endpoint Consulting, 2016) (Figure 14). Compared to the larger incineration unit, the portability, small operating footprint, and operational cost made the VEG technology to treat PFAS-contaminated soil popular. VEG treats 200 m^3 of the soil every day. The field-scale demonstration of VEG carried out by Endpoint Consulting with the collaboration of the Colorado School of Mines showed more than 99% removal of PFAS within 30 min. The efficacy of this technology toward short-chain and PFAS precursors is unknown (Endpoint Consulting, 2016).

Lab-scale low temperature (350 to 400ºC) ex-situ thermal desorption studies demonstrated 99.9% removal of PFAS (Crownover et al., 2019). However, information about the by-products and economic and environmental constraints are unavailable. The researchers have not yet well-understood the fate and atmospheric distribution of PFAS escaping from the incineration unit. Additionally, the generated hydrogen fluoride, hydrofluoric acid, and other fluorinated by-products chemically pose adverse health and safety-related issues. Therefore, proper protocol is needed to manage the exhaust of the incineration unit.

In summary, excavation and disposal of the PFAS-impacted soil in a landfill is a traditional treatment method used to remediate PFAS contaminated soil. However, considering the long-term hazardous nature of PFAS, on-site incineration prior to the landfill is recommended. Accordingly, many countries used high-thermal incineration for the soil containing ≥50 mg g^{-1} (ATP3 1342/2014). Capping the boundaries of the landfills using engineered materials also reduce the leachability of the PFAS. In highly contaminated sites like fire-fighting training facilities, soil washing is carried out to concentrate the PFAS. The concentrated liquid phase requires additional treatment, such as adsorption or degradation. On the other hand, stabilization is the best method for the washed PFAS-contaminated soil. Considering the treatment options for PFAS contaminated soil, excavation, ex-situ thermal treatment, and soil stabilization are feasible and technically established treatment options. For treating very high concentrations of PFAS (>10,000 µg L^{-1}), soil washing, or incineration is required before adopting excavation procedures.

8. REGULATIONS OF PFAS

8.1. US EPA policies

US EPA continuously develops policies and regulations to control the risks associated with the PFAS. In this regard, EPA developed the Toxic Substances Control Act (TSCA), the Safe Drinking Water Act (SDWA), and the Comprehensive Environmental Response, Compensation, And Liability Act (CERCLA).

TSCA authorizes EPA to issue regulations related to the required testing, control measures, and health and safety exposure to protect the public and environment from PFAS related components. In this regard, Significant New Use Rules (SNURs) have been developed to limit the use of a chemical identified with toxic nature. Four SNURs covering 271 PFAS were developed from 2002-2013 (US EPA, 2017a). Among those, the first three SNURs covered the voluntary phase-out of the PFOS by 3M Corporation. In 2013, SNURs informed stakeholders to notify the import of seven PFAS to the US. They also required notification from the manufacturer regarding the composition of long-chain PFCAs and the salts/ raw materials in the carpet production. The SNURs also allowed using a low concentration of some PFAS in semiconductor, metal plating, and photographic fields. According to the 2015 January SNUR, notification to US EPA is required to use PFOA and PFOA-related chemicals (US EPA, 2015). US EPA revised the SNURs related to FP dispersions containing PFOA and its salts in 2016. Based on this modification, notification is required to use PFOA-related products. The final notice of the US EPA related to the supplemental SNURs was published in July 2020, and it designates the new import and use rules for Long-Chain Perfluoro Carboxylates (LCPFAC) (US EPA, 2018c). Overall, these SNURs warrant a compulsory notification from manufacturers and stakeholders to produce, import, and use PFAS and PFAS-related chemicals.

