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PER AND POLYFLUOROALKYL SUBSTANCES (PFAS) IN THE ENVIRONMENT: A REVIEW

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ABSTRACT

Per and Polyfluoroalkyl substances (PFAS), a group of more than 7,800 synthetic heterogeneous organic compounds with different physicochemical properties, are chemically and thermally stable. Since 1945, these compounds have been used for innovative developments in the field of material sciences to provide numerous benefits to society. These compounds have oil and water-resistant properties and are used as refrigerants and fire suppressors, in the textile industry, paint industry, electronic industry, cookware industry etc.

This review documents the concentration of the most commonly used Per and Polyfluoroalkyl substances in the aquatic environment, milk samples, soil, plants, vegetables, fruits, fish and their impact on humans

Due to overuse and misuse of these compounds in different industries, these persistent pollutants are present in all the compartments of the environment, i.e. air, surface water, groundwater, river water, marine water, drinking water, soil, animal and breast milk, food chain, vegetables, fruits, and fish. In the 21st Century, these chemicals, along with antibiotics, are considered the most harmful persistent organic pollutants. The wastewater generated by households, industries, and armed force areas is a hotspot of the residual PFAS. Human health is adversely affected by consuming PFAS-contaminated plant produce and animal food and by drinking PFAS-contaminated water. On accumulation in humans, it affects the immune system, alters the lipid metabolism, endocrine activity, thyroid gland and mammary gland functioning.

Keywords: Per and Polyfluoroalkyl substances, Human, Environment, Milk, Vegetables, Fruits, Fish, Soil.

1. INTRODUCTION

Per- and polyfluoroalkyl substances (PFAS) are synthetic man-made organic compounds, that were accidently first formed in the DuPont company labin 1938. Since 1945, Dupont has been using this compound (Teflon) for coating cookware to make it non-stick cookware. These chemicals have numerous applications [1-4] used in adhesives, fire fighting foams, cosmetics, electronic items, stain and water repellent agents in the textile industry, resulting in the commercial use of these chemicals from the 1950s. Due to the synthesis of PFAS, innovative development in material chemistry for societal benefits became possible. Since then, these organic compounds have been prominent players in the betterment of human life and society. As per the US Environmental Protection Agency report [5],

approximately 7,800 PFSA structures have been synthesized, having different physical and chemical properties. The useful major applications of PFAS for modern society are given in Table 1.

Due to the presence of many C-F bonds, which is the strongest covalent bond in per and Polyfluoroalkyl substances (PFAS), the PFAS becomes thermally stable, chemically inert, and possesses hydrophobic and oleophobic properties [6]. Due to their chemical stability, they persist in the environment for a longer period of time and have high mobility [7]. These compounds are found even in remote areas like the Arctic and Antarctic [8, 9] and are also called "forever chemicals." When these compounds are uptaken by biota via the food chain are very easily bioaccumulated and biomagnified [10, 11]. The residues of per and polyfluoroalkyl substances and

their metabolites are reported in all the compartments of the environment, viz., air, groundwater, surface water, marine water, drinking water, soil, food chain, fruits, vegetables, cereals, grains, animal feeds, carpets and upholstery; cosmetics and personal care products, food wrappers and carry-out containers. Several research studies [12-15] have found the residues of PFAS in human blood, urine, breast milk, tissues, organs and in aquatic flora and fauna. According to the finding of food scientists [16,17], the presence of PFAs in the environment adversely affects the terrestrial ecosystem. The soil is an integral part of the environment, and the ecosystem is the natural source for plant growth and food production. As the protein part of the soil organic matter acts as a sorbent for PFAS [18], the soil acts as a sink/reservoir for per and polyfluoroalkyl substances

[19]. In low and middle-income countries due to water scarcity, untreated wastewater is used by farmers for agricultural usage, which cause the accumulation of these pollutants in soils. Accumulation of these undesirable organic chemicals in soils beyond their normal concentrations not only deteriorate the soil quality but also has impact on human health, food quality, and social development [20, 21], as water-soluble PFAS pollutants are transported to surface water and/or leach to groundwater resulting accumulation in plants (via roots) and animals [22].

This work aims to provide the most up-to-date review of the PFAS applications to humans, including the amount of these persistent pollutants in various environmental compartments and their impact on humans.

