Pregledni rad

Ključne reči:
ljudi; ljudska rasa; biosfera

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humans; human race; biosphere

IZAZOV U KOJEM SE NALAZIMO

THE CHALLENGE IN WHICH WE FIND OURSELVES

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Ovaj rad ima za cilj da pokaže u kakvom se haosu mi, ljudi, nalazimo – ljudska rasa i šira biosfera – sve opravdano potragom za novcem za nekolicinu. Svrha ovog rada nije da presuđuje šta je ispravno, a šta pogrešno; umesto toga, on nastoji da istakne izazove i načine kako na njih odgovoriti. Jedina vrsta kriva za ovaj haos jeste ljudska rasa, jedina vrsta koja može i mora nešto da preduzme po tom pitanju.

This paper intends to show the mess we, humans, find ourselves in—the human race and the wider biosphere—all justified by the hunt for money for the few. It is not the purpose of this paper to judge right from wrong; instead, it aims to highlight the challenges and how to respond. The only species guilty of the mess is the human race, the only species that can and must do something about it.

1. Introduction

Many news articles and scientific papers have been published over the last decade, highlighting the pollution of the biosphere. This pollution is creating a toxic blend of chemicals emitted into the atmosphere, the soil, and thereby the waterways on which we all depend [1]. No collected overview is available because manufacturers only declare the emissions of the specific chemicals they must report; the rest end up either in the atmosphere or wherever convenient and cheapest for the company, out of the eyes of the general public.

This is also what happened in Parkersburg, West Virginia. At this point, the US EPA did not exist. The DuPont Washington Works plant has had some challenges handling its waste since the 1950s. This was in the early days of producing Perfluorooctanoic acid (PFOA), internally known as C8 [2].

The C8 production in Parkersburg started around 1951, and since 1961, DuPont, now a spin-off under the brand Chemours, has known that PFOA and PFOS are toxic and are now banned in many countries. Yet it dumped waste C8 into a nearby unsafe waste dump, where overflow water from rain and waste could leak into a nearby creek leading into the Ohio River. This sounds almost like setting the scene for a criminal film, which became the topic of “Dark Waters”, which is based on the memoirs of the lawyer Robert Bilott.

The same company, DuPont, later installed a similar company near Dordrecht, The Netherlands, making the same mistake of dumping waste into the nearby river. We now know there have been pollution problems around this factory, similar to those seen in Parkersburg [3].

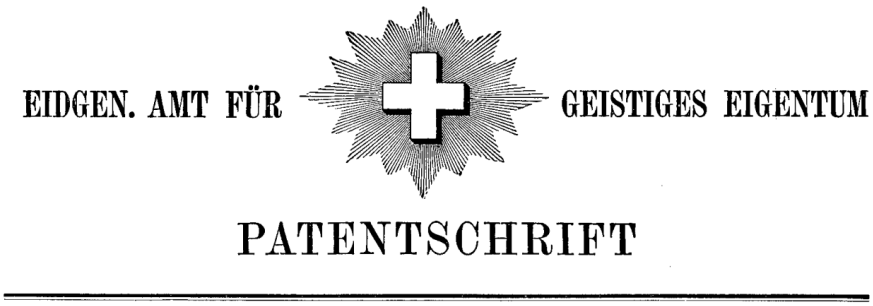
DuPont/Chemours are not the only polluters in the world. Several others produce similar products and face the same pollution challenges worldwide. Most of them – if not all – are present in

courts being held accountable for the pollution their production and products have caused to the environment and to the health of the populations around the factories. The different manufacturers are facing headwinds as they are being held accountable for the pollution caused by their production processes and products [4].

Donald R. Taves reported in 1968 that he found two forms of fluoride in human blood serum. At this time, the short notice in Nature did not mention per- or polyfluoroalkyl substances or trifluoroacetic acid. The brief note claims that fluoride in this form strengthens teeth and bones. For the same reason, fluoride is added to drinking water in many water systems worldwide. This knowledge has been the general rule, but it is now being contested and reversed in some countries [5].

However, fluorine intoxication, the title of a study by Kaj Roholm published in 1937[6], tells us that “fluorosis” was studied and taken very seriously long before Donald R. Taves performed his studies. X-ray analysis showed that the ribs in the employees' chests who had worked in a cryolite process had changed over time and came to look like marble. The workers also lost the ability to turn their heads and bend down to collect items from the floor. The latter because the joints had stiffened due to “fluorosis” of the joints.