SDWA (section 1412) requires the EPA to notify the public regarding the list of contaminants known or anticipated to occur in the public water systems. In such line, PFOA and PFOS have been included in the Contaminant Candidate List (USEPA, 2018a). Primarily, EPA included six PFAS (e.g., PFOA, PFOS, PFBS, PFNA, and PFHpA) under the third Unregulated Contaminant Monitoring Rule (UCMR). The role of UCMR is to collect the data on pollutants in drinking water suspected to develop negative impacts on

public health. The data is collected at regular intervals and reviewed periodically. From this cumulative information, US EPA released lifetime Health Advisories for PFOA and PFOS, which protects the general population from long-term exposure to PFOA and PFOS. SDWA develops non-enforceable and non-regulatory technical information to state authorities and the general public regarding the health effects, analytical methodologies, and treatment options related to PFAS contamination in drinking water (USEPA, 2016b, a). However, section 1431(a) of SDWA is designed to take immediate action for agencies where contaminants are likely to enter the public water sources (e.g., drinking water and GW) that cause substantial endangerment to public health.

CERCLA authorizes the federal government to respond to releasing hazardous substances, pollutants, and contaminants that are known to cause risk to the public (US EPA, 2020b). Specifically, section 104 (e) provides the right to the federal government to inspect the site to determine whether the release of hazardous substances is present. If there is a release, the authority has a right to decide on the cleanup. For example, PFOA and PFOS have been denoted as hazardous substances under CERCLA. Then the manufacturer will be asked to pay the recovery cost of these substances in the affected site and address the public affected by these contaminations.

Apart from these fundamental regulations, EPA has not yet finalized the regulatory standards for all PFAS. Subsequently, the SDWA act informs the use of lower limits for all unregulated PFAS as a guideline value. On the other hand, UCMR 5 is developing novel and accurate detection methods for several PFAS. CERCLA released a certified detection method for detecting PFAS from oil matrices (USEPA, 2021a). Resource Conservation and Recovery Act (RCRA) has set a primary remediation goal of 70 ppt and an interim recommendation of 40 ppt PFOA and PFOS (USEPA, 2020b). However, the cleanup guidelines under this act are not yet fully established. The Clean Air Act (CAA) develops guidelines and

regulations to control air-born PFAS pollution. Nonetheless, the absence of technical data related to the non-cancer risk of PFAS restricts the development of standardized guideline values for PFAS. CAA also focuses on developing standardized analytical methods for PFAS in the air (USEPA, 2020b). In addition to the existing frameworks, a project named "Safeguarding America's Waters, Air, and Land-PFAS 2021-2025" has been introduced, and an initial recommendation from the general public is under collection (USEPA, 2021b). The main goal of this project is to maximize funding and financial aid to the under-served and vulnerable communities affected by PFAS pollution. The President's job plan released a fund of USD 10 billion to monitor and remediate PFAS in drinking water (The White House Briefing Room, 2021).

ATSDR has framed oral Minimum Risk Levels (MRLs) for PFOA, PFOS, PFHxS, and PFNA as 3×10^{-6}, 2×10^{-6}, 2×10^{-6}, and 3×10^{-6} mg kg^{-1} day^{-1}, respectively. These MRL values have been provided based on the Low-est Observable Adverse Effect Levels (LOAEL) and No Observable Adverse Effect Levels (NOAEL) calculat-ed via laboratory and animal studies (ATSDR, 2018b). The main reason for the lack of toxic assessment of PFAS is their different toxicological endpoints. For example, PFOA and PFOS have developmental, im-munological, hepatic, and cancer endpoints, whereas PFHxS have hepatic and thyroid endpoints (Wolf et al., 2008; Viberg et al., 2013). The endpoints of PFNA are related to body weight and developmental stage (Fang et al., 2009; Wolf et al., 2010). This lack of published results on the toxicity of PFAS delays the framing of proper regulations for PFAS.

8.2. Guidelines of US EPA

US EPA is continuously regulating PFAS in different environmental media. Accordingly, 70 ng L^{-1} is used as an individual Lifetime Health Advisory (LHA) for PFOA and PFOS (US EPA, 2016c). However, different states have developed different guideline values for PFOA and PFOS. For example, the Maine directive (DEC, 2016) used 400 ng L^{-1} as an individual advisory GW cleanup level for PFOA and PFOS. Likewise, New Jersey uses 14 and 13 ng L^{-1} as a maximum contaminant level for PFOA and PFOS. The amended, proposed, and recommended guideline values for PFAS by different states of the US are summarized in Table 8.

