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PFAS IN PRODUCTS AND WASTE STREAMS IN THE NETHERLANDS

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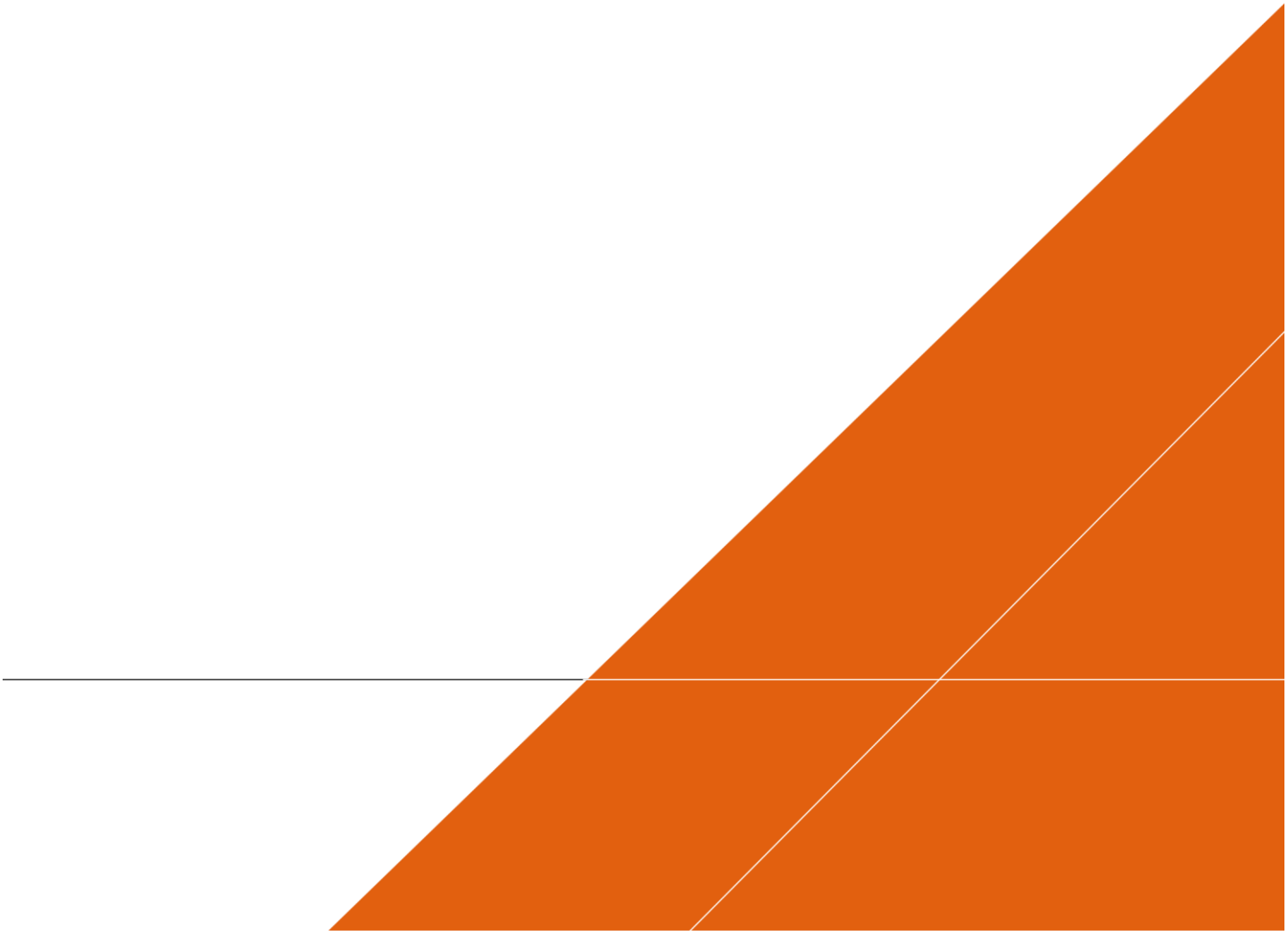

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Summary

This report contains the results of a study that aims to create insight in the presence of PFAS in products, production and recycling processes and waste. It also aims to identify significant exposure routes and release pathways of PFAS in products and waste to humans and the environment. Based on a literature review, relevant categories were identified and subsequently the most appropriate samples were selected for an analytical assessment. This study is limited to the analytical assessment of a portion of the evaluated categories and does therefore not aim to be comprehensive.

Approach

Based upon the exposure routes and release pathways to human and the environment, a selection of samples from different categories was tested. In several industries, dust samples have been taken as a screening method for the presence of PFAS.

All in all, 129 samples of products, dust, processes, recycling and waste have been tested using three types of PFAS-analyses. This three-step approach for the analyses has an increasing level of detail. The three steps consist of; (1) screening all the samples for extractable organic fluorine (EOF, screening for fluorinated compounds), (2) target analysis for 42 individual PFAS and (3) a smaller selection of samples on the Total Oxidizable Precursor assay (TOP). The TOP was mainly aimed to gain insight in the presence of PFAS-precursors and was applied to those samples that showed a high EOF and a relatively low response in the target analysis.

Analytical results

Most of the selected samples contain organic fluorine and/or PFAS. The concentrations vary greatly, between and within the sampled categories. The highest concentrations were found in several types of water- and stain repellent products, like sprays and waxes for textile/leather/carpet, floor protection and polish, windshield treatment and joint protector for bathrooms. The extensive analytical assessment also showed that in more than 90% of the products, less than 10% of the PFAS present could be explained by the PFAS-target analysis. This indicates the presence of other (unknown) PFAS. In recycled paper and paper of fireworks, relative high concentrations of PFPrA were detected, a PFAS-target compound which is usually not analysed for.

The dust samples taken in several industries, some offices and households, indicate the widespread use of PFAS. In dust, the concentrations are often relatively high. The origin of dust is usually not clear, and may also be explained by wear of clothing, carpets. However, in several industries higher levels have been measured than in household and office dust. Together with sewage sludge this is considered to be caused by wear and waste of PFAS containing products.

Interpretation contribution to the environment

Finally, a rough estimate was made of the estimated loads of PFAS in the different categories, based upon the amount of products used in the Netherlands and the concentrations that were measured in this study. The estimated loads offer a valuable impression of the contribution of the different categories to the release of PFAS to the environment.

In order of relevance the approximate loads for the categories tested in this study are:

Highly relevant (>100 kg/year):
- Water and stain repellent products, including treated textile, carpets and leather
- Paper recycling

Moderately relevant (10-100 kg/year):
- Sewage sludge
- Cleaning agents
- Fluoroelastomer products
- Pesticides

Low relevance (<10 kg/year):

- Fluoropolymer products
- Fireworks

In some cases, products with a low relevance for the environment (looking at kgs/year) can cause a direct exposure for PFAS to human. E.g. in PTFE-baking mats or cosmetics. In those categories PFAS-target compounds have been detected. The amounts are relatively low when comparing to the other tested products but can be a concern considering the direct use of the product on the skin or possible uptake via food.

Dutch Summary (Samenvatting)

Het doel van deze studie was om een inzicht te krijgen in de aanwezigheid van PFAS in producten, productie- en recyclingprocessen en afval, en om significante blootstellingsroutes en het vrijkomen van PFAS in producten en afval in beeld te brengen, zowel voor mensen als het milieu. De studie bestond uit een literatuurstudie om de categorieën in beeld te brengen, en vervolgens analyse van geselecteerde producten. De studie beperkt zich tot een deel van de beoordeelde categorieën. Vanwege het grote aantal mogelijke toepassingen beoogt de studie niet volledig te zijn.

Aanpak

Alle geanalyseerde monsters zijn geselecteerd op basis van blootstellingsroutes en vrijkomen in het milieu. Bij verschillende industrieën zijn stofmonsters genomen als screeningsmatrix voor de aanwezigheid van PFAS.

129 monsters van producten, stof, procesmateriaal, gerecycled materiaal en afval. Deze monsters zijn geanalyseerd middels drie verschillende analyses, om een zo goed mogelijk inzicht te krijgen in welke monsters PFAS aanwezig zijn, en welke PFAS-verbindingen het betreft. Voor de analyses is een drietrapsstrategie toegepast: (1) screening van de monsters op extraheerbaar organisch fluor, (2) target analyse voor 42 individuele PFAS en (3) ‘Total Oxidizable Precursors’ analyse (selectie van de monsters), om een inzicht te krijgen in de aanwezigheid van PFAS-precursors.

Resultaten

De resultaten laten zien dat een groot deel van de monsters organische gefluoreerde verbindingen en/of PFAS bevatten. De concentraties variëren aanzienlijk, zowel tussen de categorieën als binnen de categorieën. De resultaten geven daarmee een beeld over de orde van grootte waarin PFAS in deze producten voorkomen. De hoogste concentraties zijn aangetroffen in verschillende typen van water- en vetafstotende toepassingen, zoals sprays voor textiel, leer en tapijt, poetsmiddelen voor vloeren, voegenbeschermer voor badkamers en waterafstotende toepassingen voor autoruiten. In gerecycled papier en papier van vuurwerk zijn relatief hoge concentraties van één specifieke PFAS aangetroffen. Dit betreft een PFAS welke niet in de standaard analysepakketten zit.

De stofmonsters (van omgevingsstof in gebruiksruimtes) zijn genomen bij verschillende industrieën en bij een aantal kantoren en huishoudens. Uit deze resultaten komt het wijdverspreide gebruik van PFAS naar voren. In stof zijn de aangetroffen concentraties vaak relatief hoog, honderden tot duizenden µg/kg. De oorsprong van het stof is niet eenduidig en kan mogelijk worden verklaard door bijvoorbeeld slijtage van kleding en tapijten. Wel is duidelijk te zien dat bij een aantal industrieën de concentraties PFAS hoger zijn dan in huishoudens en kantoren.

Bijdrage aan het milieu

Vervolgens is de omvang van de producten die worden gebruikt in Nederland geïnventariseerd. Gecombineerd met de resultaten van de analyses is een schatting gemaakt van de bijdrage van de verschillende categorieën aan het vrijkomen van PFAS naar het milieu.

Of een productcategorie een relevante bijdrage levert aan het vrijkomen van PFAS in het milieu is niet alleen afhankelijk van de concentratie PFAS in het product, maar ook van hoe vaak en op welke manier het product gebruikt wordt. De voor de conclusies gehanteerde vrachten zijn gebaseerd op ruwe schattingen maar geven wel een indruk van de relatieve bijdrage aan het milieu. De verwachte relevantie van de categorieën in deze studie is:

Aanzienlijk (>100 kg/jaar):
- Water- en vuilafstotende producten, waaronder behandeld textiel, tapijt en leer
- Papier recycling

Matig (10-100 kg/jaar):
- Rioolslib
- Schoonmaakmiddelen
- Fluorelastomeer producten (fluorrrubbers)
- Bestrijdingsmiddelen

Beperkt (<10 kg/year):
- Fluorpolymeer producten
- Vuurwerk

In sommige gevallen kunnen producten waarbij sprake is van beperkte overdracht naar het milieu (enkele kg/jaar), wel bijdragen aan directe blootstelling van mensen. Voorbeelden zijn PTFE-bakmatten, waarin individuele PFAS zijn aangetoond, en cosmetica, waarin ook enkele PFAS zijn gevonden. De hoeveelheden zijn relatief laag vergeleken met de andere geanalyseerde producten, maar er kan wel sprake zijn van humane blootstelling.

1 THE RESEARCH QUESTION

1.1 Introduction

PFAS (per- and polyfluoroalkyl substances) are a group of substances with unique properties. The group comprises a very large number of chemicals (>4000, OECD, 2018) and have been used within a wide range of products and industries. They are now ubiquitously detected in the environment, even at remote locations. All PFAS exhibit extreme persistence, which means that they do not degrade in the environment. Several PFAS are also considered PBT substances, meaning that they are persistent, bio accumulative and toxic. Some PFAS are labelled as Substances of Very High Concern (SVHC), e.g. PFOS, PFHxS, PFOA, PFNA, PFDA, C_{11}-C_{14} PFCAs and GenX[1]. Due to the phasing out of PFOS and PFOA, other types of PFAS have been used as replacements, mainly PFAS with a shorter perfluoroalkyl chain, which are generally less bio accumulative, but more mobile in the environment. These replacements are generally less well studied with less knowledge about their toxicology.

In Figure 1, the use of different PFAS over time is summarised.

PFAS[1]	Development Time Period							
	1930s	1940s	1950s	1960s	1970s	1980s	1990s	2000s
PTFE	Invented	Non-Stick Coatings			Waterproof Fabrics			
PFOS		Initial Production	Stain & Water Resistant Products	Firefighting foam				U.S. Reduction of PFOS, PFOA, PFNA (and other select PFAS[2])
PFOA		Initial Production		Protective Coatings				
PFNA					Initial Production	Architectural Resins		
Fluoro-telomers					Initial Production	Firefighting Foams		Predominant form of firefighting foam
Dominant Process[3]		Electrochemical Fluorination (ECF)						Fluoro-telomerization (shorter chain ECF)
Pre-Invention of Chemistry /			Initial Chemical Synthesis / Production			Commercial Products Introduced and Used		

Notes:
1. This table includes fluoropolymers, PFAAs, and fluorotelomers. PTFE (polytetrafluoroethylene) is a fluoropolymer. PFOS, PFOA, and PFNA (perfluorononanoic acid) are PFAAs.
2. Refer to Section 3.4.
3. The dominant manufacturing process is shown in the table; note, however, that ECF and fluorotelomerization have both been, and continue to be, used for the production of select PFAS.

Sources: Prevedouros et al. 2006; Concawe 2016; Chemours 2017; Gore-Tex 2017; US Naval Research Academy 2017

Figure 1 Use of PFAS over time (ITRC, 2017).

[1] PFOS: perfluorooctane sulfonic acid
PFHxS: perfluorohexane sulfonic acid
PFOA: perfluoroctane carboxylic acid
PFNA: perfluorononane carboxylic acid
PFDA: perfluorodecane carboxylic acid
PFCA: perfluoroalkane carboxylic acid
GenX: 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propionic acid

As a result of increasing detections in global drinking water supplies, the food chain and biota as well as specific high profile incidents, there is increasing political and societal attention on PFAS. Most notably for the Netherlands were the incident with accidental release of AFFF near the Schiphol airport in 2008, and the discovery in 2016 of elevated PFAS levels in the blood of citizens living in the vicinity of the Chemours factory in Dordrecht. These incidents increased the focus on this group of chemicals.