Table 1: Applications of Per and Polyfluoroalkyl substances (PFAS) in different industries/for humankind

Industry	Application of PFAS
Aerospace	Corrosion protection of Phosphate ester-based brake and hydraulic fluids by altering the electrical potential at the metal surface. Used as Lubricant and elastomeric seals in Turbine-engine, Jet engine/satellite instruments. Protects wire and cables from high-temperature endurance and makes them high-stress crack resistant. Used in the propellant system as fuels and oxidizers and as aerosol propellant. PFAS are coated to protect from atomic oxygen effects.
Air-Conditioning/ Refrigerators	The Working fluid of the air conditioners and compressors contains PFAS.
Ammunition	PFAs makes them shockproof to prevent unplanned explosions and enable the ammunition for long-term storage without decay.
Automotive Industry	PFAS are used in weather resistance paint; improve the resistance of the polish to water and oil, prevent icing on the windshield in the cold countries, as sealants and bearings, engine coolants. Used in cylinder head coatings and hoses to reduce gasoline vapour emission resulting in enhanced fuel efficiency, cable and wires, andbrake pad additives.
Biotechnology	PFAS helps in cell cultivation by the enhanced supply of oxygen and other gases to microbial cells and PFAS microporous membranes prevent bacterial growth.
Building and Construction	Used in roofs of houses and Greenhouses- for protection from weathering; acts as dirt repellant and used for light. As cement additive to retard cement shrinkage. Protects wire and cables, gasket and hoses from high-temperature endurance and makes them high-stress crack resistant
Chemical Industry	Used in the production of chlorine and NaOH; in the processing of Tantalum, molybdenum, and niobium; acts as an aid in the processing of high- and linear low-density polyethene film and Fluoropolymer. These compounds act as a medium for crosslinking of resins, elastomers and adhesives and provide inert media for gaseous reactants.
Cleaning	As PFAS provides stain resistance and repels soil is used as a dry-cleaning liquid for cleaning the carpet and upholstery, is also used as a cleaning fluid for adhesives.
Effluent water treatment	Filter membranes used in the treatment plants are composed of polymeric PFAS.
Electronic Industry	PFAS acts as an inert fluid and is used for the testing electronic devices and equipment. Used for cooling of electronic and electrical equipment. These compounds dissolve the unwanted compounds deposited during the manufacturing of hard disk drives, helping in -Etching of

2. CLASSIFICATION OF PER AND POLY-FLUOROALKYL SUBSTANCES (PFAS)

The number of PFAS compounds ranges from 5600 to 7800 with different physical and chemical properties, they may be gases, liquids, solids or high molecular weight polymers, which can be classified as discussed below.

2.1. Classification based on the physical and chemical properties

2.1.1. Nonpolymers PFSA

The non polymers PFSA is classified into two classes

2.1.1.1. Perfluoroalkyl substances

These are the compounds in which all the hydrogens of the alkane are replaced by fluorine. These molecules have more than two carbon atoms with a charged functional group at one end of the chain. Their structure can be denoted as $C_nF_{2n+1}R$ where, R is a functional group that may be carboxylate, sulphonate or sulphonamides. A few examples are Perfluoroalkyl carboxylic acids, perfluoro alkane sulphonic acids and perfluoro alkane sulphonamides.

The perfluorinated alkyl substances are further divided into: (a) Perfluoroalkyl acids (PFAAs)- PFAAs includes (i) Perfluoroalkyl carboxylic acids (PFCAs) (Where R is COOH) for example perfluorooctanoic acid; (ii) Perfluoroalkyl carboxylate (where R is COO) e.g. perfluorooctanoate (iii) Perfluoroalkane sulphonic acid (PFSAs) (where R is SO_3H) e.g. perfluorooctane sulphonic acid (iv) Perfluoroalkane sulphonates (PFSAs) (where R is SO_3) example is perfluorooctane sulphonate (v) Perfluoroalkyl phosphonic acid (PFPAs) (where R is H_2PO_3) e.g. perfluoropentane phosphonic acid.

(b) Perfluoroalkyl sulphonamides (FASAs) e.g. perfluorooctane sulphonamide (where R is $-SO_2NH_2$).

(c) Perfluoroalkyl ether acids (PFEAs) (where R is O-COOH) e.g. perfluoro-2-methoxyaceticacid.

(d) Perfluoroalkyl aldehyde (PFALs) (where R is -CHO) for example perfluorooctanal

The most studied perfluoroalkyl substances are perfluorooctane carboxylate (PFOA) [(CF₃-(CF₂)₅-CF₂-COO^T] and perfluorooctane sulphonate (PFOS) [(CF₃- (CF_2) , $-CF_2$ -SO₃)].