Table 8. State-level guidelines for regulating different PFAS in GW, drinking water, and soil

(Melissa Byroade et al., 2020)

S. No	State (US)	Category	Guideline values PFAS	Guideline values ng L^{-1}	Remarks	References
1	California	Drinking water (Notification) Drinking water (Response level)	PFOA PFOS PFOA PFOS	5.1 6.5 10 40		(California Water Boards Division of Drinking Water, 2019)
2	Connecticut	Drinking water (Standard)	PFOA PFOS PFNA PFHxS PFHpA	70 70 70 70 70	Individually and combined	(Connecticut Department of Public Health, 2018)
3	Maine	GW (Standard for residential) Recommended drinking water standard Soil standards (Leaching to GW)	PFOA PFOS PFBS PFAS PFOA PFOS PFBS	400 400 4.00E+05 70 9.5 µg kg^{-1} 21 µg kg^{-1} 7,100 µg kg^{-1}	Individually and combined	(Maine Bureau of Remediation, 2018)
4	Massachusetts	Drinking water standard	PFOA PFOS PFNA PFHxS PFHpA PFDA	20 20 20 20 20 20	Individually and combined	(Massachusetts Department of Environmental Protection, 2021)
5	Michigan	proposed drinking water standard	PFOA PFOS PFNA PFHxA PFHxS PFBS GenX	8 16 6 4,00,000 51 420 370		(Michigan Department of Environment, 2019)
6	Minnesota	Drinking water Standard	PFOA PFOS PFHxS PFBS PFBA	35 15 47 2 7		(Minnesota Department of Health, 2019)
7	New Jersey	Adopted drinking water standard Proposed drinking water standard	PFNA PFOA PFOS	13 14 13		(NJAC, 2019) (Protection, 2019)
9	New York	Proposed drinking water standard	PFOA PFOS	10 10		(NRDC, 2019)
10	North Carolina	Drinking water standard Recommended GW standard	GenX PFAS	140 70	Individually and combined	(North Carolina Department of Environmental Quality, 2017)
11	Ohio	Proposed drinking water standard	PFOA PFOS GenX PFBS PFHxS PFNA	70 70 700 1,40,000 140 21	Individually and combined	(Agency, 2018)

S. No	State (US)	Category	Guideline values PFAS	Guideline values ng L^{-1}	Remarks	References
12	Texas	GW cleanup standards	PFOA PFOS PFBS PFHxS PFHpA PFNA PFDA PFBA PFPtA PFOSA PFDoA PFTA PFTrDA PFUnA PFDS	290 560 34,000 93 560 290 370 71,000 290 93 93 93 93 93 93		(AWWA, 2020)
13	Vermont	Drinking water standard	PFOA PFOS PFHxS PFHpA PFNA	20 20 20 20 20	Individually and combined	(Vermont Environmental Health Division, 2020)
14	Washington	Drinking water standards	PFOA PFOS PFNA PFHxS PFBS	10 15 14 70 1,300		(Washington Department of Health, 2019)
15	Wisconsin	Proposed GW standards Enforcement standard Prevention action limit	PFAS PFAS	20 2	Individually and combined	(Wisconsin Department of Health, 2020)
16	Alaska	GW standard	PFOA PFOS	70 70	Individually and combined	(Alaska Department of Environmental Conservation, 2019)
17	Montana	GW standard	PFOA PFOS	70 70	Individually and combined	(Montana Department of Environmental Quality, 2019)

ATSDR released a report on the toxicological profile of PFAS on human health in June 2018. In this context, the minimal risk level of PFOA, PFOS, PFHxS, and PFNA in adults and children was framed (Table 9) (ATSDR, 2018a). The US EPA released the PFAS action plan in 2019 and included PFOA, PFOS, GenX,

Table 9. Minimal risk levels of different PFAS regulated under ATSDR (ATSDR, 2018a)

S. No	PFAS	Minimal Risk Levels (MRL) (ng L^{-1})	
		Adult	Children
1	PFOA	78	21
2	PFOS	52	14
3	PFHxS	517	140
4	PFNA	78	21

PFNA, and PFHxS under the Toxics Release Inventory (TRI) (Nessa Coppinger, 2020). Most recently, EPA released a preliminary report on the Fourth Drinking Water Contaminant Candidate List to produce guidelines for other PFAS. However, no binding (common) federal regulations have been made to regulate PFAS in the US.