More research in 2017-2018 revealed that all over the Netherlands elevated concentrations can be found in soil, groundwater and surface water (Hage et al., 2018; Pancras et al., 2018b). Typically, higher concentrations are observed in cities compared to rural areas. This was an indication that the presence of PFAS in the environment is related to human activities, not only industry but also the use of products and the production of waste. The decree on soil reuse of July 2019 and the resulting stagnation in the construction and dredging industries further increased the need for more understanding of the causes of PFAS concentrations in humans and our environment.

While there is already a significant amount of literature and knowledge in many areas of PFAS understanding, during the incorporation of PFOS and PFOA in the Stockholm Convention it was highlighted that the understanding of the contribution from the multitude of sources and pathways to the environment is limited. There is a strong need to obtain a better understanding of concentrations in products and emissions in order to be able to set priorities and adequate policy.

In reaction to a question in the Dutch parliament, the State Secretary for the Environment informed the Parliament in Spring 2019 that she would commission a study to assess PFAS sources in products and waste. This report describes the results of this study with the focus on products and waste streams in the Netherlands. However, the outcome of this study may also be relevant for the other EU member states.

1.2 Objective

The goal of this study is to gain insight in the presence of PFAS in products, production and recycling processes and waste, and to identify significant exposure routes and release pathways of PFAS in products and waste to humans and the environment. The study focusses on the identification of the most relevant sources of PFAS and does not aim to be fully comprehensive.

This information gathered in this study can contribute to drafting efficient policy on PFAS and take measures to further reduce risks to humans and the environment.

1.3 Approach and parallel studies

The study consists of 2 phases:

1. Inventory and sampling plan. The inventory comprises a literature review, screening of environmental data and interviews with international experts.
2. Collection and analyses of samples on selected processes and waste streams.

The first phase was executed separately from the second. The second phase consisted of a consecutive process of selecting, collecting and analysing the samples.

The results have been interpreted and described in this report.

Phase 1

In order to obtain the best possible understanding of which products or waste are the most relevant exposure routes, two different approaches are used and combined:

1. A literature review and interviews regarding the presence of PFAS in products and waste (the sources).
2. A review of data regarding the presence of PFAS in soil, water and human blood (the receptors).

The second approach was added because the information on amounts and types of PFAS in products and waste can be scarce and very often not documented. By only performing a literature review on PFAS in products and waste it is easy to overlook major pathways. Therefore, it was decided to add the information about environmental and blood data.

At the end of phase 1, the data regarding sources (products, waste) and receptors (environment, humans) are combined to estimate of the most relevant product and waste streams to be sampled and analysed.

Parallel studies

Simultaneously to this study, two other investigations are being conducted concerning PFAS in products in the Netherlands by Rijkswaterstaat (RWS) and the Netherlands Food and Consumer Product Safety Authority (NVWA). These two studies will focus on discharges (wastewater) and sources of PFAS to these discharges, and on the presence of PFAS in food contact materials. Therefore, the current study will not focus on these types of products and waste streams. Overlap between the studies has been avoided as much as possible. The study of RWS has been finished (Jans and Berbee, 2020), the study of the NVWA is still ongoing.

Phase 2

Phase 2 consists of the execution of analyses for 129 samples. The samples were collected between March and October 2020.

2 IDENTIFYING RELEVANT PRODUCTS AND WASTE STREAMS

2.1 Methodology

It is well known that PFAS can be present in many industrial and household products and waste streams. To get the best possible overview, the inventory and literature research consisted of the following elements:

- Review of core studies regarding PFAS sources. Four published literature sources are the basis for the summary of the list of products and production processes containing PFAS:
  - kissa, 2001;
  - fluorocouncil website;
  - hekster, 2002; and
  - kemi, 2015.

  Some of these studies are relatively old but remain relevant. Where PFOS and PFOA have been used in the past, other PFAS are now likely to be used as alternatives.
- Six interviews with scientists concerning PFAS in products:
  - R. Berbee, Rijkswaterstaat, The Netherlands.
  - M. Janssen, Centre for Safety of Substances and Products, RIVM, The Netherlands.
  - I. Cousins, University of Stockholm, Sweden.
  - D. Borg, Swedish Chemical Agency.
  - C. Staude, German Environment Agency, department Chemicals.
  - Z. Wang, ETH Zürich, Department of Chemistry and Applied Bioscience.
- A search on the websites of several PFAS producers (3M, Chemours, Daikin, Unimatec) about applications in which PFAS are currently being used.
- Gathering information about PFAS in the environment (not comprehensive) as indications for where we could link PFAS in the environment with PFAS uses.
- Collecting a wide range of studies and articles on analyses of PFAS in products, in addition to the core studies and the websites of the producers.

In this chapter, the main decisions and findings that led to the final result of phase 1 - a sampling plan - are described which consider the following key factors:
- Properties and suitability for use.
- Uses of PFAS and an estimate of relevance.
- Presence and distribution patterns in the environment, human blood and dust.
- Linking sources and receptors to PFAS pathways, to priorities in a sampling plan for phase 2.

2.2 PFAS properties and suitability for use

PFAS have unique chemical and physical properties. Fluorinated surfactants are physically and chemically stable, are hydrophobic (repels water) as well as oleophobic (repels oil) (Kissa, 2001). This makes them very favourable for a multitude of uses.

Unique properties and related suitability for use:
- Fluorinated surfactants can lower the surface tension of water more than hydrocarbon-based surfactants. This makes them powerful wetting agents (Kissa, 2001).
- They are useful emulsifiers and dispersants (Kissa, 2001).
- Can be used as mold release agents (Kissa, 2001).
- Improve wetting power, so increase levelling of paints and floor polishes (Kissa, 2001).
- They improve the open time in paints, and are effective anti-blocking agents (website 3M).
- Oil and water repellent, so are soil and dirt resistant, and oil stains are easily removed (website 3M).
- They have a strong chemical stability, so fluorinated compounds can be used in situations where hydrocarbon-based compound will decompose (high temperatures, strong acid or alkaline conditions, oxidizing agents, etc.) (Kissa, 2001).

PFAS are relatively expensive compared to their hydrocarbon equivalents. So, they are mainly used when other compounds do not perform adequately. However, the high price of PFAS can be compensated by the low amounts that are needed for the application, and long service life of the PFAS. Concentrations as little as 50-150 ppm (mg/kg) (0.005%-0.015%) can be sufficient (Kissa, 2001).

PFAS can be used in different forms:

- As non-polymeric / single molecules (e.g. PFOS, PFOA). These type of PFAS are the focus of this investigation. The PFAS non-polymeric molecules can be divided into:
  - Perfluoroalkyl acids (PFAA's). This group comprises different types of acids like:
    - Perfluoroalkyl sulfonic acids (PFSA's), e.g. PFOS (C8), PFHxS (C6).
    - Perfluoroalkyl carboxylic acids (PFCA's), e.g. PFOA (C8), PFHxA (C6), PFBA (C4).
  - PFAS Precursors (compounds which can degrade to PFCA's and PFSA's), e.g. 6:2 FTS, 8:2 diPaP.
  - Perfluoroethers, e.g. GenX.
  - Some PFAS (single) molecules are quite long and are sometimes also defined as polymers (which are exempted from restrictions) (Vestergren, 2019).
- As polymers. This group can be divided into:
  - Fluoropolymer; polymers with a fluorinated backbone.
  - Side-chain fluorinated polymers; polymers with a hydrocarbon backbone and with fluorinated side chains. These side chains can be broken down to PFCA's.
  - Other fluorinated polymers, with perfluorocarbons built into the backbone, like e.g. fluorinated polyurethane.

This investigation focusses on the non-polymeric PFAS present in products and on the non-polymeric PFAS that can leach out of polymeric PFAS.

In many cases it is not clear which PFAS are present in products. Notification on the material safety data sheets (MSDS) is not required for concentrations <0.1% (1 g/kg), and the exact composition is often notified as confidential business information. Furthermore, a REACH registration starts at 1 tonne/year and polymers are excluded from registration.

PFOS and PFOA are being phased out by the European Union (with some exemptions). Maximum allowable levels are 0.001% for PFOS (10 mg/kg) (EU, 2019), 25 ppb (25 µg/kg) for PFOA and 1000 ppb (1 mg/kg) for PFOA-related substances (EU, 2017). With the phase out of PFOS and PFOA, alternatives have been developed to replace PFOS and PFOA, which, most applications, are other PFAS e.g. GenX or 6:2 FTS. The OECD (Organisation for Economic Co-operation and Development) has identified 4730 PFAS-related CAS-numbers. Of these compounds, 4186 (88%) are likely to degrade to perfluoroalkylacids (PFAAs), like e.g. PFOS, PFOA and their analogues (OECD, 2018).

2.3 Uses of PFAS

2.3.1 Textile, leather and carpet industry

PFAS are being used in the textile, leather and carpet industries for their stain and water repellent properties. They are being used in products like furniture, outdoor clothing, shoes, tents, ropes, tablecloths, umbrella's, car seats, carpets etc.

- PFAS can be used in the production of fibres and yarn, however, they are usually applied to the end products: cloth or carpets.
- Durable water repellent membranes like e.g. Gore-Tex®, consist of two PFAS-containing layers; a layer of expanded PTFE for the breathability and a layer of usually side-chain fluoropolymers, which is applied (sprayed) to the outer layer of the fabric. PTFE is a PFAS, which is considered to be inert. However, the end-of-life fate of the PTFE is not well known, and during production of PTFE PFAS-monomers can be released or be present as residual processing aid in the product.
- Usually side-chain fluoropolymers are being used as a surface treatment on the garment to make them soil- and water repellent. These compounds consist of a carbon backbone (not fluorinated), with fluorochemicals attached to the side, being fluorotelomer alcohols or perfluoroalkane sulfonamidoethanols (Holmquist, 2016). During use or wear or washing, the side-chain groups can be released from the fabric and are then being released into the environment as non-polymeric PFAS.

- It is estimated that most of the protective PFAS layer on carpets will be released to the environment during its life span (Hekster et al., 2002).
- As far as is known, a Dutch overview of textile manufacturers and / or textile impregnators is not available. According to MODINT2, an estimated 10 to 30 companies in the textile and carpet industry in the Netherlands use impregnating agents (de Groot et al., 2013).
- C8-based fluorochemicals (e.g. PFOS, PFOA) were generally regarded as the best finisher in the textile industry. These have been replaced by short chain PFAS, but now these finishes are also coming under increasing pressure. The sector is looking for non-fluorinated alternatives.
- The municipality of Genemuiden is the 'carpet capital' for the Netherlands: 60% of the floor covering in the Netherlands comes from this town at the river Zwartewater (website Canonvannederland). How many of these companies use PFAS impregnating agents is unknown (de Groot et al., 2013).
- In tanneries, PFAS are also being used as a finishing step to provide soil, stain and water repellence and during finishing of the leather for homes. Concentrations of fluorochemical in the final products are indicated to be 0.25-0.5 g/kg (Kissa 2001).
- PFAS used within the leather industry were typically acrylate, methacrylate, adipate and urethane polymers of N-ethyl perfluoroctanesulfonamidoethanol (EtFOSE), with a usage rate up to 15% of the fibre weight. These products might also have contained PFOS as an impurity up to 2 wt%. Nowadays PFBS3-based products are being used, as well as short-chain fluorotelomer based PFAS polymers (UNEP-POPS, 2017). According to RIVM-study 'Impregneermiddelen', the Dutch leather industry works with C4-PFAS polymers as an alternative to PFOS substances (de Groot et al., 2013).
- The use of PFAS in the Netherlands for textile, leather and carpet industry has been estimated to be 25-35 tonnes per year for carpet and leather applications (Hekster et al., 2002) (excluding textiles). It is estimated that the majority of the PFAS used for carpet, textile and leather is emitted to the environment during production and use.

Waste and reuse

- Based on the information above, it can be expected that PFAS might be present in waste from the textile, leather and carpet industries, wastewater and sludge of the WWTP.
- PFAS will also be present in used carpets and textiles which are incinerated in municipal incinerators or have been sent to landfills (also in the past).
- Carpets are being reused as raw materials for the cement- and automotive industry. Outdoor furniture and flower pots can also be made of reused carpets.
- In the cement industry the carpet flocks are being used as fuel, and as additive to cement.
- The flocks can also be used as equestrian surfaces, as coating for drainage tubes, in road construction and in the production of clothing, and will therefore be reused in a variety of applications.
- Dust from carpets will eventually end up in household waste. In the Netherlands, this waste is incinerated. It is not yet clear what the fate of PFAS is at the current conditions at the waste incinerators (temperature below optimum temperature for complete PFAS degradation).

2.3.2 Paper

Oil and greaseproof paper is another major use of PFAS, which is mainly being used for food packaging (e.g. pizza boxes, popcorn paper and other fast-food packaging). In an RIVM-study in 2018, it was concluded that PFAS are used in these types of products, but that there is currently not enough information about these PFAS to make reliable risk assessments (Bokkers et al., 2018).

Although oil and greaseproof paper (Ersatz paper) is not produced in the Netherlands as far as known, papers with a PFAS treatment may be a major source of PFAS in the Netherlands due to import of these types of papers. In 2002 it was estimated that 60-105 tonnes of PFAS are imported annually into the Netherlands via paper (Hekster et al., 2002).

[2] MODINT is the Dutch business network of manufacturers, importers, agents and wholesalers of clothing, fashion accessories, carpet and (interior) textiles. There are approximately 600 companies connected.
[3] PFBS: perfluorobutane sulfonic acid

A few PFAS are mentioned in the list of allowable processing aids for paper (Warenwetregeling verpakkingen en gebruiksartikelen)[4], it can therefore not be excluded that these types of PFAS are being applied in the Netherlands in the paper industry.

PFAS are not only being used for oil and greaseproof paper, but also in folding cartons, carbonless forms and masking papers (UNEP-POPs, 2011).

Waste and reuse

- PFAS might be present in wastewater and sludge of the WWTP of paper industries.
- Papers are being reused about 7 times. PFAS might be present in the recycled paper pulp.
- Inks might also contain PFAS. These also end up in the recycled paper pulp.
- At the end of the recycling lifetime, the paper fibers cannot be used anymore, since they are not strong enough and contain too many residues. This paper is being dried and the remaining minerals are being reused as calciferous binding agent in e.g. the construction sector.

2.3.3 Food related

PTFE (food related)

PTFE (polytetrafluoroethene) is a non-stick coating which is applied in many household products, like pans and kitchen utensils. PTFE is a PFAS-polymer which does not produce PFAS monomers, however, during the production of PTFE PFAS like PFOA or GenX (or other perfluoroethers) are being used as a polymerization aid. PFOA has been phased out in Europe, but the use of PFOA in other countries has increased, so PFOA might still be present in imported articles. The restriction on PFOA use in the EU from 4 July 2020 should resolve this issue, for products with PFOA concentrations > 25 µg/kg. In the finished articles, other PFAS monomers like GenX might still be present as residuals from the production process (in concentrations <0.1%).