Sources of the PFAS in the Environment

2.1.1.2. Polyfluoroalkyl substances

These are the compounds in which all the hydrogens are not replaced by fluorine, they have one or more nonfluorine atoms. Non-fluorine atom are usually H or O attached to at least one carbon atom in the tail. Generally, these compounds are named with n:x prefix where n denotes the number of fully fluorinated carbon atoms and x denotes the number of non-fully fluorinated carbon atom(s). Polyfluorinated PFAS can be divided in to the following classes:

- (a) *Fluorotelomer Compounds:* These are those polyfluoroalkyl substances that are produced by the fluorotelomerization process. This class includes- (i) Fluorotelomer alcohols (FTOH): $[(CF₃-(CF₂)₇ -$ CH₂- CH₂OH)] (ii) Fluorotelomer sulfonic acids (FTSA) (iii) Fluorotelomer carboxylic acids (FTCA).
- (b) *Perfluoroalkane sulfonamido compounds:* These compounds contain a fully fluorinated carbon chain with one or more $CH₂$ group(s) attached to the sulfonamido tail, e.g. n-ethyl perfluorooctanesulphamido ethanol, $[(CF_3-(CF_2)_7-CH_2-SO_2N(C_2H_5))$ $-CH_2-CH_2OH$]. This class includes (i) perfluoroalkane sulphamido ethanols (FASEs) (ii) perfluoroalkane sulphamido acetic acid (FASAAs) (iii) N-alkyl perfluoroalkane sulphonamides (N-alkyl FASAs).

2.1.2. Polymer PFSA

These are large molecules formed by combining many of the same or identical small molecules called monomers**.** The polymeric PFAS can be divided into following:

2.1.2.1. Fluoropolymers

These are the compounds in which most of the hydrogen atoms attached to the carbon of the monomer are replaced by fluorine atoms. Common examples are Poly tetra fluoro ethylene (PTFE), polyvinylidene fluoride (PVDF).

2.1.2.2. Side chain fluorinated polymers

These compounds contain the non-fluorinated carbon chains with a side chain of the poly/perfluoroalkylic group, e.g. Fluorinated urethans, Fluorinated acrylates.

2.1.2.3. Perfluoropolyethers (PFPE)

These are the compounds in which carbon atoms are attached with oxygen besides fluorine, e.g. Perfluoropolyether-benzophenone (PFPE-BP).

2.2. Classification based on the number of carbon atoms in the chain

2.2.1. Long-chain PFAS

These are compounds with six or more fully fluorinated

carbon atoms e.g. PFSAs, PFCAs.

2.2.2. Short-chain PFAS

Those PFSAs which have six or fewer carbon atoms are called short-chain PFAS, e.g. perfluorobutanoic acid, perfluorobutane sulphonic acid.

3. ENVIRONMENTAL SOURCES OF PER AND POLYFLUOROALKYL SUBSTANCES (PFAS)

Per and Polyfluoroalkyl substances are oil and water repellent, chemical resistant and are used in the firefighting foams, textiles, electrical, non-sticking metal coatings, laundry and cleaning industries, painting, printing and paper industries, oil extraction and mining, medical devices, pharmaceuticals, pesticides, skin creams, cosmetics, photography, chrome painting etc. The short-chain PFAS are highly mobile and are more stable and are easily accumulated in the environment [23], while long-chain PFAS are easily accumulated in humans, animals, soils and sediments. The major sources of exposure to the citizenry are public water systems, drinking water wells, surface water of lakes, ponds, groundwater, food packaging, food items sold in the market, fish, indoor dust from carpets, textiles, etc.

4. ROUTES OF CONTAMINATION

4.1. Human intake of the Per and Polyfluoroalkyl substances (PFAS)

Humans are exposed to Per and Polyfluoroalkyl substances (PFAS) either by direct exposure or indirect exposure [24, 25].

- (i) *Ingestion (direct exposure):* It occurs via the gastrointestinal route, i.e. by up-taking contaminated food and other feeds, drinking contaminated water, milk and other drinks via the mouth.
- (ii) *Dermal/via permeable membrane (Indirect exposure):* Dermal uptake means absorption through the skin. Skin creams and cosmetics are a few examples of dermal uptake.
- (iii) *Inhalation (Indirect exposure):* Inhalation uptake occurs via inhalation of the polluted air as dust fumes or contaminated vapours.
- (iv) Cleaning products, car polishing, wood, stone and floor are the other routes of human uptake of PFAS [26].

Per and Polyfluoroalkyl substances (PFAS) present in sediments are bioaccumulated in the small aquatic organisms present in the sediments. These toxic compounds become available to wildlife animals and humans who eat these aquatic organisms as food.