2. The history of fluorine chemistry



EIDGEN. AMT FÜR GEISTIGES EIGENTUM

PATENTSCHRIFT

Nr. 59000 4. Januar 1912, 7 Uhr p. Klasse 108 a

HAUPTPATENT

Walter LACHMANN, Hamburg-Schulau (Deutschland).

Verfahren zur Kälteerzeugung.

Figure 1. Patent application for using Methyl Chloride (R-40)

One of the early working fluids used up to about the 1960s internationally was methyl chloride, which was very toxic and flammable.

The history of fluorine chemistry started in Belgium, where the scientist Frédéric Jean Edmond Swarts (1866-1940) was a professor at the University of Ghent. His father, Théodore Hubert François Swarts (1839-1911), was also a professor of chemistry at the same university.

One of the main works of Frédéric Jean Edmond Swarts was in the field of fluorine and developing various solutions and compounds, of which one of them later became known as Freon 12, but his work also showed how other alkanes and alkenes could be made. A little overlooked is that Frédéric Jean Edmond Swarts also found stannic fluoride (tin tetrafluoride), which is used in toothpaste today. One of Professor Swarts’s inventions was how to produce trifluoroacetic acid (TFAA) (table 1), although it caused him some trouble because it was challenging to handle the acidity and toxicity.

One of the students under Professor Swarts was a young man by the name of Albert Leon Henne (1901–1967), who, after his studies at Brussels University, where he earned his PhD in 1925 [7, 8], with a dissertation titled "The Stereoisomers of Chloroiodo-ethylene", went to the USA, where he

became a US citizen in 1933. Mr Henne later became a Professor of chemistry at the State University of Ohio, where he came in contact with Thomas Midgely (May 18, 1889 – November 2, 1944).

Thomas Midgely was later credited with, among other inventions, the invention of CFC-12 together with Albert L. Henne. Looking closer, the real achievement of the two was that they solved Professor Swarts's challenges with controlling the acidity and toxicity of the trifluoroacetic acid (TFAA) during the process. What they did not understand or wilfully forgot to mention was the potential pollution of the global environment that might follow their inventions when the products later became industrialised.

In the movie produced about a century later, Dark Waters, it became clear that DuPont and its supplier 3M started to realise these risks. In the 1960s, but the commercial interests in the use of these compounds and the money involved blurred the judgment of the company decision-makers, who decided to keep the information secret from the public and authorities for decades.

It was not until the case portrayed in the film shed light on the issue following the unnatural deaths of a herd of cows that had been drinking polluted water from a creek. It took years and a lot of time in courts and years of providing evidence of the risk before the ball started to roll. The first scientific papers began to emerge between the years 2000 and 2010 [9] and have rapidly increased ever since, researching a variety of compounds and their impact on human health and environmental consequences, but also the commercial risks taken with damages to brand name and time in various courts.

Matts Bäckström, a professor at the University of Stockholm, in the 1940s, suggested that instead of calling the fluorinated hydrocarbons Freon, they be called Swartsan in recognition and honor of Professor Swarts being the real inventor. Bäckström later gave up the idea and accepted the ASHRAE capital R.

ASHRAE was formed as the American Society of Heating, Refrigerating, and Air-Conditioning Engineers by the merger in 1959 of the American Society of Heating and Air-Conditioning Engineers (ASHAE), founded in 1894, and the American Society of Refrigerating Engineers (ASRE), founded in 1904 [10].