PFAS polymers can also be used in tubing, hoses, gaskets, sealants and filters of food processing equipment (Kissa, 2001).

Silicon baking forms

Silicon baking forms have also shown to contain PFAS (Blom and Hanssen, 2015), which likely originate from the use of PFAS as mold release agents. Aluminium foil may be coated with PFAS as an anti-blocking agent (Kissa, 2001).

2.3.4 Cleaning agents

PFAS are also being used in household and industrial cleaning agents e.g. carpet spot cleaners, dish washer liquids, degreasers, oven cleaners, cleaners for hard surfaces, precision cleaners and windshield wiper fluids (Kotthoff et al., 2015, Kissa, 2001). They can substantially enhance the cleaning power, and promote a rapid runoff of rinse solutions. PFAS are also used for de-greasing of metal surfaces.

2.3.5 Coatings and polishes

The oil and water repellency is a useful property for several types of coatings and polishes. In carwash polishes PFAS are being used to make the water droplets fall off the surface easily. Also, other water repellent coatings for e.g. tiles and floor polishes contain PFAS. PFAS are also being used as anti-fogging agents, for e.g. greenhouses, but also for glasses, mirrors etcetera (Kissa, 2001).

PFAS are being used in paints, e.g. water based latex paints, for the levelling properties (Kissa, 2001). The amounts used are estimated to be low, since there are cheaper alternatives (Kemi, 2015; Hertzke, 2012).

[4] https://wetten.overheid.nl/BWBR0034991/2017-01-01. E.g.:
- ammoniumbis(N-ethyl-2-perfluoroctaansulfonamideethyl)fosfaat
- copolymeren van 2-(perfluoroctylsulfonylaminomethyl)ethylmethacrylaat, 2,3-epoxypropylmethacrylaat, ethoxyethylacrylaat en methacryloylmethyl-trimethylammoniumchloride
- perfluoralkyl(C6-C16)(C6-C18)fosfaten van bis(2-hydroxyethyl)amine

Also, in several types of ink (printing inks, ball point pens, marking pens), PFAS are being used to improve levelling ink (Kissa, 2001).

2.3.6 Fire-fighting foam

PFOS has been used extensively in AFFF (aqueous film forming foam) in the past, because of the strong resistance against high temperatures and excellent film forming properties. Alongside the phase out of PFOS, a shift towards fluorotelomer based foams has occurred, with fluorosurfactant foams now mainly using C6-fluorotelomers (fluorotelomers with a fully fluorinated backbone of 6 carbon atoms). It is known that AFFF also contain a significant proportion of proprietary PFAA precursors, which are not detected with regular PFAS analyses (Barzen-Hanson et al., 2017). However, once within the environment or the human body these PFAA-precursors can biotransform to detectable, regulated PFAAs such as PFHxS and PFOA. Firefighting foam may be directly released into the environment from regular fire training, incident response or accidental release unless a containment and treatment system is present. The amount of PFAS used in firefighting foam was estimated to be 1-4 tonnes/year in the Netherlands in 2002 (Hekster et al., 2002). In 2009 it was estimated that 16.000 m^3 of AFFF was left in stocks (Bruinen de Bruin, 2009), with a PFAS content of 5% this coincides with 0,8 tonnes of PFAS. Currently several fire departments are in a transition to use fluorine free foams (F3-foams). These F3-foams have been evaluated by the European Chemicals Agency (ECHA) and the European Commission (final report issue 3, 2020), and are considered generally available and technically feasible.

Waste and reuse

- Foam used for fire protection in buildings has to be periodically be replaced by new foam. Legacy foam stocks are still present at several sites.
- The concentrate is processed by a waste processing facility. It is not clear whether all foam is being processed in the right way and being incinerated. It might also be the case that it is being processed by a biological wastewater treatment plant where PFAS may be released at the surface water without treatment and accumulated within biosolids.

2.3.7 Other industries

PFAS are being used in many other industries such as fluoropolymers in gaskets, hoses, seals etc. PFAS also have also been used in lubricants, as emulsifiers, dispersants, mold release agents, wetting agents and for their water and oil repellence.

Fluoropolymer production

In Europe, 52,000 tonnes of fluoropolymers are sold annually, which are used in several industries like transport, chemical industry, electronics and cookware (PlasticsEurope, 2017). PTFE is the most important fluoropolymer (70%), FEP and PVDF are the next most widely used fluoropolymers (Gardiner, 2015).

In the production of PTFE and other fluoropolymers, PFAS are being used as polymerization aid. For example, PFOA and GenX have been used by Chemours in Dordrecht for many years, and have caused elevated levels of these compounds in the area around the factory (Bentum, 2017) via dispersion in air and subsequent deposition. Also at a site where PTFE dispersions have been dehydrated, increased levels have been found in soil, groundwater and surface water up to several km from the site (Bentum, 2018a). The PTFE production plant in Dordrecht has emitted several tons of PFOA and GenX per year to the surface water and the air (Roelandse, 2017; Zeilmaker et al., 2016), and via waste (IL&T, 2019).

It is known that the produced PTFE slurries or dry powders still contain some PFAS molecules like GenX as impurities. It is unclear whether these impurities cause a contamination at the sites where these semi-finished PTFE products are being processed.

Fluoropolymer applications

It is unclear whether the impurities in semi-finished fluoropolymers cause a large release of PFAS to the environment at processing and application sites. These sites might be sites where, for example, fluoropolymer coatings are being applied, like coated pipelines for the chemical industry, where fluoropolymer parts are being molded, or kitchen utensils are being coated.

Aerospace and aviation

In aerospace and aviation PFAS have been used in brake and hydraulic fluids (Kissa, 2001), lubricants, interior, fluoropolymers are being used in many types of tubing, seals, wire and cables etc.

Galvanic industry

It is well known that PFOS has been used as a mist-suppressant in the chrome plating industry, but also in other applications in the galvanic industry. PFAS might be used to promote the flow of metal coatings, prevent cracks during drying, prevent corrosion and reduce mechanical wear (Kissa, 2001). In an inventory in 2009, the amount of PFOS used in the metal plating industry was estimated to be 0.4 tonnes/year (worst case) (Bruinen de Bruin et al., 2009).

Buildings and construction

Coated fabrics and metal roof coatings are being used as large architectural membranes, e.g. above stadiums. PFAS are also being used in paint coatings, adhesives, sealants and caulks. Other applications for PFAS are as additives to cement to reduce shrinkage (Kissa, 2001), as mold release agents and to use as an oil and water-repellent coating.

Automotive

In the automotive industry, PFAS are being used as fluoropolymers in several types of rubbers, seals, o-rings and valve packings, but also in lubricants, textiles, engine oil coolers, and surface treatment of the outer parts of the car. PFAS may be used as mold release agent for tyres, one of the most commonly used and fast wearing products.

Rubber

Fluoropolymer types of rubber (e.g. Viton) may contain monomers as residues, furthermore, PFAS monomers are being used as mold release agents, mainly in the rubber industry (website Daikin, 2019).

Electronics

In electronics, PFAS are being used as fluoropolymers in hard disk drives, cell phones, printed circuit boards and optical fibres. They can also be mixed into polycarbonate as flame retardant (website 3M, 2019). They are also being used as etching and resistant materials.

Energy

PFAS are being used in lithium batteries and fuel cells (website Fluorocouncil, 2019), but also as anti-fogging agent or to obtain a quick wash off on solar panels.

Oil and gas

In the oil and gas industry, PFAS polymers are being used in several types of seals, hoses, resistant piping (website Fluorocouncil, 2019), but also to stop water from blocking natural gas wells (website 3M, 2019).

Semiconductors

PFAS are being used in the semiconductor industry as etching and resistant materials, but also as drying and cleaning fluids, wetting surfactants, plasma and vapor deposition machinery. Fluoropolymers are being used in gaskets, o-rings, tanks, valves, pumps and piping (website Fluorocouncil, 2019).

Healthcare and hospitals

At healthcare and hospitals fluoropolymers are being used in filters, tubings, o-rings, seals and gaskets, but also in defibrillators, implants, pacemakers etc. Some PFAS are also used in certain medicines. The packaging of medicine might also be treated with an oil and water-resistant coating. Other uses are oil and water-resistant coatings for personal protection (PPE), garments, drapes and curtains. (website Fluorocouncil, 2019).

2.3.8 Miscellaneous uses

Pesticides

The main known use of PFAS in pesticides has been the use of PFOSA against leaf-cutting ants. This product has not been allowed in Europe. However, there are some PFAS that are being used as dispersants or adjuvants in certain herbicides and fungicides, to aid wetting and penetration of the active ingredients (Kissa, 2001, Kemi, 2015), for this purpose, relatively low concentrations could be sufficient (0.1%) (Kemi, 2015). It is not clear whether, and to what extent, PFAS have been used for this purpose in the Netherlands.

Many of the highly selective pesticides that are used currently are based on fluorinated building blocks (website Unimatec, 2019). In most highly selective pesticides the fluorinated group of the pesticides is quite short, and it depends on the definition of PFAS whether these compounds are highlighted as PFAS.

Cosmetics

PFAS are being used in water resistant mascara and to make eye shadow more brilliant. PFAS are also being used in suntan lotions, sunscreens, lip gloss, foams and conditioners (website Unimatec, 2019). PTFE is being used for dental floss.

Skiwax

PFAS are being used in skiwax to improve gliding properties and have been detected in high levels in the blood of ski waxing personnel (Nilsson et al., 2010). Just recently, the International Ski Federation has decided to ban PFAS in all competitive ski disciplines from the winter of 2020/2021 (website ChemicalWatch, 2019).

Artificial grass

PFAS have also been detected artificial grass and in the backing of the turf, according to Ecocenter (2019). This could be due to the use of PFAS as in the production of the plastics and rubbers, they could be blended into the plastic as an additive, or added as a coating layer to reduce friction (website Ecocenter, 2019, Lambert et al., 2005).

2.3.9 End of life industries

As PFAS are very recalcitrant, they are being detected at many end-of-life industries, like landfills, wastewater treatment plants (including sludge), compost facilities, waste incineration and waste processing sites.

2.4 PFAS in the environment and human

The use of PFAS in so many products and industries has had the consequence that PFAS are found, usually at low concentrations, ubiquitously in the environment and biota. PFAS are found in the blood of people all around the globe. As another important indicator of transport mechanisms, the presence in topsoils may be important.

In this paragraph, we briefly describe findings of the presence of PFAS in the environment, the presence of PFAS in blood as indicator of changing PFAS usage, and PFAS in dust as a possible pathway and screening indicator for the presence of PFAS in products.

2.4.1 PFAS in the environment in the Netherlands

In the Netherlands, we have a background concentration of PFAS in the topsoil of 1-2 µg/kg d.s (Wintersen et al., 2020). These are low concentrations but have recently caused a significant issue in the Netherlands regarding soil reuse. Furthermore, PFAS are man-made chemicals, they do not occur naturally. The data shows that the concentrations in urban areas are usually higher than the concentrations in the rural areas. Long range transport of PFAS within the atmosphere and subsequent deposition can be an explanation for the elevated PFAS levels within topsoil.

However, long range atmospheric PFAS transport and deposition does not explain the differences observed between the urban areas and the rural areas. It is very likely that the widespread use of PFAS containing products also contributes to the increased levels of PFAS at short to intermediate distances.

In 2016-2018 several soil and groundwater investigations were undertaken to assess the presence of contaminated hotspots concerning PFAS in the Netherlands (Pancras et al., 2018a). The major source zone locations for PFAS are fire training areas (due to the use of PFAS containing AFFF in firefighting activities) and in the Netherlands, the Teflon production site of Chemours in Dordrecht is a major source for PFOA and GenX (via air deposition), since it has emitted tons of PFAS in the past. Increased concentrations of mainly PFOA are being found up to 50 kilometers from the site (Wintersen et al., 2020). Other suspected sites, like landfills, chemical industry and the metal industry are considered potentially linked to the presence of PFAS as well.

PFAS have been detected in the Dutch environment at significant concentrations at;

- A PTFE production site.
- At a company where PTFE slurries have been dried.
- Fire training areas.
- Fire incident areas.
- Landfills.
- Metal industry.
- A company where jerrycans / IBCs of AFFF were being rinsed and reused.
- Higher concentrations than background in sediments in areas with greenhouses (the source of these concentrations has not been identified yet).
- N-EtFOSAA is often detected in surface water sediments.
- Higher concentrations of PFOS in the soil than background in coastal areas.
- Higher concentrations of PFOS and PFOA in topsoil of urban areas than in rural areas.
- (Pancras et al., 2018b; Wintersen et al., 2020; van Bentum et al., 2018b, 2019).

2.4.2 Blood

With the phase out of PFOS and PFOA, temporal trends in the types and concentrations of PFAS in blood can be seen. Over a period from 1996 to 2014 Glynn et al. (2012, 2015) observed the following trends in blood of primiparous women in Sweden:

- Decreasing concentrations of PFOS and PFOA.
- Increasing concentrations of PFBS, PFHxS, PFNA and PFDA.

The increase of the PFBS and PFHxS are attributed to the exposure to PFHxS in drinking water. In addition to the known PFAS, more and more unknown organofluorines are detected, which may include other PFAS. A study of Chen et al. (2016) concludes that 2 – 44 % of total organic fluorine (TOF) in blood samples can be accounted for by the measured PFAS and Yeung & Mabury (2016) shows a trend of identified and non-identifiable extractable organofluorine (EOF) which is clearly illustrated in the image below (Figure 2). In the image it can also be seen that the total EOF concentrations are comparable over the years (around 20 µg F/l), the fraction of identifiable PFAS decreases and the fraction of PFAS that could not be identified increased in the last couple of years.