8.3. Regulations, policies, and guideline values implemented in other countries

The timeline taken by the EPA to draft the regulations for PFAS is considered as long. The reason is associated with the lesser number of evidence in the toxicological profile of all PFAS. This type of inevitable delay results in the amendment of different regulations in other countries. EU developed PFAS chemical legislation under the scheme name of REACH and actively participated in developing guidelines and regulations for long-chain and short-chain PFAS. Regulation EC 1935/2004 envisions the use of PFAS in food contact materials; it provides information on the use of allowed PFAS composition in food contact materials. EC 98/83 includes a limit value for a sum of 20 individual PFAS as 0.1 µg L^{-1} and 0.5 µg L^{-1} as the limit value for total PFAS (Kwiatkowski et al., 2020). GW Directive 2006/118/EEC measures the substances that cause potential risk to humans. Accordingly, PFAS has been added to the Ground Water Watch List (GWWL) and is monitored by 11 states in the EU. Based on the developed dataset, GW Directive amended GW regulatory guidelines for some of the PFAS (Dauchy et al., 2019). Water Framework Directive (2000/60/EC) uses Environmental Quality Standards (EQS) to see the chemicals that destroy the quality of surface water (Laitinen et al., 2014). PFOS and its derivatives have been categorized under EQS, and regular monitoring has been done. EC 1881/2006 allows calculating the Tolerable Daily Intake (TDI) for PFOA, PFOS, and its salts/derivatives. Based on this monitoring EU directive released a risk assessment for some PFOS, PFOA, PFNA, and PFHxS (Young et al., 2021). Moreover, the EU government also framed different directives such as industrial emission (2010/75/EU), European Pollutant Release and Transfer Register (E-PRTR) (USEPA, 2018b), water (APHI, 2021), land, and soil legislations.

The proposed maximum PFAS level in drinking water by the German Ministry of Health is 300 ng L^{-1} for PFOA and PFOS (Wilhelm et al., 2010). This guideline value is proposed by evaluating the toxicology of PFOA and PFOS in all types of populations. Italian National Health Institute proposed less than 30 ng L^{-1} and less than 500 ng L^{-1} as guideline values for PFOA and PFOS (Mastrantonio et al., 2018). Additionally, EPA guidelines of 70 ng L^{-1} for PFOA and PFOS are amended in the places where high concentrations of PFAS are found. The Spanish government often monitors the concentrations of PFAS in different locations. Nonetheless, no specific guidelines have been proposed for PFAS.

Canada has taken a step to develop drinking water screening value ng L^{-1} for PFNA (0.02), Perfluorovaleric acid (PFPeA) (0.2), PFHxA (0.2), PFHpA (0.2), PFHxS (0.6), PFBS (15), and PFBA (30) (Abunada et al., 2020). Also, the government is actively participating in the new guideline development for another regulated PFAS (NCCEH, 2019). The Department of Health, Food standards Australia developed drinking water guidelines for PFOA and PFOS as 560 and 70 ng L^{-1} (Australian-Government, 2019). It also created a guideline value for recreational water quality for PFOA and PFOS as 10,000 and 2,000 ng L^{-1}. These two guidelines propose a TDI value of 160 and 20 ng L^{-1} for PFOA and PFOS (Australian-Government, 2019). Moreover, the intergovernmental agreement is introduced by the Australian government. Through this agreement, Australian citizens have the right to advise new threshold values for PFAS based on the health effects.