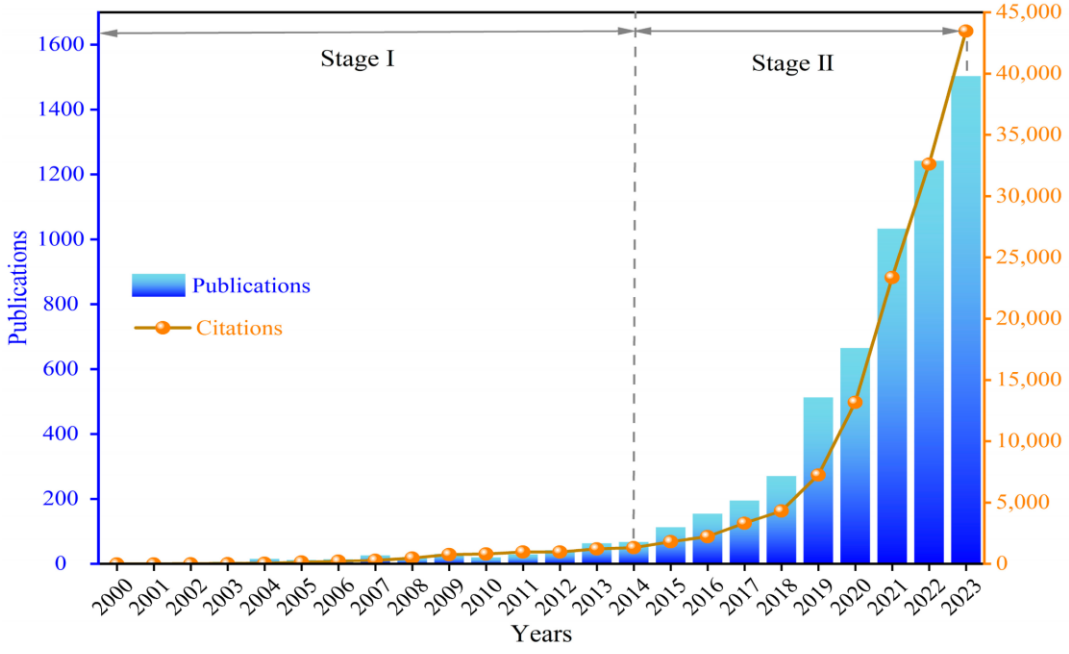


Figure 2. Number of publications and citations related to PFAS from 2000 to 2023

The number of publications covering the threats from PFAS and TFAA pollution has increased significantly from 2010 onward, as shown in Figure 2. Many and very different angles to the problems with per- & polyfluoroalkyl substances are one part of the problems humans have created, but many others are also surfacing. The pollution caused by vaccines, penicillin, and other antibiotics has been known for years. A portion of these substances is caught in wastewater cleaning systems, and then distributed on farmland, where it supposedly helps fertilise the soil. At the same time, these compounds are washed out and end up in the drinking water. In particular, antibiotics have been a worry because overexposure degrades some of their efficiency. Where should we go next if penicillin cannot be used anymore?

Table 1 The names have been clarified according to Buck et al. 's (2021) supplement. In the literature, trifluoracetic acid is often called TFA, where it should be TFAA. ***Error! Bookmark not defined.***

Perfluoroalkyl carboxylic acids (and selected salts)	FORMULA	CAS NUMBER	ACRONYM
Trifluoroacetic acid	CF_3COOH	76-05-1	TFAA
Perfluoroalkyl carboxylate anions	FORMULA	CAS NUMBER	ACRONYM
Trifluoroacetate	CF3COO-	14477-72-6	TFA

3. PFAS affects the human body

In connection with the COVID-19 pandemic, it was studied whether the PFAS could have added to the death toll seen [11]. From other studies, it is known that PFAS can affect the immune system. The study states, “Therefore, PFAS may have the potential via immunotoxicity to exacerbate COVID-19 respiratory symptoms or more generally the severity of the disease through a direct or indirect mechanism”. Another study [12] on this topic says, “Several studies have demonstrated that exposure to many metals, both toxic and essential, at both sub-chronic and chronic levels may result in immunotoxic effects, especially concerning cytokines.” The challenge is that most of the tests done in laboratories are made on mice and other animals and to convert the data to translate it into how it will affect the human body.

A review study [13] on early life exposure to PFAS and its effect on children raises a valid point in the discussion: Does it make sense to study what effects one component of an array of components has on the systems in the body, because of the variety of pollution levels with the individual components? It is still poorly understood how the components affect the body as a group and how they affect each other.

4. The inconvenient truth

In the year 2006, Al Gore, former Vice-President of the United States and presidential candidate, published a book identifying the planetary emergency of global warming. Both the CFCs, HCFCs, and HFCs are potent global warming gases. What Mr. Gore does not consider are the breakdown products these gases also produce, especially the unsaturated hydrofluoroalkenes branded as HFO, to distinguish them from the HCFs, which they are. The inconvenient truth is that ozone depletion and global warming are not the only emergency planetary challenges humans have introduced to the planet and the biosphere.

“Olefins: Acyclic and cyclic hydrocarbons having one or more carbon-carbon double bonds, apart from the formal ones in aromatic compounds. The class olefins subsumes alkenes and cycloalkenes and the corresponding polyenes [14] and “Olefin, compound made up of hydrogen and carbon that contains one or more pairs of carbon atoms linked by a double bond. Olefins are examples of unsaturated hydrocarbons (compounds that contain **only** hydrogen and carbon and at least one double or triple bond).