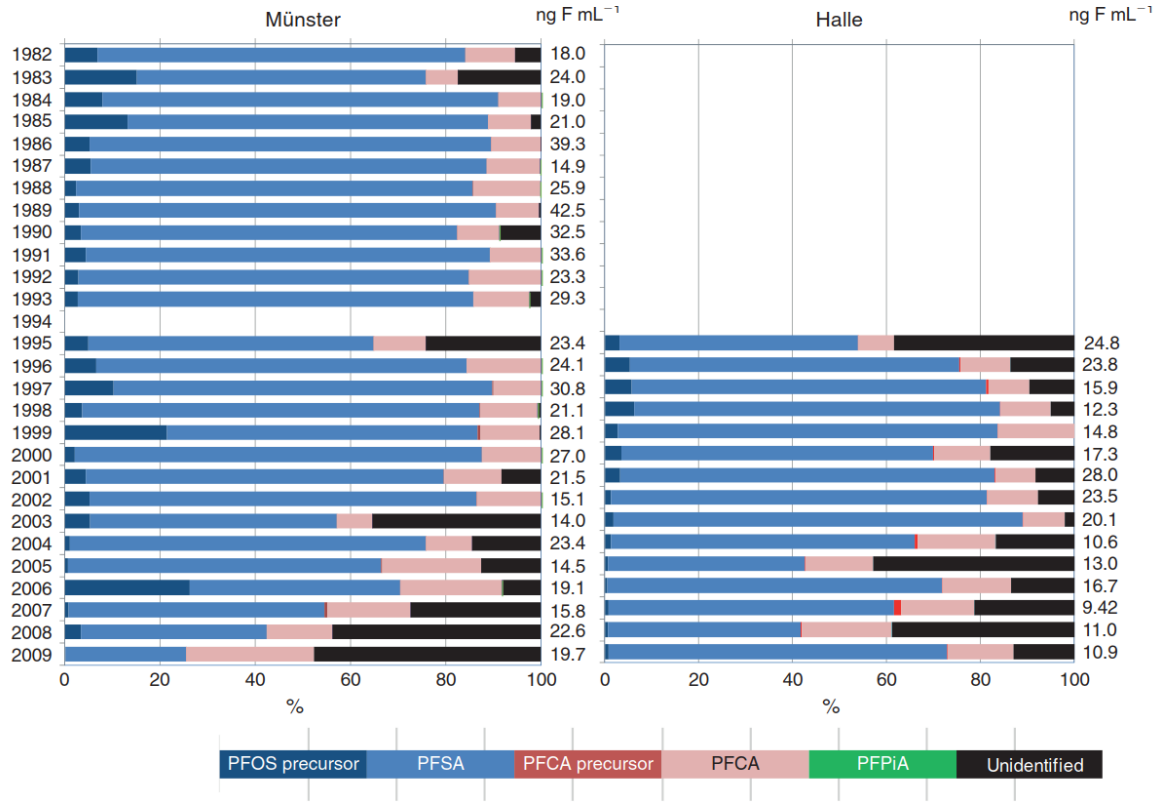

Figure 2 Composition and concentrations (ng F/ml) of EOF in German blood plasma samples (Yeung and Mabury, 2016)

According to Wu et al. (2015), the serum concentrations are related to different factors for children or adults and even considering the known factors not all identified PFAS can be explained. Children usually have higher concentrations in their blood than adults, which can be explained by children coming into contact with their environment more and eating more food relative to their bodyweight which means the intake of PFAS per kg body weight is also higher.

In a more recent study conducted in Flanders (Belgium), some additional factors have been found. The relationship between PFAS concentrations in blood and the following factors have been documented (Wu et al, 2015, Colles et al., 2020):

- Residential dust concentrations.
- Occupational exposure.
- Frequency of wearing waterproof clothes.
- Having used fire extinguishers.
- Frequencies of eating fish and red meat.
- Eating fast food and butter/margarine.
- Eating microwave popcorn.
- Eating offal and locally grown food.
- Use of cosmetics.

Altogether, there is an observed trend of increasing number of PFAS and PFAA precursors being used, including unidentifiable organofluorine compounds. At the same time several factors are identified as possible explanations for increased PFAS concentrations in blood.

2.4.3 Dust

Several studies have shown that PFAS are present in household dust at concentrations around 100 µg/kg (Lucattini et al., 2018). Dust might therefore be a contributor to the background concentrations of PFAS that are found in the soils in the Netherlands.

In a study of Bjorklund (2009), dust in several houses and offices was analysed. The dust in offices and apartments contained the highest concentrations of PFOS and PFOA. Dust from houses, day care centres and cars contained less. In this study no clear correlations have been found between potential emission sources and the concentrations found in dust. However, the highest concentrations of PFAS were being found in the office of a major newspaper publisher.

Also, in the study from Harrad et al. (2019), where PFAS was measured in dust in the indoor environment in Ireland, there was no significant difference between samples with a present putative PFAS source and samples where this source was not present. The measured concentrations in this research were lower than mentioned in the studies of Lucattini et al., (2018) and Bjorklund (2009), with PFBS being the dominant PFAS (17 µg/kg PFBS, ∑ PFAS (6) = 32 µg/kg).

Haug et al. (2011) analysed dust in Norwegian homes on PFAS. They found correlations between PFAS concentrations and the presence of synthetic rugs or Gore-tex clothes.

Eriksson and Kärrman (2015) included polyfluorinated phosphate esters (PaP) into their analyses of dust. They concluded that the concentrations of PAPs were higher than those of the other PFAS classes. Concentrations of several hundreds to thousands of µg/kg were observed. Furthermore, the presence of PAPs in household dust is related to lifestyle. The study clearly shows that there is a need for analytical assessment of the precursor compounds like PAPs in the exposure studies.

In Sweden, very high levels of PFAS (specifically perfluorinated phosphonic and phosphinic acids) have been detected in household vacuum cleaner bags in the city of Skellefteå (up to 2000 µg/kg). To narrow down the origin of these high levels, vacuum cleaner bags of several industries in the area around the city were analysed. Dust samples of six industries were also sampled. The earlier findings of extremely high concentrations in the households in the communities could not be confirmed by the study, but several industries showed clearly higher concentrations than in households, with total concentrations of PFAS ranging to more than 5000 µg/kg:

- Paint industry.
- Plastic industry.
- Rubber industry.

Elevated concentrations were also measured at a smelter, which might be due to PFAS residues in the electronic scrap or in the recycling process (Weiss et al., 2019). The main conclusion of the study is that there is an ongoing use of PFAS in these industries, and that the industries are not always aware of PFAS being present in their production processes.

Furthermore, dust sampling might be a good method to detect PFAS, even in industries where it is not clear where and whether PFAS are being used.

2.5 Quantities of PFAS used in the different industries

The amounts of PFAS used in the different industries is one of the remaining questions concerning PFAS.

Information on actual quantities is scarce. In 2002 some estimations have been made concerning the use of PFAS in carpet, leather, paper, as polymerisation aid and in fire-fighting foams. These numbers summed up to 87-145 tonnes of PFAS/year (excluding textiles) in the Netherlands (Hekster et al., 2002). Since 2000, 3M has phased out their products based on C6, C8 and C10-perfluoroalkane sulfonyl fluoride (PASF)-based chemistry, and replaced them with C4-based chemistry (Wang et al., 2014). Furthermore, PFOA has been phased out for the major part in the period of 2010-2015 in the western countries, via the PFOA-Stewardship program. Since the study in 2002, there has been a shift towards C4-based chemistry and short-chain fluorotelomer products.

However, in other countries like India, Poland, China and Russia the use of PFOA has not been phased out, and the production of PFOA and the use of PFOA as processing aid for fluoropolymer production has increased in these countries to similar levels as it was in Japan, Western Europe and the USA (Wang et al., 2014).

In the Netherlands, PFOA is estimated to have been emitted from the fluoropolymer production plant in Dordrecht in about 3.5 tonnes per year around the year 2000, which could have been higher before 1998 (5-14 tonnes/year, Zeilmaker et al., 2016). GenX is being used in the fluoropolymer production since 2012, and the emission to air and water has been minimized (de Kort et al., 2019).

The shift in uses of different types of PFAS, the use of PFAS-precursors, PFAS-polymers and PFAS which are in many cases noted as “confidential business information” make it difficult to estimate how much PFAS is actually being used.

Recently, a comprehensive overview of the uses of PFAS has been published (Glüge et al., 2020). In this article, more than 200 uses (64 use categories) have been identified for more than 1400 individual PFAS. The uses have been assigned to the (individual) PFAS and vice versa. However, it was very complicated to assign quantities of PFAS to the different uses.

Two inventories were used to estimate quantities of PFAS which could be assigned to the different uses, based on the US Toxic Substances Control Act (TSCA) and on the SPIN database (Substances in Preparations in Nordic Countries) of Denmark, Finland, Norway and Sweden. The REACH database has not been evaluated for this purpose, since in the REACH database the amount of substance reported refers to the chemical and not to the type of use (Glüge et al., 2020).

It is striking that where in the past the majority of perfluorooctane sulfonic acid based compounds (e.g. PFOS-related compounds) was being used in the textile and paper industries (Wang et al., 2017), in the evaluation of the SPIN database and the TSCA database, the amount of PFAS coupled to the textile and paper industries is relatively low, as given in Figure 3 and Table 3 of the paper of Glüge et al., 2020. However, there is still much uncertainty, e.g. in the TSCA database, more than 80% of the volume entries is confidential business information (Glüge et al., 2020), which means that the table is being based on 20% of the entries where the actual compounds are being mentioned.

The highest amounts of non-polymeric PFAS in the Nordic countries are being used in building and construction, electronic industry, electricity, gas, steam and air conditioning, flame retardants and extinguishing agents. 90% of the non-polymeric PFAS (5650 tonnes in the period of 2000-2017) used concerned 1H-pentafluoroethane (HFC-125), a compound usually not considered/included in most PFAS evaluations (C2-PFAS. Also not analysed in this research). 470 tonnes were used in flame retardants and extinguishing agents (it was not possible to distinguish these), the remaining 180 tonnes of non-polymers are being used in other categories. The amounts used in floor covering (incl. carpets) and textiles is low, as well as the amounts used in paper packaging.

In the TSCA-evaluation, the highest amounts of PFAS (non-polymers) have been found in the categories functional fluid for machinery manufacturing, electrical equipment, appliance and component manufacturing, industrial gas manufacturing (air conditioner/refrigeration) and other chemicals. The majority of these applications use 1H-pentafluoroethane, similar to the SPIN-database.

3 SELECTING SAMPLING STREAMS

There is a multitude of products and waste streams that may contain PFAS, and it is key to focus on the pathways or streams that are suspected to contribute most to the exposure to PFAS. Based on the review undertaken and expert judgement, it has been estimated how relevant the presence of PFAS in the products and waste streams might be for exposure to humans and the environment. This expert judgement is based upon several considerations:

- **Quantities and concentrations used:**
  The major indication of relevance for selection are the amounts and concentration levels of PFAS used in an industry or a product. The PFAS processing industry (Chemours, Custom Powders) is obvious. But moving down the distribution chain, the data on quantities are scarce and, in many cases, it is not notified on the product. Concentrations <0.1% (1 g/kg) are in most cases not notified on the material safety data sheets (MSDS), and the exact composition is often notified as confidential business information. Impurities of precursors can also be present without notification. In the sampling plan an estimate is made based upon the number of references in the literature or the suitability of PFAS for the specific use.

- **Dust is a major pathway:**
  Several research articles have indicated that exposure to household dust is a significant source, after ingestion of food and drinking water. The diffuse presence of PFAS in the topsoil in the Netherlands, with higher concentrations in urban areas than in rural areas can, can plausibly be explained by contributions of diffuse applications via dust. Furthermore, higher concentrations of PFAS in blood of children, compared to adults, are for a significant part attributed to intake of dust. Another transport route is via air, which at short distances could be due to the use of volatile PFAS like e.g. FTOHs (fluorotelomer alcohols). In the selection of samples, not only solid products are considered, but also the dust in ventilation systems or vacuum cleaner bags, which are frequently used in PFAS investigations. They have the benefit that they are an outcome of multiple possible sources or types of products (e.g. different carpets/treatments).

- **Primary and secondary sources:**
  Besides the primary sources, secondary sources like wastewater treatment plants (WWTP) and dust are interesting to investigate since they can provide information on (i) the widespread distribution of PFAS and (ii) they can be used as screening matrices to trace the presence of PFAS back to their original sources. They can be viewed as some sort of memory of historical emissions, or a collective grab sample of multiple sources. Therefore, at several industries dust samples are included, as well as sludge samples from WWTPs. Water from WWTPs is being investigated in parallel research, commissioned by STOWA (in progress).

- **The impact of many diffuse sources:**
  PFAS have penetrated our everyday products to a high degree. Whereas isolated industries may have a strong local impact, the presence in carpets, rubber products and other consumer products can mean that PFAS are released to the environment at a much wider scale, and most probably have a significant contribution to the level of PFAS in blood, soils etc. Products that are more widely used and can emit PFAS are more considered than products with less widespread usage. As example, paints, greasing products, tyres can be found everywhere, and the production of dust by tyres is obvious.

- **Environmental concentrations:**
  At several locations in the environment, elevated level of PFAS have been measured, which cannot be explained by (clear) direct sources of PFAS, e.g. in the surroundings of greenhouses, which might be due to pesticides used, coatings for glass used, or perhaps the use of recycled carpet as a substrate material.

- **Already available data and parallel research:**
  The PFAS content of some products is already well investigated. An example of this are firefighting foams. They are a very well-known source of PFAS, and the transition from Aqueous Film Forming Foams (AFFF) to Fluorine Free Foams (F3 foams) is well on its way. Products with sufficient data on PFAS, like AFFF, are not added to the sampling list.

Parallel to this study there are two other studies on PFAS (RWS and NVWA). The sampling plan of both studies is taken into account in the choice of sampling and care is taken to the added value of the studies, not to repeat the same samples. Since the NVWA is investigating food contact materials for PFAS, these items are left out of our sampling plan.

3.1 Samples

This investigation focusses on products, dust and solid waste (and reuse) streams. It focusses on the streams which are most suspected for the presence of PFAS. It is, in most cases, not clear whether PFAS are actually present in these products, which PFAS might be present and at what concentrations. The analyses are therefore focussed on providing broad PFAS analysis (see paragraph 2.5) which will give more insight to the assessment. For the sampling and analysis, a stepwise approach was followed in which the initial sampling and analysis could result in adaptation of the subsequent samples in the sampling plan as well as subsequent sample selection.

An overview of the taken samples is given in Table 1. The initial sampling plan has been subject to change, throughout the project, based on the results of the sampling and analyses, and also on other developments, like the COVID-19 pandemic and developments in the parallel studies. In the table, the relevance has been estimated based on the information gathered during the inventory and completed with expert judgement. The relevance is based upon the expectation of amounts used, risk of emissions to the environment, type of PFAS etc. For example:

- The relevance of sampling carpets is estimated to be high, because it is well known that large quantities of PFAS are used, carpets are present in nearly all living spaces, and most probably the cause of indoor dust and high human exposure.
- Hydraulic fluids for the aircraft industry are expected to be of moderate relevance because the amount of PFAS use is limited, their use is limited to airport maintenance facilities and expected to be well controlled.

Furthermore, the table shows the number of samples to be tested and a short explanation is provided.

As can be seen in Table 1, different types of samples have been. The total amount of samples is 129 for phase 2, comprising:

- 29 dust samples (for screening industries and reference samples, plus 1 reference sample for the dust collector).
- 83 product samples.
- 17 process/waste samples.

The samples have been gathered and analysed in batches. The progress of sampling and analyses has been discussed and the sampling plan was adjusted during the process.