5. PFAS IN THE AQUATIC ENVIRONMENT

Manufacturing industrial discharges, domestic wastewater discharges, sewage sludge, sewage water, effluent from landfills and air emissions are the major entry sources of these persistent pollutants in to the aquatic environment. The concentration of these compounds in the wastewater depends on the source of effluent, the physic-chemical properties of the pollutant (i.e. the number of carbon atoms in the chain, the functional group attached) and water solubility as most of the PFAS are persistent i.e. they cannot be evaporated. These pollutants enter to surface water, river water, groundwater, drinking water, marine water and agricultural fields via wastewater [27, 28]. According to literature review, the total concentration of the PFAS in wastewater ranges from 0.0ng/L to 143ug/L, in river water ranges from a few ng/L to 496 ug/L, in surface water up to 84 ug/L and in drinking water ranges up to 8300 ng/L. The concentration of the different PFAS in water samples is given in Table 2.

6. PFAS IN THE PLANTS AND VEGETABLES/ CROPS

The major constituents of the human diet are vegetables, fruits and cereals (i.e., field crops) and meat, milk, and eggs i.e., animal origin food. Globally, due to changes in dietary habits, to reduce malnutrition, population explosion, and prosperity, the demand for vegetables and fruits has been increasing since the beginning of the $21st$ century. The use of treated or untreated wastewater for irrigation (the survey of literature denotes that approximately 20 million hectares of land are irrigated by raw wastewater or partially treated wastewater contaminated by PFAS), amendment of soils with sewage sludge or paper-fibre biosolids, and atmospheric deposition are the main sources of soil contamination by PFAS. From soil, these PFAS are transported to plants via roots. Globally, 10% of the world population consumes food grown on the wastewater irrigated or manure amended soils. The accumulation of the different PFAS compounds depends on number of the carbon atoms in the chain of the pollutant and the amount accumulated in soil and plant type. The accumulation of PFAS in the vegetation parts

of plants was greater than in reproductive and storage parts. The accumulation of PFAs in the different parts of plants were in the order of the leaves $>$ stem/shoot $>$ root > fruit. Accumulation of PFAS in plants damages cell structure, and organelle functions, perturbs photosynthesis, protein synthesis, carbon and nitrogen metabolism, gene expression, etc. [29].

The accumulation of different PFAS in different parts of plants, vegetables and crops is documented in Tables 2 and 3.

7. PFAS IN FISH AND OTHER AQUATIC ORGANISMS

Food and Agriculture Organization (FAO), with 195 members, a specific agency of the UN, was established in 1945 to eliminate hunger and malnutrition. The worldwide population is increasing and is expected to be 9.1 billion by 2050. As per the FAO report, since the last decade, the number of undernourished and malnourished people in developing countries is increasing. Despite the innovative techniques, agricultural production has not been able to solve this problem. Fish and other aquatic animals are the food source for billions of people across the world. It is a general belief that fish is a healthy source of nutrients, protein, omega-3 fatty acids, vitamin D, calcium, vitamin B complex, vitamin A, iron, zinc, essential fatty acids, micronutrients, and lysine. Due to poverty, deficiency in agricultural food production, and to cope with the malnutrition, the consumption of fish and other aquatic animals is continuously increasing. The global consumption per capita has increased from 9 kg in 2013 to 21 kg per capita in 2021 and is expected to reach 40 kg per capita by 2050. The increasing concentration of PFAS in the aquatic medium affects fish and other aquatic animals. As PFAS is associates with protein-rich tissues, these organic pollutants persist for a longer period of time and are bioaccumulated in the gills, liver, lungs, muscles, and intestines of fish via protein, organic matter, and the food they consume [30]. Bioaccumulation of these pollutants (PFAS) in fish organs causes metabolism disturbance, reproduction disruption, oxidative stress, developmental toxicity, thyroid disruption, nuclear receptor activation, reactive oxygen species induction, interaction with the membrane etc. [31]. The concentrations of different PFAS in fish and other aquatic animals are given in Table 4.

Table 4: Total concentration of PFAS in Different Species of Fish

8. PFAS IN THE MILK

Per capita milk consumption throughout the world (i.e., developed and developing countries) is increasing, which may be due to economic prosperity, population explosion, and changes in dietary habits. India, the top producer globally produces approximately 18% of the global milk production. Several researchers [32-34] have reported residues of PFAs in breast milk and animal milk. In the USA, more than 80% of breast milk is contaminated with PFAS. PFAS binds to the β lactoglobulin protein present in the milk. Exposure of PFAS to infants via breastmilk affects immunity, hormonal function, development of the nervous system, vaccine response, kidney function, obesity, asthma etc. The data on the concentration of PFAS in milk is recorded in Table 2.