The use of hydrofluoroolefins (HFO) is therefore a marketing invention. Buck et al. (2011) [15] show that HFO-1234ze, CASRN 29118-24-9, should be included in the definition of PFAS, see figure 3. The hydrofluoroolefins have a very short atmospheric lifetime and therefore also have a very low GWP; however, they are known to break into trifluoroacetic acid TFAA, which is the shortest chain of perfluorinated carboxylic acid (PFCA) and the perfluorinated carboxylates. TFAA is one of millions of compounds categorised as per- and polyfluoroalkyl substances (PFAS). Industry has convinced the US administration that TFAA should be excluded from the PFAS definition, while the OECD includes it [16].

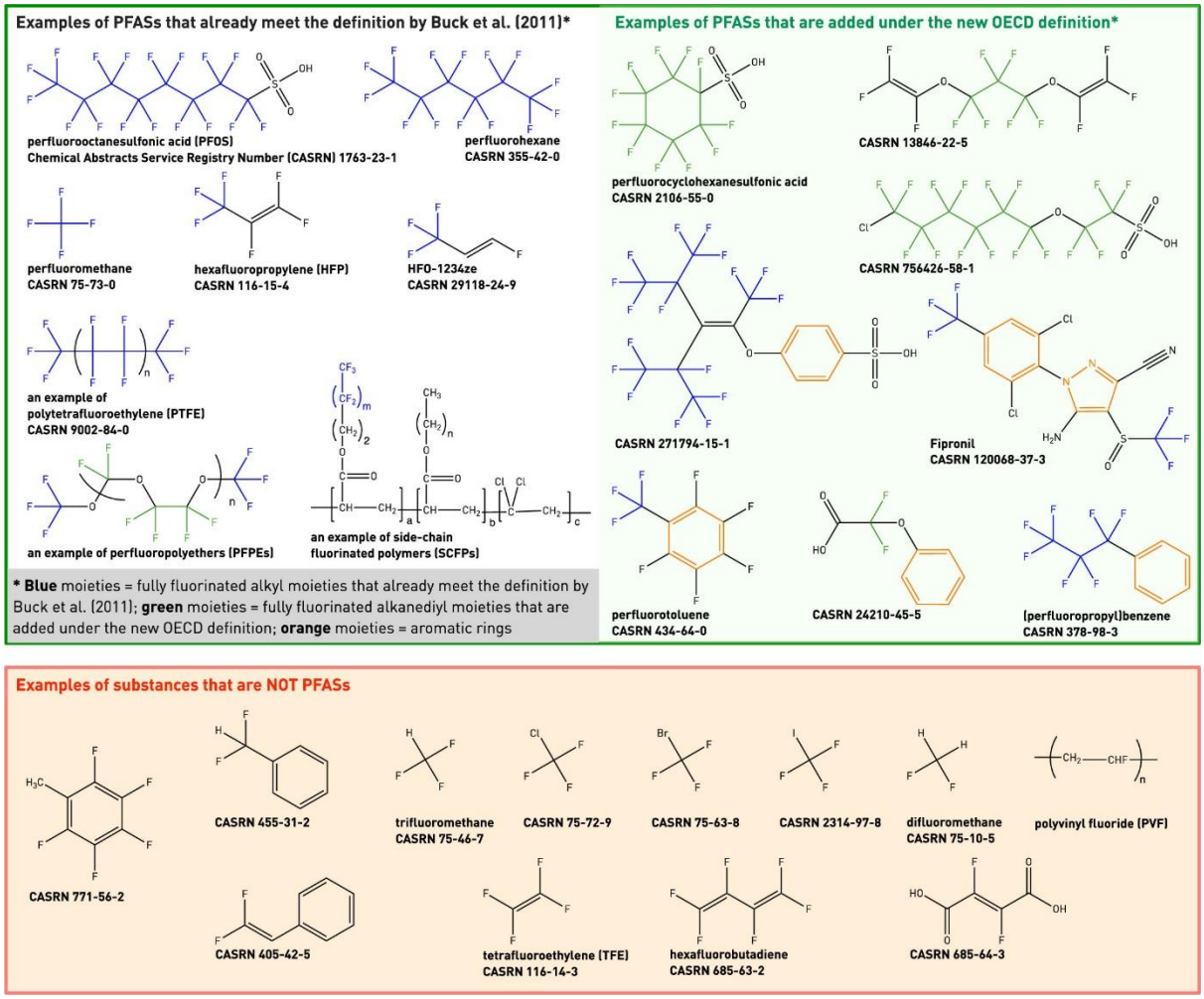


Figure 3. Definitions and examples of substances that are not covered by the OECD definition of TFA [17]

An interesting OECD list of high-production-volume chemicals [18], which is defined as products produced in levels above 1000 tons per year in at least one member country, on point 151, we find 76-05-1 trifluoracetic acid [19]. This tells us that TFAA, often called TFAA, is not only a breakdown product originating from refrigerants, but also an industrial product. TFAA is a precursor in producing fluorinated compounds, e.g., pesticides, refrigerants, adhesives and sealants, and solvents.