Table 1 Samples from different products, processes, recycling and waste streams

Industries and products	Expected relevance	# samples	Rationale
Textile, leather and carpet industry			
Carpet industry			
Carpet	high	11	3 Dutch carpets. Major source of dust, ubiquitous, recycled carpet fluff (4), 3 dust samples carpet industry and sludge from a wastewater plant in the area of carpet production
Textile industry			
Outdoor gear (non-Gore-Tex sprayed etc.)	high	3	Use of PFAS expected and broadly used product. Has been combined with other outdoor textile applications

Industries and products	Expected relevance	# samples	Rationale
Footwear	moderate	0	Limited expected contribution compared to outdoor gear, similar exposure mechanisms
Other outdoor textile applications (tents, awning cloth,)	mod/high	4	2 dust samples, 3 textile samples above and 2 sails from sailmakers.
Ropes	low	0	Not considered to be relevant
DWR (Goretex)	moderate	0	Known content
Sprays	high	3	Water and stain repellent sprays
Leather industry			
Furniture	high	2	Leather sample and dust from furniture industry, broad use expected and widely used product
Clothing	moderate	0	Limited compared to furniture or footwear
Sprays and creams for footwear/leather	high	3	Sprays, creams are widely used.
Paper industry			
Oil and greaseproof paper	high	12	Considered to be very relevant and additional to NVWA study. 4 types of paper tested (non-food related). 8 paper recycling related process streams have been tested
Carbonless paper	moderate	0	Limited quantities
Double walled paper	moderate	0	Limited quantities
Food related			
PTFE coated pans and other kitchen aids			
Coated materials	moderate	6	Ideal medium to test import of PFOA containing PTFE from Asia. 3 pans testes, and 3 PTFE baking mats
Other food related			
Processing equipment; Tubing, hoses, gaskets, sealants, filters	low	0	Low exposure and emissions expected
Silicon baking forms	moderate	3	Direct contact with food, possibly used as mold release agent, thus secondary contaminant
Aluminium foil	low	0	Not really expected as PFAS containing
Cleaning agents			
Household cleaning agents			
Dish washer detergent	moderate	4	Dishwasher cubes and rinsing aid.
Degreasers, oven cleaners, specific surface cleaners	moderate	5	Popular, cheap cleaning agent, bbq cleaner, abrasive, bathroom cleaner and phone cleaner tested.

Industries and products	Expected relevance	# samples	Rationale
Industrial cleaning agents			
Cleaners for hard surfaces	moderate	0	Industrial use is very specific, not widely used
Precision cleaners	moderate	0	Industrial use is very specific, not widely used
Coatings and polishes			
Carwash	high	3	Three carwash products tested
Anti-fogging/fouling greenhouses	moderate	2	Increased levels of PFAS in soil near greenhouses are found, cause not clear. Analyses are combined with coatings solar panels and windshield treatment fluids.
Paints	moderate	3	Two paints and one outdoor protector analysed
Floor polish	high	3	Many households, PFAS possible
Inks	moderate	3	Dust of printing industry
Water repellent coating	high	1	Overlap with anti-fogging and windshield treatment fluids
AFFF			
Firefighting, AFFF	high	0	Well known
Other industries			
Fluorpolymer/-chemical industries			
PTFE production; semi-finished articles	high	0	PTFE production is not tested. Finished articles are tested
Fluoroelastomer application	high	3	3 Viton/FKM rubbers tested
Fluoropolymers	high	3	3 finished articles tested (PTFE, ETFE, FEP)
PTFE/fluoropolymer application, e.g. lined hoses	high	4	Dust samples at sites where fluoropolymers are being processed into products.
Aerospace/aircrafts			
Brake and hydraulic fluids	moderate	0	hydraulic fluids are extremely well monitored in the aircraft industry, PFAS are present, amounts are estimated to be low
Tubing, seals	low	0	Low exposure, and emissions expected
Galvanic industry			
Mist suppressant in chromium plating	Moderate	0	Known application of PFASs (PFOS (former), 6:2 FTS (current) used as mist suppressants)
Improved levelling	Moderate	0	Low exposure and emissions expected
Reduce sulfuric acid mist in copper production	Moderate	0	Low exposure and emissions expected

Industries and products	Expected relevance	# samples	Rationale
Automotive			
Rubbers, seals, rings, valve packings	low	0	Seals low emissions, fluorinated rubbers and tyres are taken into account in category fluoroelastomers and rubber industry
Lubricants and sprays	moderate	4	Spray with and without PTFE, engine treatment, general lubricant
Textiles	moderate	3	Car textiles are made especially stain and grease repellent.
Engine oil coolers	moderate	0	Little info, most probably not relevant
Car lacquers	moderate	2	Widely used sprays
Windshield treatment fluids	moderate	2	Direct release to the environment. Water repellence. Will be combined with coating greenhouses and solar panels
Car treatment (wash/wax)	moderate	0	See coatings and polishes
Buildings and construction			
Fluoropolymer-coated fabrics	moderate	0	See tents
Fluoropolymer-based metal roof coatings	low	0	Combined with water repellent coating
Paint coatings	moderate	0	See coatings and polishes
Adhesives, sealants and caulks (kit)	Moderate	3	used everywhere, also in households, water repellent kits etc.
Cement additives	moderate	0	Tested in RWS study
Water repellent coating	moderate	1	Big surfaces, direct release to the environment, quantities unknown
Rubber industry			
Fluorinated rubbers (fluoroelastomers)	moderate	0	See fluoropolymers and elastomers
Mold release agent	high	4	Car tyres, source of dust globally, cheap tyres, and dust of rubber industry
Plastic industry			
Fluorinated plastics (PTFE, FEP)	moderate	0	Is already part of PTFE slurries
Mold release agent	high	3	Dust, high concentrations of PFAS found in a Swedish study
Electronics			
Etching and resist materials	moderate	5	Dust sampling at electronic industries and technical rooms
Hard disk drives, cell phones	low	0	Low emission expected
Printed circuit boards	low	0	Low emission expected
Optical fibers	low	0	Low emission expected
Energy			

Industries and products	Expected relevance	# samples	Rationale
Lithium batteries	low	0	Low emission expected
Fuel cells	low	0	Low emission expected
Photovoltaic solar panels	low	0	Sprays and surface treatments are part of windshield treatment or anti fouling and fogging treatments greenhouses
Oil and gas			
Seals, hoses, resistant piping	low	0	Low emission expected.
Preventing the blocking of wells by water	high	0	Difficult to obtain
Semiconductors			
Etching and resist materials	moderate	0	Highly controlled and dust free environment expected. Application in process, not in product
Drying and cleaning fluids	moderate	0	Highly controlled and dust free environment expected. Application in process, not in product
Wetting surfactants	moderate	0	Highly controlled and dust free environment expected. Application in process, not in product
Plasma and vapor deposition machinery	low	0	Highly controlled and dust free environment expected. Application in process, not in product
Gaskets, o-rings, tanks, valves, pumps, piping	low	0	Highly controlled and dust free environment expected. Application in process, not in product
Healthcare and hospitals			
Filters, tubing, o-rings, seals, gaskets	low	0	Low emission expected
Defibrillators, pacemakers etc	low	0	Low emission expected
Packaging for pharmaceuticals	low	0	Low emission expected
Drapes and curtains	moderate	0	Comparable to outdoor gear, tents
Garments	moderate	0	Comparable to outdoor gear
Oxygen delivery to tissues	moderate	0	Low amount expected
Miscellaneous			
Pesticides/herbicides	moderate	3	Little indication of amounts used, but environmental data suggest use near greenhouses. Two pesticides and one sludge from WWTP near greenhouses tested
Cosmetics (waterproof mascara, foundation, suncream, powder)	high	4	Cosmetics are known to contain high levels of PFAS. Also high exposure because of direct application to the skin.
Dental floss (PTFE)	moderate	0	Low emission expected
Ski wax	low in NL	0	Expected to be no significant source in the Netherlands
Artificial grass	moderate	2	Newspaper article mentions PFAS containing artificial turf, worth checking

Industries and products	Expected relevance	# samples	Rationale
Fireworks	moderate	3	Fireworks are used outdoors. Paper packaging around fireworks may be water resistant.
End of life industries			
Unintended release, waste processing facilities	high	0	ILT study waste Chemours (IL&T, 2019), part of study RWS
Sewage sludge	moderate	2	WWTP sludge is sometimes used as soil treatment. This treatment is officially not allowed in the Netherlands. Four samples taken (including the one mentioned at carpet and pesticides)
Waste incineration	low-moderate	0	Part of study RWS
Carpet reuse	high	0	Flocks of the carpet industry, see carpet
Paper Sludge	moderate	1	Paper sludge is used in cement industry and as inoculum
Reference dust			
Household dust	high	3	Reference dust in regular households
Office dust	high	3	Reference dust in offices
Total amount of samples:		**129**	

3.2 Analytical plan and method

Since the group of PFAS comprises >4000 individual compounds, it is not possible to analyse for all of these individual PFAS (also since they differ in characteristics like volatility, adsorption, solubility in solvents, and because of the lack of analytical standards). The analytical plan is focussed on gaining as much information as possible in a cost-effective manner. This focusses on gaining insight into:

- Total amount of extractable PFAS.
- Presence of most important PFAS-substances.
- Presence of PFAS-precursors.
- Short- and long chain PFAS.

For this purpose, a stepwise analytical approach is being taken consisting of:

1. Screening on organic fluorine (total organic fluorine -TOF or extractable organic fluorine - EOF) using CIC (combustion ion chromatography).
2. Analysis of an extensive set of individual PFAS using LC-MS-MS (liquid chromatography tandem mass spectrometry).
3. Conduct a TOP analysis if there is a significant difference between the TOF/EOF and the individual PFAS analysis.

These analyses will be executed stepwise: based on the results of the first two steps (screening and target analysis) samples are selected for the TOP analysis. The selection of samples will be based on the detected concentrations in the previous step. Reasons to do an additional analysis can be;

- When results are different than expected.
- When concentrations of samples from the same product group vary.

The method of these analyses are explained in more detail below. The analyses have been executed by SGS in Antwerp, which is a lab with strong capabilities in speciality analysis (including PFAS) in several media.

3.2.1 Analytical plan

Screening on total organic fluorine (EOF)

The first step consists of a screening for extractable organofluorine (EOF). The analysis gives a total amount of fluorine which is bound to organic molecules, it is not specific for certain PFAS and gives no insight into chain length. It is however a good and cost-effective screening method for a large set of PFAS, including precursors which are not being detected using the standard analytical PFAS analyses that focus on individual substances.

Since a large part of the samples concerns solids, but there are also liquids being sampled, all samples have been analysed using an extraction method, to be able to compare the results between samples. Furthermore, analysing for EOF compared to TOF (total organic fluorine) has the advantage that for samples with known presence of fluoropolymers (like PTFE containing materials), the PTFE itself is not analysed, but the amount of organofluorine that can be extracted.

The detection level for EOF is in the order of magnitude of 100 µg/kg (lower if possible), which depends on the matrix analysed. This detection level is rather high compared to the individual PFAS screening, and therefore low levels of EOF will not be detected. However, the analysis does detect whether a significant amount of PFAS is present.

The EOF analysis is not specific for the group of fluorinated organic surfactants that have been the main focal point of environmental and health concerns. EOF is a measure for all extractable organofluorines including, for example, non-ionic fluorohydrocarbons. Therefore, with this method, also other fluorinated compounds (which are not PFAS) can be detected. To gain more insight in the presence of individual PFAS, samples are subjected to the PFAS-target analysis and (a selection to) the TOP-analysis.

Individual PFAS (PFAS-target analysis)

All samples have been subjected to analysis of individual PFAS using LC-MS-MS (liquid chromatography tandem mass spectrometry). Looking at individual PFAS, a set of PFAS has been chosen which can be analysed by an analytical laboratory with established methods (table 2), this includes the PFSA’s, PFCA’s, fluorotelomer sulfonic acids (FTS), polyfluoroalkyl phosphate esters (PaP), perfluoroethers and some other precursors.

Fluorotelomer alcohols are not included in the set of individual analysis, since they are volatile and require a different type of analysis. However, they will be detected using EOF and TOP.

Table 2 List of PFAS included in analysis.

PFAS Group	Individual PFAS	Short/long chain	Remarks	Detected with EOF	Detected with TOP
Perfluorosulfonic acid (PFSA's)	PFBS (C4) PFPeS (C5) PFHxS (C6) PFHpS (C7) PFOS (C8) PFDS (C10)	Short Short Long* Long Long Long	Standard PFAS	Yes	Yes
Perfluoro carboxylic acids (PFCA's)	PFPrA (C3) PFBA (C4) PFPeA (C5) PFHxA (C6) PFHpA (C7) PFOA (C8) PFNA (C9) PFDA (C10) PFUnDA (C11) PFDoDA (C12) PFTriDA (C13) PFTeDA (C14) PFHxDA (16) PFOcDA (18)	Short Short Short Short Short Long Long Long Long Long Long Long Long Long	Standard PFAS	Yes	Yes
Fluortelomersulfonic acids	4:2 FTS 6:2 FTS 8:2 FTS 10:2 FTS	Short Short Long Long	Used as replacement for PFOS/PFOA in AFFF and galvanic industry	Yes	Yes
Fluortelomeralcohols	- -		The fluorotelomer alcohols are not foreseen in this analysis, since they are analysed using a different type of analysis (volatile PFAS). They are being used for water repellence in different types of products.	Yes	Yes
Polyfluoroalkyl phosphate esters ((di)PaP) and phosphinates	6:2 PAP + 6:2 diPaP diSAMPAP 6:6 PFPI 6:8 PFPI 8:8 PFPI	Short Long Short Long Long	Used in e.g. paper industry	Yes	Yes
Other precursors	PFOSA PFOSAA N-MeFOSA N-EtFOSA FOSAA N-MeFOSAA N-EtFOSAA	Long Long Long Long Long Long Long	Precursors, several of them also being analysed in the standard set of parameters for soil investigations. N-EtFOSAA is often being detected in sediments in the Netherlands.	Yes	Yes
Perfluoroethers	GenX ADONA F53B: major component (9Cl-PF3ONS)	-	GenX is a processing aid (replaces PFOA) in Teflon production. Other perfluoroethers are Adona (3M) and F53B (China)	Yes	No

PFAS Group	Individual PFAS	Short/long chain	Remarks	Detected with EOF	Detected with TOP
	F53B: minor component (11Cl-PF3OUdNS)				
Other PFAS	FHUEA FDUEA	-	Partly fluorinated and/or unsaturated PFAS	Yes	Unknown

* For PFSA's, PFHxS is considered as a long-chain compound. For PFCA's and PFAS-precursors, the C6-compounds are considered to be short-chain compounds.