8.4. Limitations and recommendations to current EPA's regulation or policies for PFAS

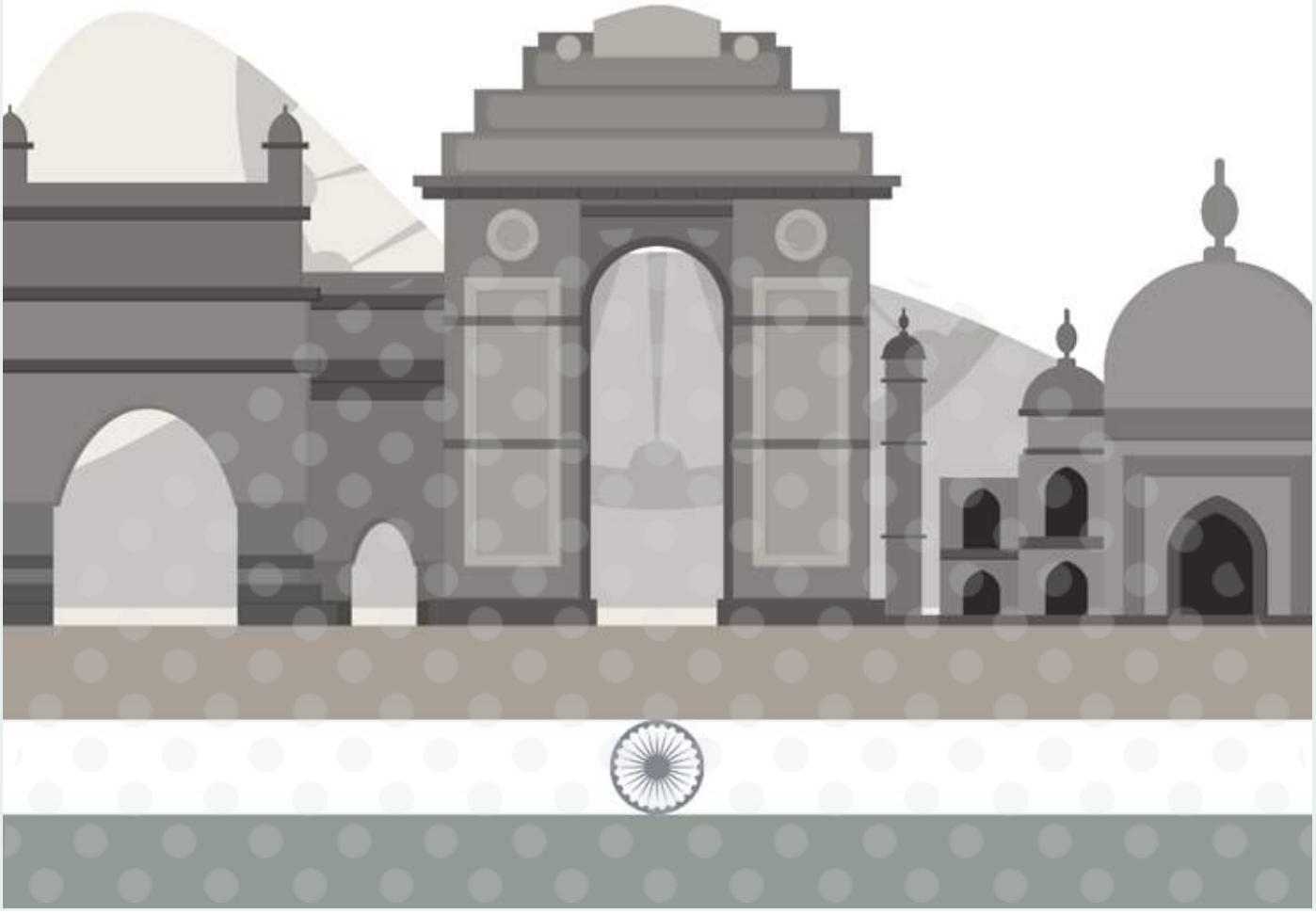
Though EPA has put forth diverse efforts to reduce PFAS pollution and protect the general public, several loopholes need to be fixed in the EPA's action plans (Langenbach and Wilson, 2021).