However, TFAA is also a degradation product from several gases and other compounds ending up in the environment and thereby also in the waterways and food of any kind humans consume, with no discrimination between meat eaters or vegans. The degradation products from PFAS are in the rain, dust, both in and outdoor air, in the soil, and in the groundwater. The sources are a long array of products from cosmetics, pharmaceuticals, medicals, clothes, carpets, furniture, electronics like mobile phones and television, and many others, and we can go on.

In a paper [20], we see how new substitutes of previous compounds turn out to be a bigger problem than the compounds they substitute. It concludes in short that **GenX** exhibited the lowest removal (79%) and defluorination (33%) after 7 hours of photocatalysis compared to **PFOA** (100% removal, 69% defluorination) and **PFOS** (100% removal, 51% defluorination). The degradation rate of GenX was significantly lower than that of PFOA and PFOS. GenX produced more ultra-short chain transformation products that accumulated in the water phase due to poor adsorption on the photocatalyst. The degradation plateau for GenX was reached after 2 hours, while PFOA took 4-7 hours, and PFOS was still degrading after 7 hours. The lower degradation and defluorination of GenX are attributed to its poor adsorption on the catalyst and steric hindrance. The short-chain products referred to here are trifluoroacetic acid (TFAA). See figure 3.

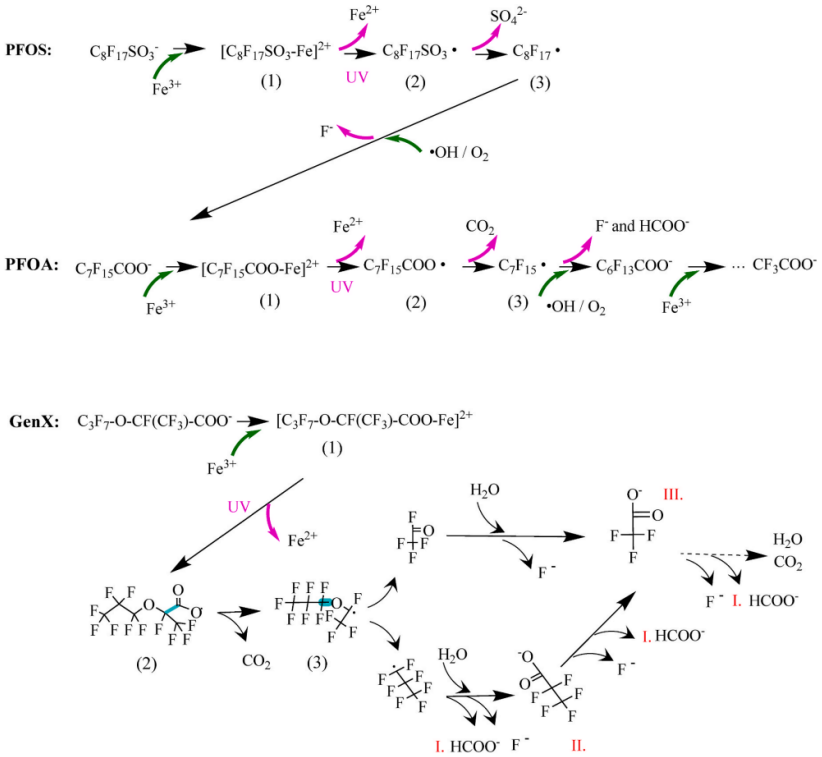


Figure 4. The degradation pathway of PFOA, PFOS, and GenX

RIWA-Meuse, the Association of River Water Works along the Meuse, has published an annual report explaining what tests the association is testing for, and the methods used [21]. For diabetes patients, Metformin and Guanylurea, in Table 1, are indicators of the number of persons with diabetes 2 in the area since Guanylurea is a reaction product of Metformin. The report from 2023 [22] includes more information about the individual points.

The table tells a story about the number of water pollutants to watch. Here, the PFAS group is not specified, but a group of the 22 most common in the area is often considered and added to make the combined level. The question in many cases is the lack of scientific evidence and limits before disruption of the human endocrine system, vaccine immune system, etc., of the more than 14000

commonly used products in modern society. We know that there are more than 7 million and counting PFAS substances around [23], so are we just waiting for the worst to happen before banning them all?