TOP analysis

When a large difference is observed between the results of the EOF analysis and the sum of individual PFAS, other PFAS are likely to be present but their identity is unknown. For an evaluation of the possible emission of all PFAS, it is important to know whether these unknown PFAS can (bio)transform into the persistent end products PFAA (perfluoroalkyl acids, like PFOS, PFOA and similar). It is also important to know whether these are long chain PFAS (subjected to current restrictions) or short chain PFAS (mobile and persistent PFAS). These compounds might be e.g. fluorotelomer alcohols, which have not been analysed using the PFAS-target analysis, or PFAS with a different head group, which makes it difficult to detect in a PFAS-target analysis.

The TOP analysis (total oxidizable precursors) has been specifically developed to identify the PFAS present in samples where total (extractable) organic fluorine could not be explained by the individual PFAS measurements. By means of an oxidative digest, the slow aerobic biodegradation of precursors in the environment is mimicked and the precursors are rapidly transformed into the end-products, PFAA's, which are analysed by LC-MS/MS before and after the oxidative digest. The major benefits of the TOP analysis are that it gives insight in the chain length of the PFAS precursors and provides a measurement of the total PFAS present.

A smaller selection of the total amount of samples (20) has been subjected to a TOP analysis. The selection of samples for the TOP analysis is therefore based on the results collected: if the difference between the EOF and the target PFAS analysis is high, an additional TOP analysis can be conducted to explain a (much) higher share of the EOF concentration than can be done with PFAS target analysis only.

3.2.2 Method

For all analyses, the sample extraction is similar: (when available) 5 grams of sample was extracted with 40mL LC-MS grade methanol (MeOH). Dependent of the sample characteristics, this sometimes needed to be modified to a different volume (e.g. carpet fibres had to be extract with a higher volume of MeOH to ensure complete submersion of the sample). After extraction the obtained extract was filtered on a 0.22 µm filter (mixed cellulose ester) to remove particulates.

Extractable organic fluorine:

- 10 mL of the extract was taken off and evaporated until near dryness. The remainder was diluted with 0.5 mL MeOH and brought over in a sampling boat, after which the content was pyrolyzed in the horizontal furnace under a constant humid Ar/O2 gas stream. The resulting combustion gasses where scrubbed by the gas absorption unit. After the combustion has been completed a part of the absorption solution is automatically injected into the Ion chromatograph to determine the fluorine content.
  - Combustion: Mitsubishi Chemical Analytech HF-210 (horizontal furnace) + ASC-270LS (automated sample changer) + GA-211 (gas absorption unit)
  - Analysis: Thermo ICS-2100 Ion chromatograph, equipped with AG-18 Guard column and AS-18 Analytical column and conductivity detector

PFAS Target analysis:

- 0.5 mL of the extract was taken off, spiked with a mixture of labelled 13C-PFAS compound and diluted to 1 mL with water. This analytical solution was analysed by UPLC-MS/MS (Water Xevo-TQ-XS UPLC-MS/MS).

TOP analysis:

- 1 mL of the analytical solution was taken off and the methanol was evaporated off, since it would interfere with the TOP-reaction. To the residue 50 mL water was added and 13C8-PFOSA is added to monitor the oxidation. A strong oxidant (sodium persulfate) is added to the solution and the solution is made alkaline by addition of sodium hydroxide until pH 13. The mixture is heated during 6 hours on a hotblock at 85°C. After oxidation the solution is neutralized until pH 6.5, before cooling it overnight to stop the reaction. After the resting period the extraction standards for quantification are added and a solid-phase extraction (SPE) sample clean-up is performed. The solution is analysed by UPLC-MS/MS. The goal with this approach is to determine whether the organofluorine compounds, which were not detected by the target PFAS-analysis can be converted to the detectable PFAS compounds. This is also the reason why 13C8PFOSA is added prior to oxidation: when the oxidation is successful, the 13C8-PFOSA will be completely converted to 13C8-PFOA.

4 RESULTS

In total, 129 samples have been collected and analysed for EOF and PFAS-target analysis. A selection of the samples (20 samples) has been analysed for TOP. A complete overview of the analytical data of the EOF and PFAS-target analysis has been given in Appendix A. The analytical data of the TOP-analysis is given in Appendix B. In this chapter, the results are summarized. The results have been evaluated in Chapter 5.

4.1 EOF and PFAS-target data

The PFAS-target data concerns 45 individual PFAS. These PFAS have been categorized into short- and long-chain PFCA's, PFSA's, precursors, perfluoroethers and other PFAS (see Table 2).

In the figures below, overviews of all the data are given, per type of product (product, process, recycling, waste), sorted from low concentrations to high concentrations. These figures focus at three types of results: PFAS-target data, extractable organic fluorine, and the ratio of these two; the percentage of organic fluorine that can be explained by the PFAS-target data (a ratio of 0.68 fluorine/PFAS-target has been used for this calculation[5]). Note that the scale between the EOF and PFAS-target data differs. The detected EOF concentrations are significantly higher than the PFAS-target data.

EOF data is based on the extraction of organofluorine compounds using a methanol extraction. For two types of samples this may result in an overestimation of the amount of extractable organic fluorine:

- Fluoroelastomers have a low compatibility with methanol. Parts of the elastomer itself may be dissolved into methanol. This may result in a high concentration of extractable organic fluorine.
- Water can be mixed with methanol. Therefore, water-based samples might get mixed with methanol, and both organic and inorganic fluorine will be detected.

Both types of samples are highlighted in Appendix A.

It has to be noted that EOF is a measure for all extractable organofluorines (see paragraph 3.2.1). This doesn't necessarily mean that PFAS are present that are the focus of this investigation.

[5] The PFAS-target compound each contain a different fraction of fluorine. To compare the sum PFAS-target with the EOF concentrations a ratio of 0.68 has been used, based on the fluorine ratio in the perfluorinated backbone, multiplied by 90% to account for the non-fluorinated head of the molecule. This number is used for an indicative comparison.

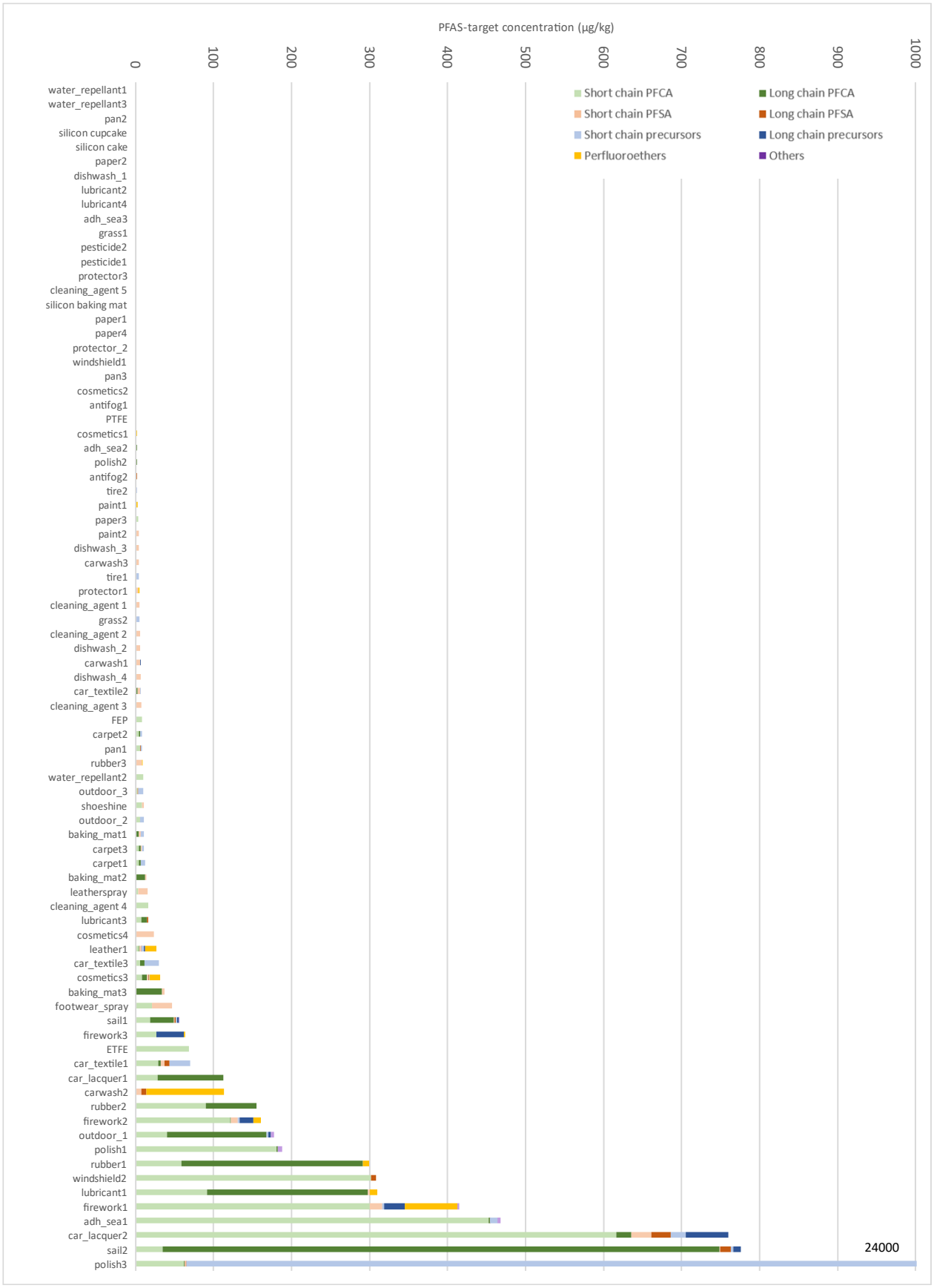

Figure 3 Results PFAS-target analysis for the for the samples categorized as 'product'

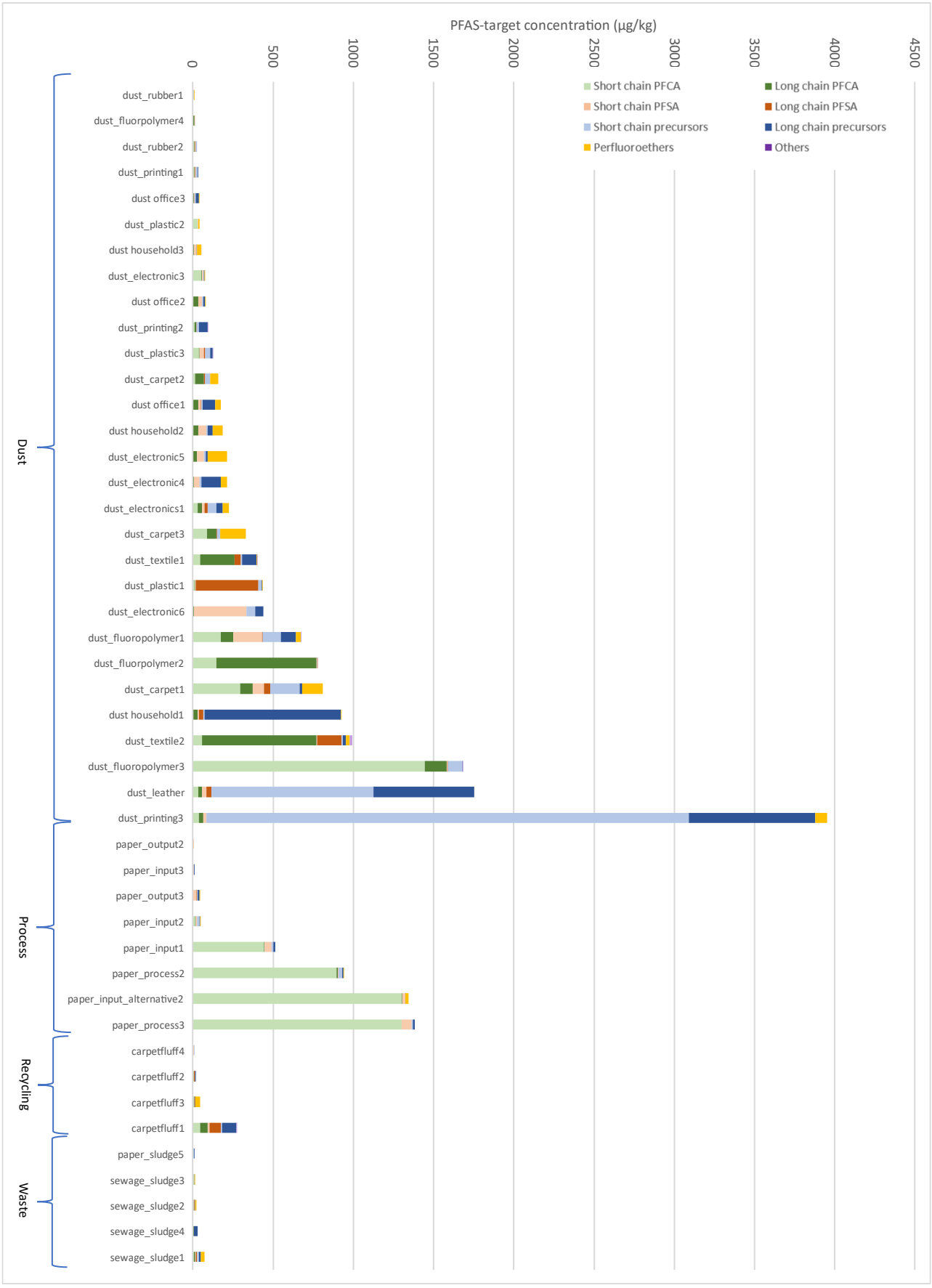

Figure 4 Results PFAS target analysis for the samples categorized as 'dust', 'process', 'recycling' and 'waste'