- Effective and standardized methods are required to analyze complete subclasses of PFAS: EPA's present action plans focus on the PFOA and PFOS. However, several thousands of subclasses of PFAS used in the market. Therefore, it is essential to frame standardized protocol to quantify a broad spectrum of PFAS rather than focusing on two of the majorly used PFAS.
- The existing method limit of detection 40 ppt is higher considering the current national health advisory guideline value of 70 ppt. Considering the RCRA action plan using a lower value for the method detection limit may help reduce the health impacts of PFAS.
- Although PFAS considered an air pollutant and is verified through several air trajectory models (e.g., Community Multiscale Air Quality (CMAQ) modeling system), no guidelines or standardized methods were published by EPA. In this regard, other agencies, such as Occupational Safety and Health Administration (OSHA) should help frame air sampling methods and inhalation exposure dosages.
- Generally, the absence of frequent monitoring and a lesser number of sample collections largely miscalls the concentration of PFAS in the study area. Therefore, temporal and seasonal water sampling is required.
- It is necessary to include more PFAS under the SDWA. This way, further accumulation and spread of PFAS-related components in the environment can be minimized.
- Generalized method to identify the PFAS hotspots is lacking. Also, some of the developed treatment options in the field scale are uneconomical. Therefore, resilience is needed to frame guidelines for detection, monitoring, and remediation of PFAS from the PFAS contaminated site.
- EPA has not focused on the adverse effects of short-chain PFAS, commonly known as GenX or PFAS alternatives. These alternatives are identified as more mobile (soluble) than long-chain PFAS, and these pollutants are considered less toxic than PFAS. However, till now, EPA has not yet released any statement or study related to these short-chain alternatives.
- In most cases, the analytical method is used to quantify the individual or a set of PFAS. But it is not mentioned clearly in the results, which leads to analytical bias related to the concentration or pollution caused by individual PFAS or a group of PFAS.

8.5. Regulations of PFAS in India

India has set the first step to regulate PFOA and PFOS by adopting the standard methods used for sampling and analysis. Accordingly, the BIS announced three methods for sampling and measuring PFOA and PFOS (BIU, 2020). The methods are,

* ISO 3696:1987: Water for analytical laboratory use– Specification and test methods
* ISO 5667-1 Water quality sampling- Part 1: Guidance on the design of sampling programs and sampling techniques
* ISO 8466-1:1990 Water quality– Calibration and evaluation of analytical methods and estimation of performance characteristics– Part 1: Statistical evaluation of the linear calibration function.
* Apart from these methods, no phase-out or guidelines have been developed by the Indian government.



8.6. Recommendations

- *PFAS are omnipresent pollutants that are commonly used in most industrial and consumer-related products. As the toxicity of PFAS is well-known from the existing epidemiological studies, high-quality toxicological and epidemiological studies are to be conducted among the Indian population.*
- *The association between onset chronic diseases (e.g., anemia, inflammation, and carcinogenicity) and women/children need to be assessed.*
- *Periodic biomonitoring programs on the environment (ecological) and humans need to be framed to develop valuable evidence for population-related advocacy policies.*
- *Public and political awareness programs related to a behavioral change at the individual and social level to restrict the use of PFAS-containing products need to be initiated. This way, one can reduce the persistent exposure to PFAS.*
- *On the industrial side, transparent general data sharing on the use of substituent chemicals for PFAS should be practiced.*
- *Periodic sampling of the sites that use PFAS-containing products should be analyzed to understand the environmental risk quotient associated with effluent discharge. More specially, the "dark water" scenario should not be repeated in India.*
- *Stringent guidelines for regulating the PFAS and PFAS-related products should be prepared by referring to the aforesaid periodic monitoring datasheet. In this way, the health and ecological risk can be minimized.*
- *Inventory for PFAS-related concentrations in different consumer products should be investigated through accredited laboratories. Based on the concentration-related toxicity, a product warning label should be prepared and used on the product to enhance public awareness.*
- *At the treatment level, the efficacies of the existing water and WW treatment processes should be evaluated. As activated carbon-based adsorbents provide higher adsorption capacity for a wide range of PFAS, a protocol for adopting the activated carbon technology for industries should be developed.*
- *Overall, fortunately, the concentrations of PFAS in humans and the environment are at a safer level as of now. However, their persistence has already ringed an alarm to design a safeguard system to minimize PFAS contamination in day-to-day activities. Also, it is an obligatory duty to preserve the environment and the public from this hazardous and toxic pollutant. Lastly, the chances of preventing PFAS pollution are available, and the government should make an immediate effort to control the PFAS pollution to preserve the incredible environment and public.*

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Annexure- II

Item No.05 Court No. 1

BEFORE THE NATIONAL GREEN TRIBUNAL
PRINCIPAL BENCH, NEW DELHI

Original Application No. 548/2024

News item titled "IIT Madras study reveals presence of ,forever chemicals, in Chennai lakes drinking water" appearing in The Hindu dated 07.04.2024