Table 2. RIWA-Meuse has organised the groups of compounds tested for in three groups

Industrial counds and consumer products	Pharmaceuticals and endocrine disrupting chemicals (EDC's)
PFAS*	Lithium
Dibromomethane sulfonic acid	Valsartanic acid
Dichloromehtane sulfonic acid	Valsartan
Dibromoacetic acid	Metformin
Tribromomethane	Guanylurea
Trichloroacetic acid (TCA)	2-hydroxibuprofen
Hexa(methoxymethyl)melamine (HMMM)	Candesartan
Melamine	Hydrochlorothiazide
8-hydroxypenillic acid	Lamotrigine
Ethylenediaminetetraacetic acid (EDTA)	Tramadol
1,4 Dioxane	N-formyl-4-aminoantipyrine
Trifluoroacetic acid (TFA)	Bisphenol A
Methenamine/urotropine	Ketoprofen
Cyanuric acid	Naproxen
Diethylenetriaminepentaacetic acid (DTPA)	Vigabatrin
Nitriloacetic acid (NTA)	**Pesticides, biocides and their metabolites**
Sulfamic acid	Chlorate
Totyltriazole	S-metolachlor
Bromate	Metolachlor-ESA
	Metolachlor-OA
	Aminomethylphosphonic acid (AMPA)
	Glyphosate
	Prosulfocarb
* PFAS is a group of substances	Propamocarb

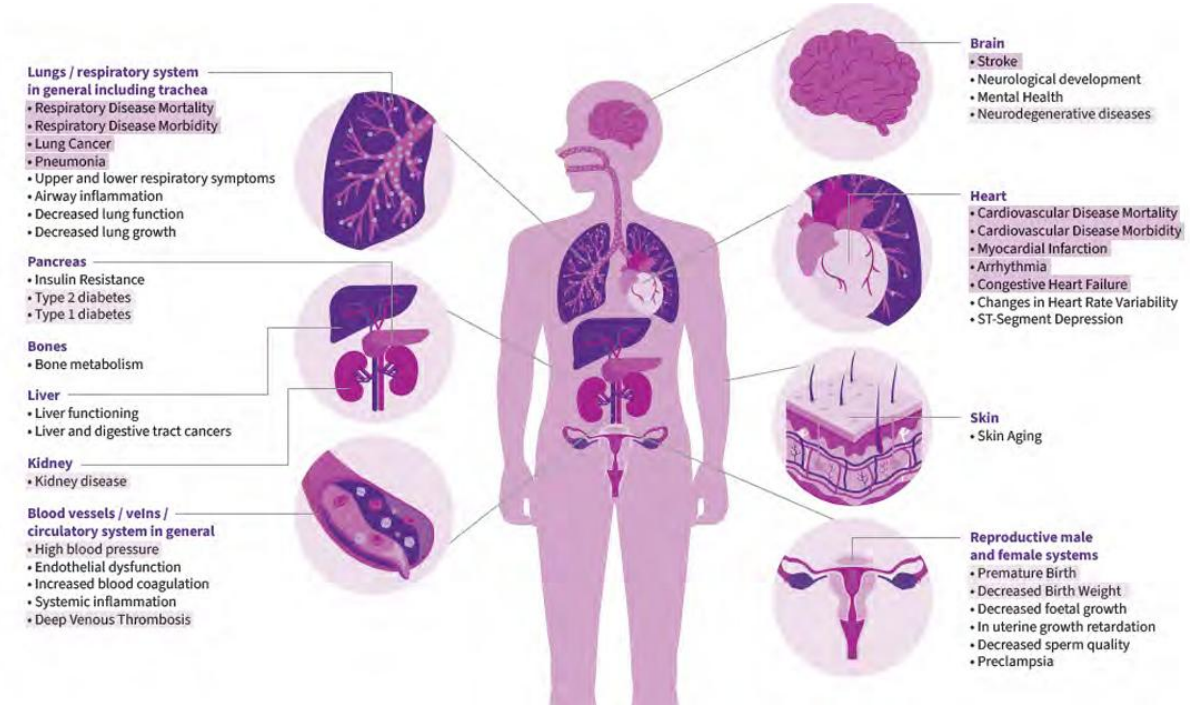


Figure 5. Organs and systems affected by air pollution – a list of diseases

5. Air pollution

"Air pollution is responsible for 6.7 million premature deaths every year" [24], says WHO. Of these, 3.2 million die prematurely from domestic air pollution. Most people live where air quality exceeds the WHO's recommendations [25]. The main topics are the nanoparticles known as $PM_{2.5}$ and PM_{10}, which both enter the body when humans breathe. The $PM_{2.5}$ and smaller particles can enter the bloodstream, where they can react with the cells.