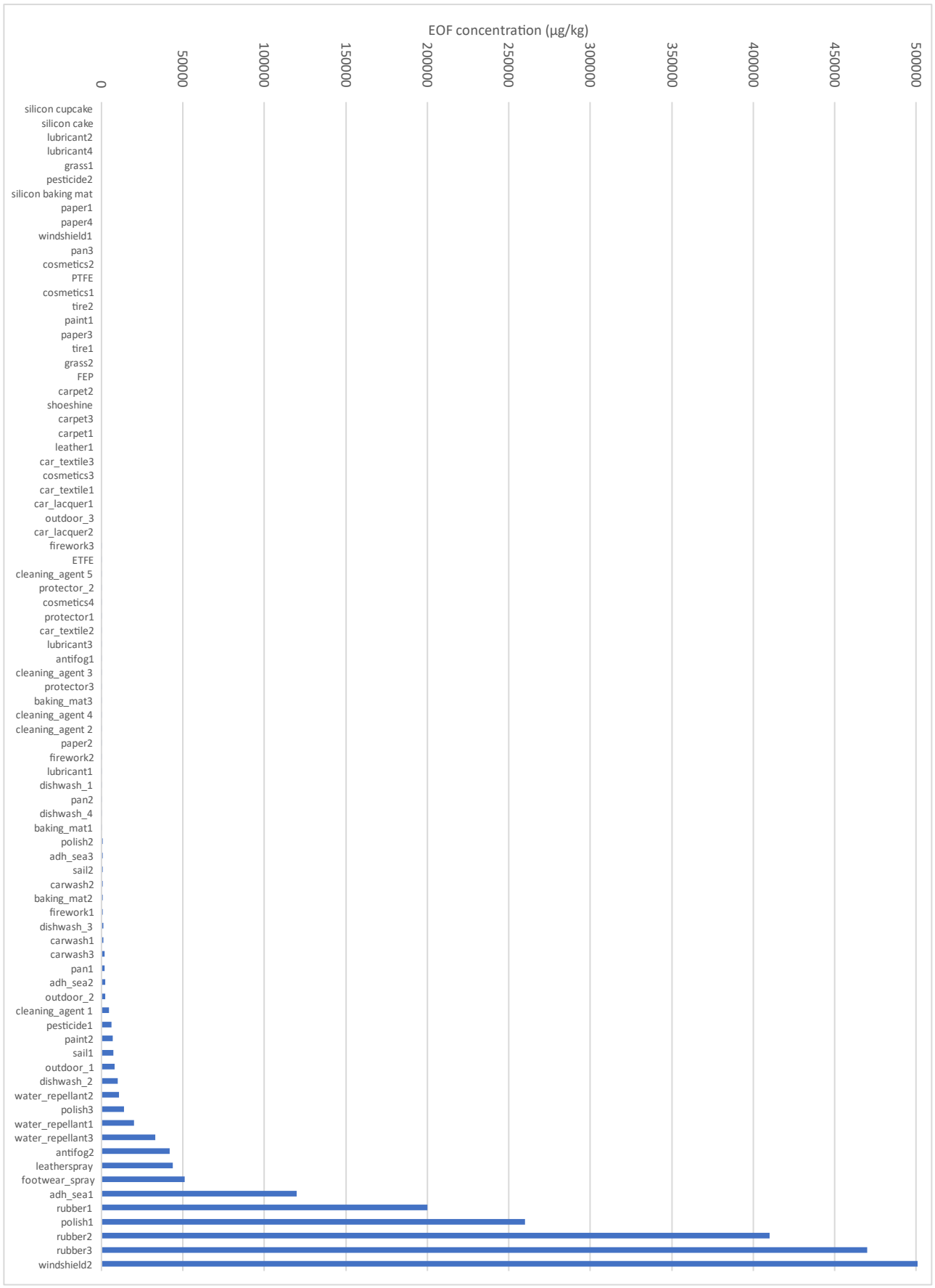

Figure 5 Results EOF analysis for the for the samples categorized as 'product'

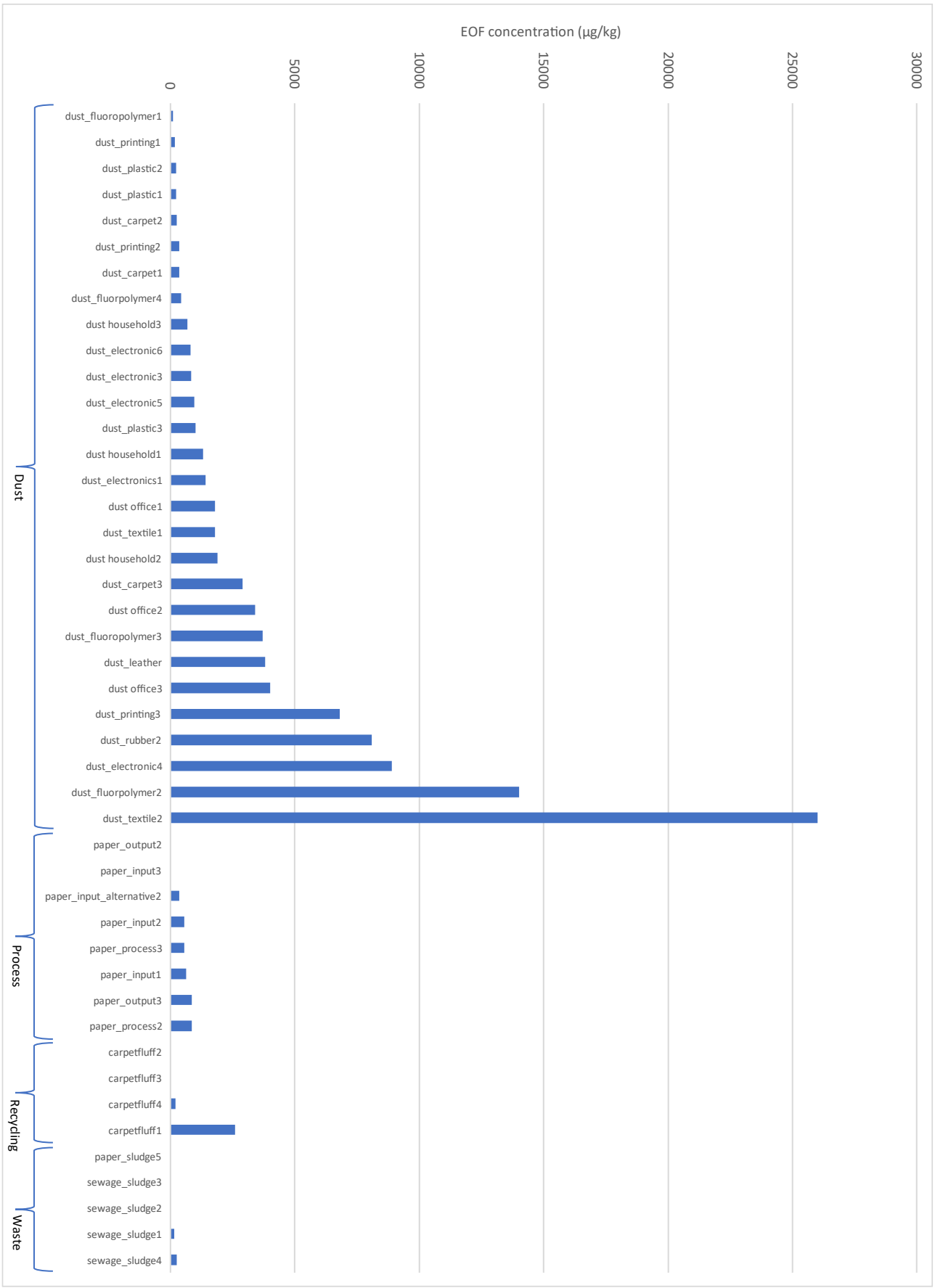

Figure 6 Results EOF analysis for the samples categorized as 'dust', 'process', 'recycling' and 'waste'

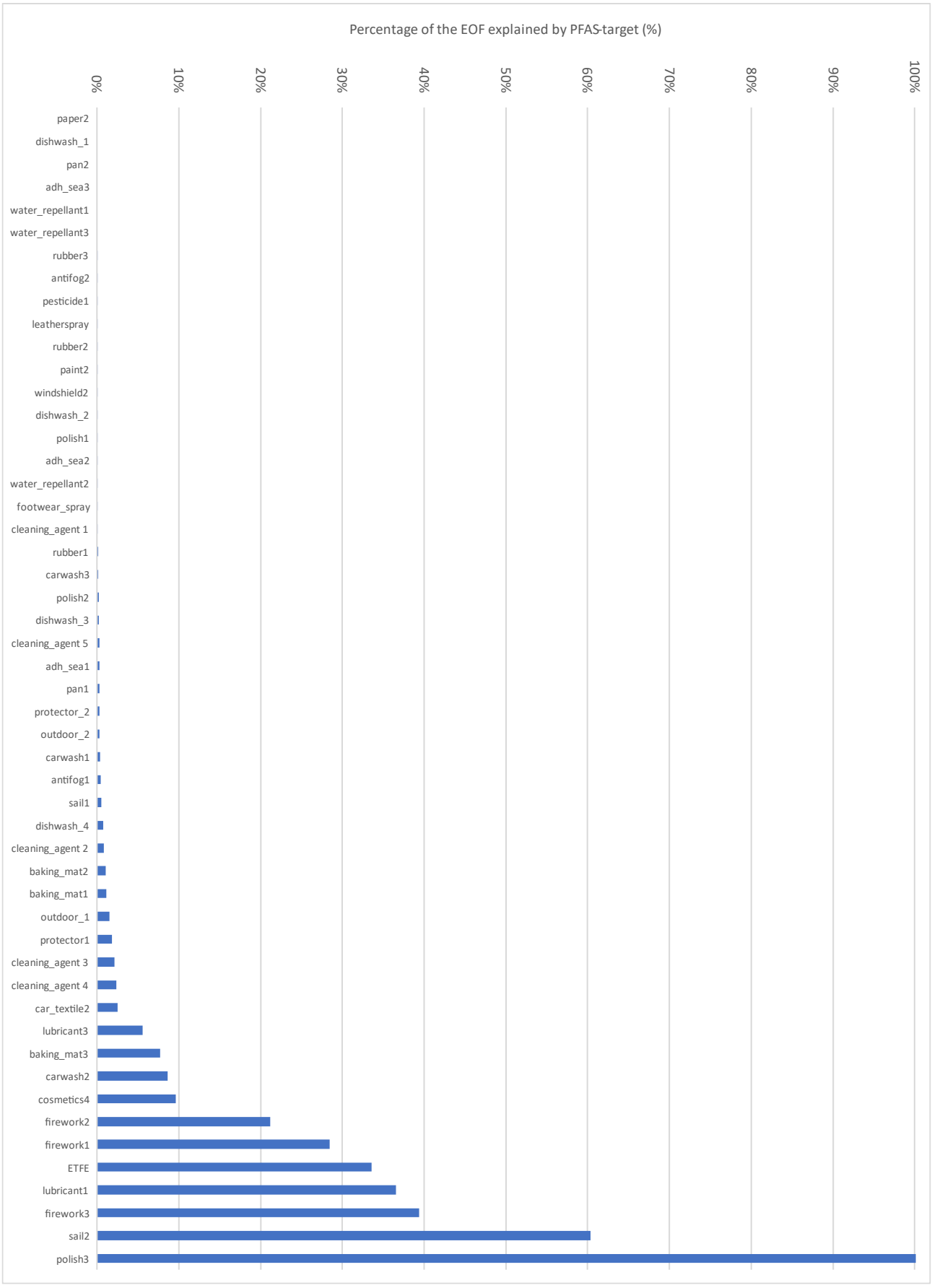

Figure 7 Results percentage of EOF data that can be explained by the PFAS-target data for the samples categorized as 'product'

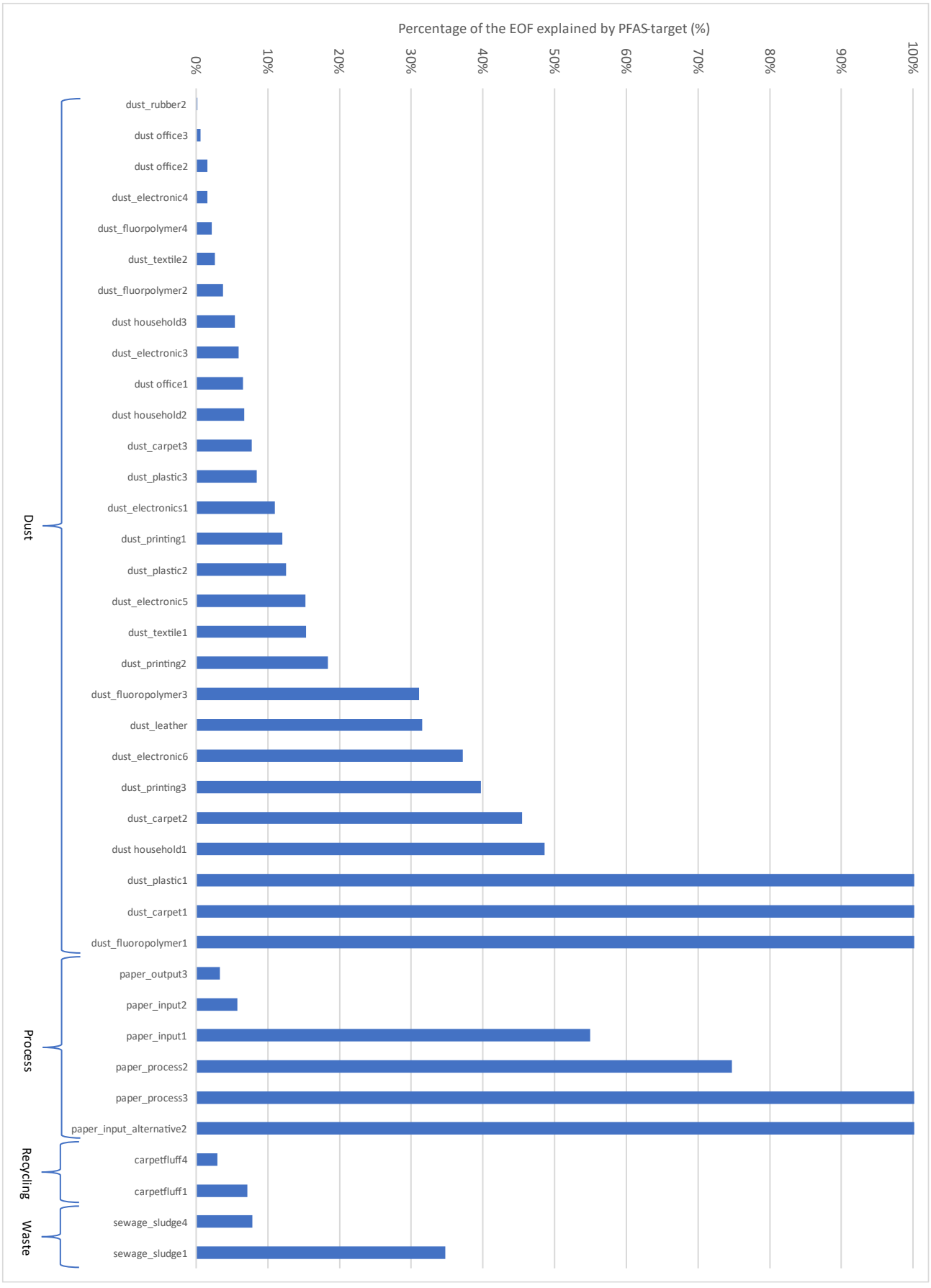

Figure 8 Results percentage of EOF data that can be explained by the PFAS-target data, for the samples categorized as 'dust', 'process', 'recycling' and 'waste'

4.2 Results TOP-analysis

The results of the TOP-analysis are shown in Appendix B and in the figures below. 20 samples have been selected for TOP-analysis, based on a large difference between the EOF-results and the PFAS-target results.