Date of hearing: 17.05.2024

CORAM: HON'BLE MR. JUSTICE PRAKASH SHRIVASTAVA, CHAIRPERSON
HON'BLE MR. JUSTICE ARUN KUMAR TYAGI, JUDICIAL MEMBER
HON'BLE DR. A. SENTHIL VEL, EXPERT MEMBER

ORDER

1. This original application is registered *suo-motu* on the basis of the news item titled "IIT Madras study reveals presence of ,forever chemicals, in Chennai lakes drinking water" appearing in The Hindu dated 07.04.2024.

2. The matter relates to the presence of 'forever chemicals' i.e. pre and polyfluroalkyl substances (PFAs) in Buckingham Canal, Adyar river and Chembarambakkam lake in Chennai (Tamil Nadu) as per a recent study conducted by IIT Madras.

3. The news item explains that PFAs are a vast family of synthetic chemicals found in everyday products such as non stick cookware, upholstery, industrial material, food packaging etc. These chemicals do not degrade easily in the environment. They are persistent in the environment and could lead to adverse health effects such as liver damage, low infant birth weights, hormonal imbalance, fertility issues, immune system effects and even cancer.

4. The news item further alleges that the findings, published in *Environmental Sciences Europe*, revealed high levels of PFAS in groundwater samples from Chennai. While groundwater samples were collected from in and around the Perungudi dumpsite, surface level samples were taken from different points along the Buckingham Canal, Adyar river, and Chembarambakkam lake. Samples from a water treatment plant (WTP) near the lake were also tested. All the samples were quantified for eight target PFAS.

5. As per the news item, it was found that the concentrations of all eight target PFAS was increased in the treated water of the water treatment plant, compared to the raw water. The target PFAS' concentration ranged in varying quantities up to 136.274 ng/L in the groundwater; 59. 838 ng/L in Adyar river; 60.174 ng/L in Buckingham canal, up to 33.316 ng/L n Chembarambakkam lake, and up to 23.952 ng/L in the raw water of the water treatment plant. (as compared to the United States Environmental Protection Agency's health advisory levels in drinking water, the measured amounts of PFOA (77.61 ng/L) and L-PFOS (8.12 ng/L) are each around 19,400 times and 400 times higher. The advisory limits are 0.004 ng/L and 0.02 ng/L.

6. The news item states that Industrial emissions, untreated domestic wastewater discharge, and open dump sites have been suspected to be significant sources of contamination, highlighting the need for further investigation to fully assess the extent of PFAS contamination in Chennai

7. The above matter indicates violation of the Water (Prevention and Control of Pollution) Act, 1974 and the Environment Protection Act, 1986.

8. The news item raises substantial issue relating to compliance of the environmental norms and implementation of the provisions of scheduled enactment.

9. Power of the Tribunal to take up the matter *suo-motu* has been recognized by the Hon'ble Supreme Court in the matter of *"Municipal Corporation of Greater Mumbai vs. Ankita Sinha & Ors." reported in 2021 SCC Online SC 897*.

10. Thought a study has been conducted in Adyar River and Chembarambakkam lake in Chennai but the same situation may be prevailing in other stream/water body also. CPCB is required to consider the study result and inform the parameters which have been prescribed and the remedial action which is proposed.

11. Hence, we implead the following as respondents in the matter:

(i). Member Secretary, Central Pollution Control Board, its, Parivesh Bhawan, East Arjun Nagar, Delhi-110032.

(ii). Member Secretary, Tamil Nadu Pollution Control Board, 76 Mount Salai, Guindy, Chennai – 600032.

(iii). Collector/District Magistrate Chennai, Fourth Floor, M. Singaravelar Maaligai, 62 Rajaji Salai, Chennai, Collectorate, Chennai - 600001

12. Let notice be issued to the above respondents for filing their response at least one week before the next date of hearing.

13. List on 03.09.2024

Prakash Shrivastava, CP

Arun Kumar Tyagi, JM

Dr. A. Senthil Vel, EM

May 17, 2024
OA No. 548/2024
HB