Human exposure to per- and polyfluoroalkyl substances (PFAS) is ubiquitous [26]. Prenatal and early-life exposures to PFAS have been consistently associated with adverse health effects in children, primarily metabolic and immune system disorders, yet we still know little about the underlying mechanisms that might explain these associations [27, 28].

Plastic nanoparticles can enter the cells in the brain where they will stay and potentially be a precursor for Parkinson's disease [29, 30]. Nanoplastics and PFAS are proven to disrupt biomolecular structures and function [31]. Fluorinated containers can react to become perfluoroalkyl carboxylic acids [32]. World Economic Forum suggests rethinking the future of plastic by organising recycling, like it is being done with bottles in many regions of the world – an effective after-use plastics economy [33]. Currently, only about 9,5% in 2022 [34, 35] out of 400 million tons of plastics were recycled, and 50% ends up in landfills, 22% evades waste management and ends up in the environment, contributing to the pollution in oceans and ecosystems. One of the reasons for the difficulties of recycling is the different substances used in the plastics to give it different properties and colours. Once mixed, it is not possible to separate them again, it seems.

A Danish expert group under the Danish EPA has published a report looking at the TFAA problem. One of the tasks was to identify the source of the substance. There are two primary sources in Denmark: the fluorinated gases and the fluorinated pesticides used in agriculture. Apart from the pesticides, there are different sources for TFAA emissions, e.g., spray cans, anaesthetics (e.g., halothane and isoflurane), burning of fluoropolymers, and other perfluoro substances. Many of the emissions end up in the water when it rains.

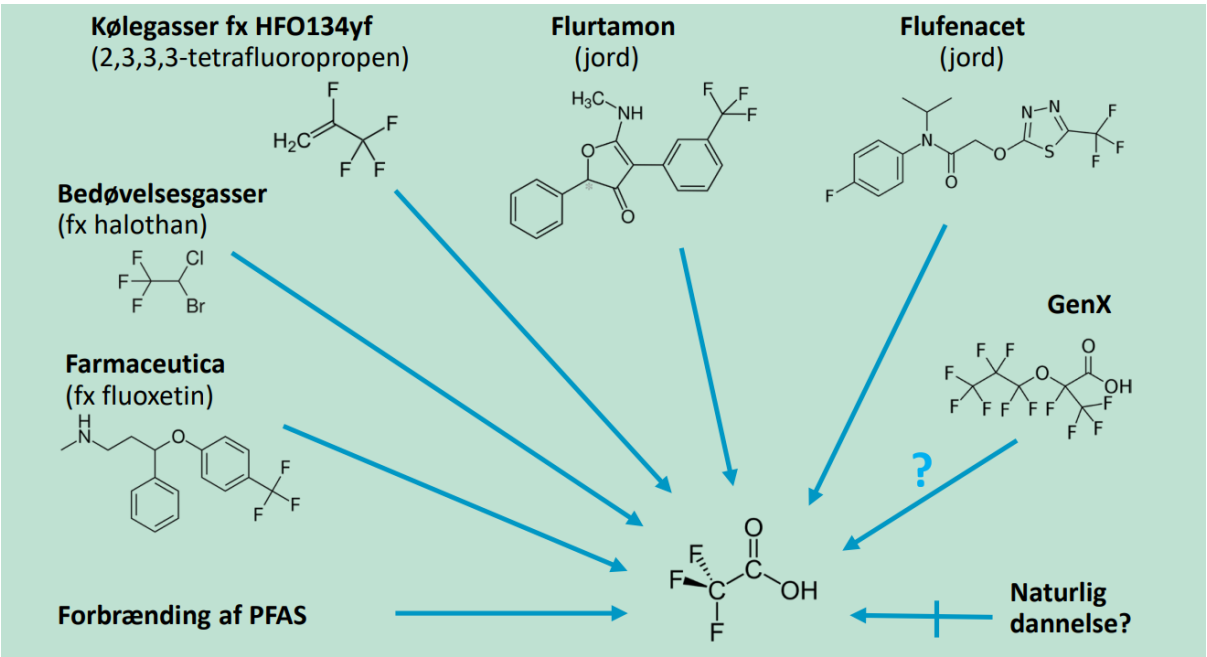


Figure 6. Among the sources, the Danish EPA has found that the refrigerants are the primary contributor to the TFAA emissions [36]