9. PFAS IN SOIL

Soil is the most important natural source on earth as it supports the production of the human food system, fodder for animals, fibre etc. Healthy soil is essential for human life and their well-being. Franklin D. Roosevelt correctly interpreted that "*The nation that destroys its soil destroys itself*." But for the last 60 years, due to anthropogenic activities, the health of the soil has been deteriorating. Soil is an important reservoir of pollutants, e.g. potentially toxic metals, pesticides, antibiotics, PFAS etc.

PFAS are released in agricultural soils through irrigation water, biosolids applications, AFFFs discharge, and atmospheric dispersion [35, 36]. The PFAS pollutants are either sorbed by soil particles or dissolved in the soil solution. From the soils, these pollutants are either absorbed by the roots of the cultivated plants or leached into the groundwater. Many agricultural scientists [37- 39] have reported that soils globally are contaminated with PFAS. The concentration of the different PFAS compounds in soil is given in Table 2.

10. IMPACT OF PFAS ON HUMANS

The impact of these chemicals on humans depends on the route of exposure, duration of exposure and concentration of the chemical [40]. A number of studies [41-43] have reported that PFAS, mainly PFOA and PFOS, in humans affects the immune system, alter the lipid metabolism, endocrine activity, thyroid gland and mammary gland functioning. These chemicals also cause delays in the mammary gland development, obesity, increased miscarriage risk and low sperm count [44]. In their studies, Temkin [45] found that these toxic chemicals in humans increase blood cholesterol and that these chemicals are also linked to kidney diseases, testicular cancer, kidney cancer, increased uric acid

concentration in the blood, and immunity/decreased response to vaccines. Zeng *et al.,* [46] reported that PFOS in humans decreases cell activities and increases ROS levels, decreases mitrochondrial membrane potential with enhanced apoptosis and autophagy. Increase of endoplasmic reticulum stress by PFAS in humans has been reported by Louisse *et al.,* [47]. Redox imbalance and abnormal autophagy with decreased glutamatmine synthase activity by PFOS have been found by Li *et al.,* [48]. Borghese *et al.,* [49] discovered that exposing pregnant women to PFHxS for a longer period of time increases the chances of preeclampsia development by three fold. Blood glucose level and PFOS exposure had a positive significant correlation [50].

11. CONCLUSION

- PFAS are considered one of the the most toxic pollutants of the $21st$ century as they are present in all the compartments of the environment, viz., air, groundwater, surface water, river water, drinking water, house hold wastewater, soil, manure, milk, vegetables, crops fruits, and fish.
- As PFAS compounds possess the strongest covalent C-F bond, these compounds are thermally and chemically stable and possess hydrophobic and oleophobic properties. Due to their chemical stability, they persist in the environment for a longer period of time with a high mobility. These compounds are found even in remote areas like the Arctic and Antarctic and are also called "forever chemicals."
- In biosolids and manure, the concentration of these pollutants ranged from ng/g to ug/g. The concentration of these pollutants in surface water near industrial level and AFFF discharge points ranged up to mg/L. The concentration of these pollutants in soils near the AFFF discharge points ranged up to 7 mg/kg.
- In the agrosystem, PFAS are introduced via manure amendments and irrigation by contaminated water and impacts the ecological balance in soils.
- Short-chain PFAS compounds translocate very easily into plant components that are consumed by humans or animals as fodder, while long-chain PFAS compounds remain in the root part.
- As these pollutants contain functional groups such as carboxyl, sulphonyl, sulphonamide, amine groups glycosylation of the pollutants in the plants occurs.
- PFAS are bioaccumulated in the liver, muscle and fillet part of the most consumable fish (rainbow trout) worldwide.
- Ingestion of the PFAS through animal and/or plantbased foods as well as drinking of PFAScontaminated water has impact on human health. The accumulation of these pollutants in humans affects the immune system, lipid metabolism, endocrine system, and reproductive system and alters development. These pollutants also have cancerogenicity. Health impacts on humans depend on concentration, route and duration of exposure.

Future Suggestions

- The production and utilization of these pollutants must be reduced.
- Researchers must find some alternate, less toxic, more efficient compounds.
- It is a need of the hour to develop effective technology to remove these pollutants from wastewater.
- Efforts must be made to create awareness to the citizenry regarding the negative impacts of these pollutants

Conflict of interest

The authors declare no competing intrests.

Declaration

No original data has been used in this review, all information as accessed from published work.

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