The Danish EPA expert group writes, “In terms of percentage, TFA-related PFAS account for the largest accumulation in society (36%) and are also the most significant contributor to emissions to the air and water compared to PFAAs (including precursors) and PFAS polymers. The input in the TFA category comes primarily from F-gases (hydrofluorocarbons, HFCs, including hydro-olefins, HFOs), which account for 73%, and from pesticides, which contribute 26%. F-gases are almost exclusively responsible for the significant emissions to air (200 t/year), whereas pesticides are estimated to contribute most to emissions into soil (172 t/year). Atmospheric deposition over land is estimated at 10.3 t/year (almost exclusively TFA) [37].”

As seen from Figure 6, the report concludes that the natural formation of TFAA does not happen based on available scientific literature studies. Research in the broader source of pollution with TFAA has shown that the single most significant source of TFAA emissions is HFO. However, the numbers differ from country to country, depending on whether fluorinated gases are produced in the area and by industries. Climatic conditions and dominating wind directions are spreading the gas emissions and pollution. Countries with intensive farmland activities tend to have more TFAA coming from pesticides than others.

A portion of PFAS is released into the environment during production and throughout the life cycle of manufactured goods, leading to contamination of food and water, including the drinking water and food we consume. The same extraordinary stability and durability that make PFAS valuable to consumers and in industrial manufacturing now pose a significant threat to the environment and human health. “The stability of C–F bonds renders PFAS resistant to degradation by both biotic and abiotic processes, leading to their gradual accumulation in humans over time, which can lead to chronic health effects such as cancer, abnormal immune system function, hormonal disruption, kidney and liver diseases, and adverse reproductive and developmental outcomes [38].”

6. Conclusions

Humans created fluorinated hydrocarbons, and humans have enjoyed the benefits of these compounds without knowing the risks of using them. The industry kept its knowledge to itself for decades, and the truth was only revealed after a lawyer had to investigate after a herd of cows died from poisoning. The presence of the natural formation of TFAA has been laid to rest and is not worth mentioning again.

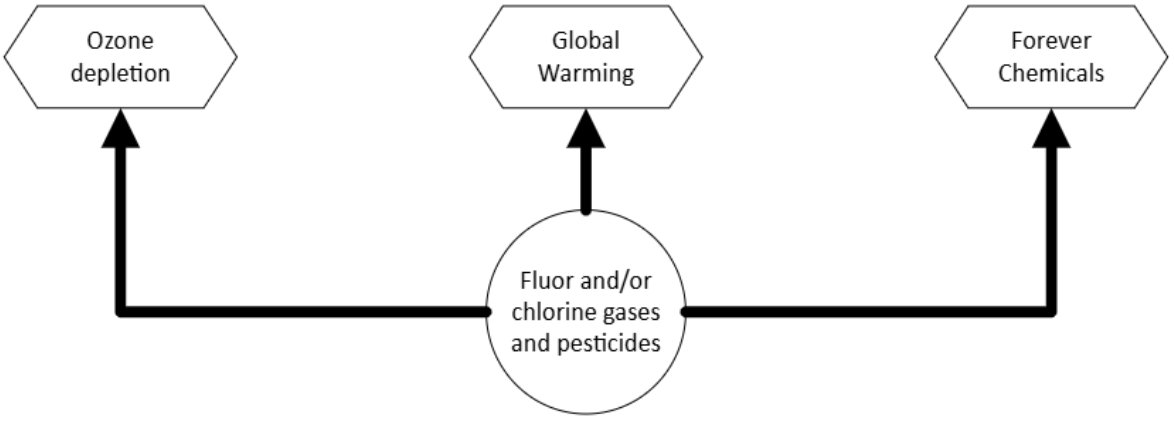


Figure 7. The per- and polyfluorinated hydrocarbons have caused different problems over time. The issues are distinct, yet also part of the same problem.

For the refrigeration and heat pump industry, the way out of this mess is relatively easy, because the solutions are there already. Using the information provided by the Danish EPA, which points out

that the formation of trifluoroacetic acid (TFAA) mainly comes from refrigerants and pesticides, the industry can change within a short time to fluids that do not contain any fluorine, e.g., ammonia, carbon dioxide, various hydrocarbons, air, and water. The solutions have been used for over 150 years and will be available for eternity.

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