The TOP-analysis mimics the transformation of PFAS-precursors into PFCA's and PFSA's (the dead-end daughter products). The difference between the concentrations after and before TOP is caused by the oxidation of PFAS-precursors.

Out of the 20 samples tested:

- 12 samples show a strong increase of PFAA's. This indicates the presence of PFAS-precursors. In the product samples this mostly concerns short chain PFAS like PFPeA (C5) and PFHxA (C6).
- In 4 samples, the PFAA-concentration before and after oxidative digestion stays roughly the same. There is no clear indication for additional PFAS-precursors in these samples.
- In 4 samples, the PFAA-concentration seems to decrease after TOP oxidation, this can have the following reasons:
  - There is a difference in the detection limits before and after TOP oxidations. After TOP oxidation, the detection limit is higher, and only the results above detection limit are shown.
  - Other PFAS are present, which do not show up in the TOP-analysis; e.g. PFPrA in paper.
  - In some other samples PFBS seems to disappear. There is no good explanation for this effect, but it might be the case that some PFAS disappear during the evaporation phase of the TOP-analysis. The samples have been reanalysed which showed the same results.

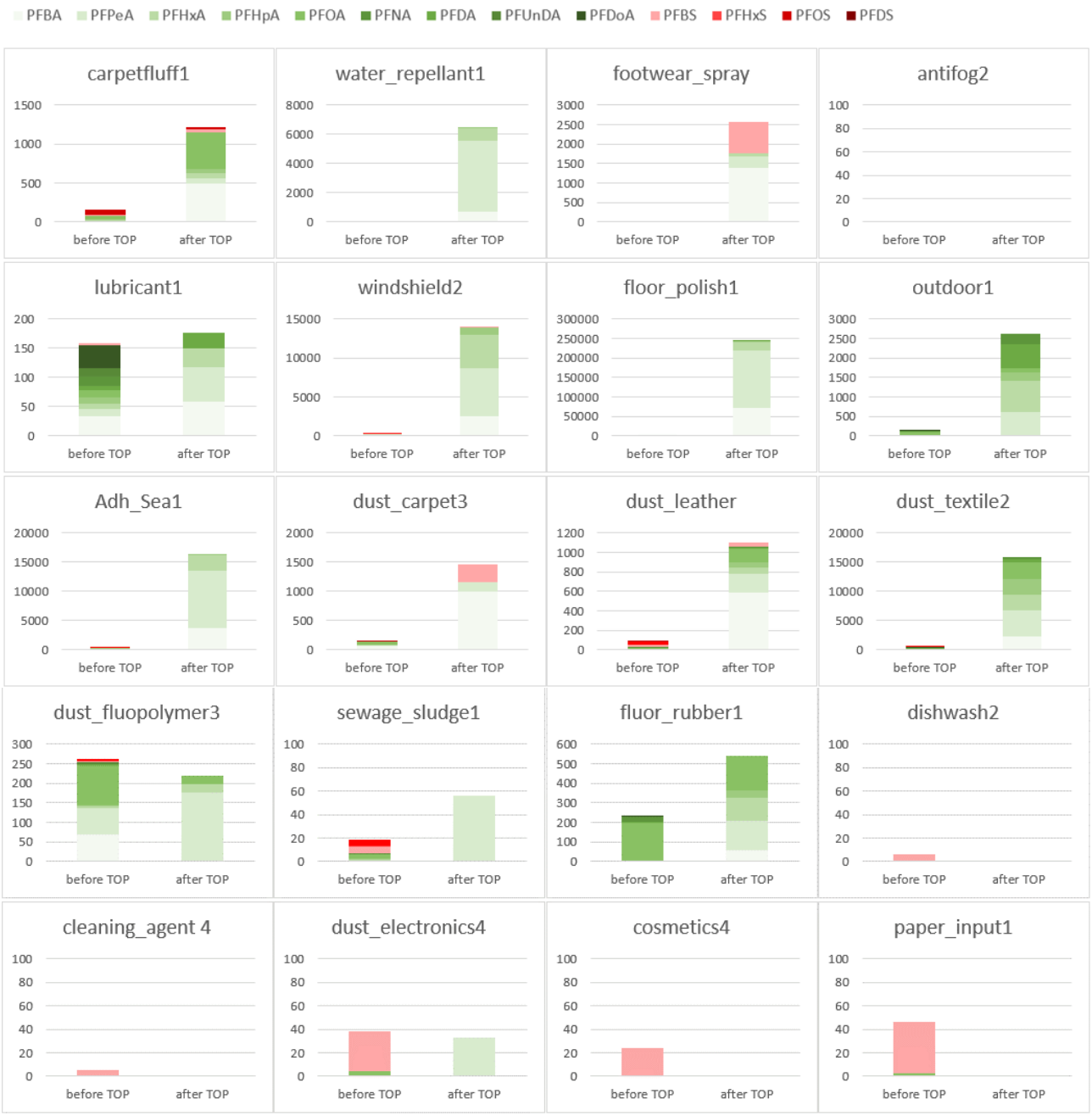

Figure 9 Results TOP analyses. Values on the y-axis are given in µg/kg

5 EVALUATION OF THE RESULTS

It has to be noted that this investigation concerns a wide screening of materials.129 different samples have been analysed, which vary widely in composition, type of material and application. The results can be used to give a general idea about the presence of PFAS in different applications, but cannot be directly applied to an entire category of material, since the PFAS-concentrations within the different categories vary a lot and only a limited amount of 2-4 samples have been analysed per type of application.

5.1 General observations

The samples have been categorized into 5 different categories:

- Product samples. This category concerns a wide variety of mainly consumer products, like e.g. textile sprays, kitchen utensils, cleaning agents, make up etc. Also some products have been included which are used in different processes, like fluoropolymers, fluoroelastomers.
- Dust samples. These samples are taken as a screening matrix, to check whether increased concentrations are measured. This is done at different factories, where it is not clear whether or not PFAS are being used, but also in offices and households to check whether general wear of carpets or the use of sprays etc. can lead to PFAS concentrations.
- Process samples. This category concerns mainly paper pulp samples from different stages from the paper production process at three paper factories. This also includes some paper streams which concern recycled paper.
- Recycling samples. This category concerns recycled carpets and textiles.
- Waste samples. This category concerns wastewater treatment sludge.

5.1.1 Product samples

52 out of 83 product samples (63%) contain detectable EOF concentrations (above 100 µg/kg). EOF is very high in oil and water repellent applications like textile and leather sprays, joint protector for bathrooms and floor polish. The EOF is extremely high in the three fluorinated rubber rings tested (FKM, fluoroelastomers, Viton). Fluorinated rubbers are used in several industries as rubbers, seals, hoses, o-rings and valve packings. In these samples, it is plausible that part of the rubber has been solubilized during the extraction with methanol, and the fluorinated rubber itself is being detected. Nevertheless, in these samples relatively high amounts of PFCA's have been detected.

Extractable organic fluorine concentrations are usually a few orders of magnitude higher than the PFAS-target concentrations. This can also be seen in figure 7. In most cases, the PFAS-target concentrations explain less than 10% of the EOF-concentrations.

From the 83 product samples, 23 samples show a PFAS target concentration above 25 µg/kg (28%) of which 14 samples show a PFAS target concentration above 100 µg/kg (17%). The PFAS target compounds in the samples with the highest concentrations mainly concern short- and long chain PFCAs. The sample with the highest concentration PFAS-target, being a floor polish, contains mainly short chain precursors, in this case being 6:2 PaP and diPaP-compounds.

The highest concentrations are found in:
- Oil and water repellent applications, floor protection and polish, windshield treatment, joint protector for bathrooms, car paint.
- Fluorinated rubber rings (FKM, fluoroelastomers).
- Paper used in fireworks.
- Fabrics for sunscreens and sails (awning).

Relatively low concentrations have been found in:
- Silicon baking forms.
- Artificial grass.
- Tires.
- Non-PTFE containing lubricants.

Within certain product categories, the concentrations can vary highly between samples.

The fact that PFAS-target compounds usually explain less than 10% of the extractable organic fluorine, indicates that other PFAS and PFAS-precursors might have been used in these products.
For 13 product samples, this has been tested using the TOP-analysis. Of these samples, 7 samples show an increased concentration of PFCAs after the TOP-analysis. This indicates that other PFAS precursors are present, which are in most cases short chain precursors (PFHxA and PFPeA). One of the samples shows only slightly increased PFAS-levels (lubricant). The other 5 product samples show no indication for PFAS precursors. The cheap antifog shows no increase of PFAAs, while it does contain a very high EOF. Probably other fluorinated compounds are present which do not transform into PFAAs or anorganic fluorine that has been extracted due to the water-based product. The same observation is seen for the dishwashing agent and cleaning agent.

Overall, especially products with water and dirt resistant properties contain PFAS.

5.1.2 Dust samples

Dust has been collected and analysed, to serve as a screening matrix. In dust samples, generally relatively high concentrations of PFAS-target have been detected (several hundreds to several thousands µg/kg). Furthermore, a higher fraction of the EOF can be explained by the PFAS-target compounds (compared to products). This indicates that either more target compounds are present in the original product, or that degradation products of precursors are collected in dust. Concentrations within the dust categories can vary highly among samples.

PFAS-target concentrations of 100-200 µg/kg can be considered as background concentrations. These concentrations have been detected in household dust and dust at several offices. One of the household samples showed a high concentration of N-EtFOSAA, but this could possibly be explained by a new carpet bought a couple of months before sampling. Therefore, the results of this sample are not considered to be background concentrations.

In the figure below, the results of the EOF-analysis (left), PFAS-target analysis (middle) and percentage of EOF (right) that could be explained by PFAS-target, is given. When looking at dust as a screening matrix, clearly elevated concentrations are found (left and middle figure) in the fluoropolymer industry, textiles and carpets. In the printing industry, the concentrations EOF and PFAS-target are low, except at the printing room at one of the sampled offices. It is not clear what causes the high concentrations in this sample, there can be several sources, e.g. the ink, paper, circuit boards used (no carpets or floor polishes). For the electronic industry, plastic industry and rubber industry, the concentrations are not very clearly elevated, but might still indicate the use of PFAS.

Almost all dust samples, including reference samples, contain PFOA. Exceptions are one printing industry sample, one rubber industry sample and one plastic industry sample. PFOA has just recently been restricted within REACH (4 July 2020), it is expected that PFOA in dust originates from the degradation of PFOA-precursor-containing products, which slowly transform in the environment to PFOA.

The five dust samples that have been subjected to the TOP analysis all show an increase in PFAS concentration, which indicates the presence of PFAS precursors.

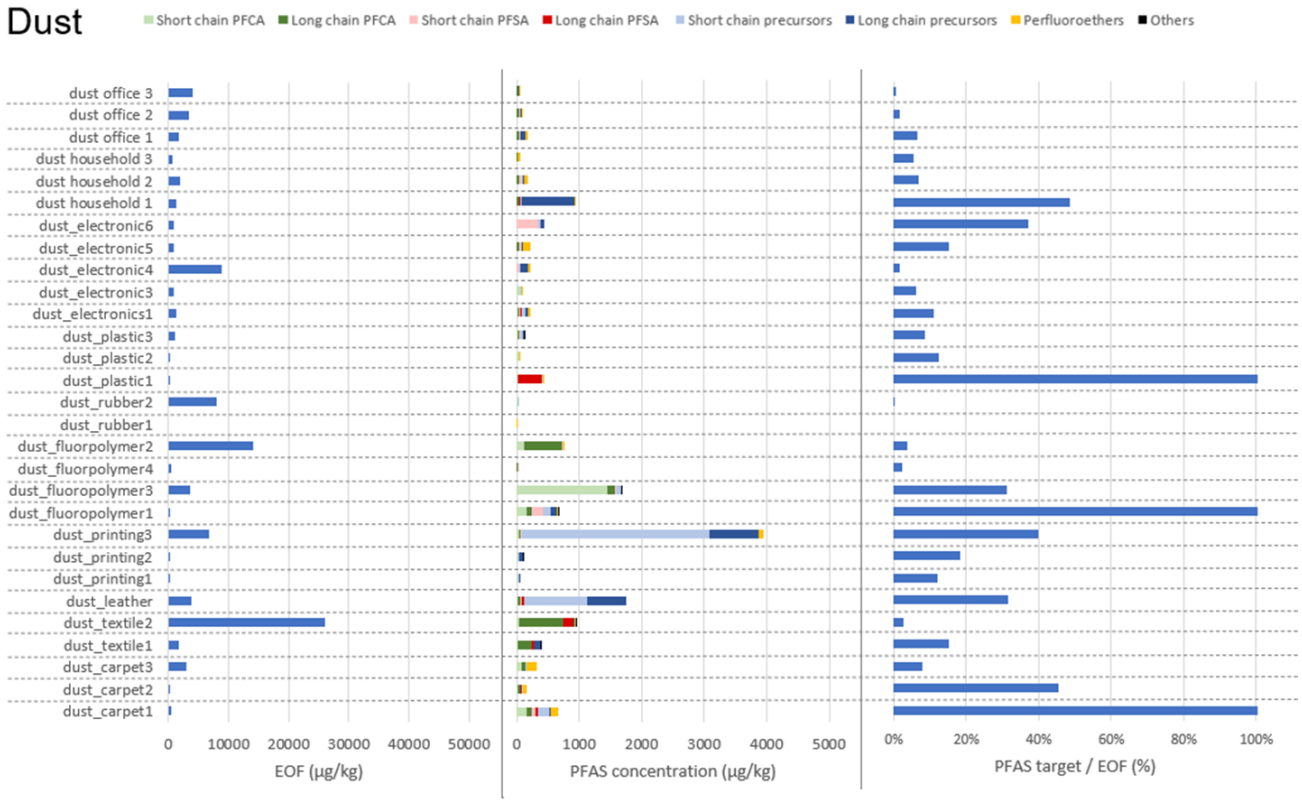

Figure 10 Results EOF, PFAS target analysis and percentage EOF found with PFAS target in all dust samples

5.1.3 Process samples

The process samples selected concern mainly paper pulp samples from different stages from the paper production process at three paper factories. This also includes some recycled paper pulp.

The paper pulp samples contain relatively high PFAS-target concentrations in 4 out of 8 samples (500-1300 µg/kg). This can, for the major part, be explained by short chain PFCA's, in this case PFPrA (C3-PFCA).

PFOS has not been detected in the paper pulp samples. Recently a survey, conducted by Rijkswaterstaat (Jans en Berbee, 2020), showed elevated levels of PFAS in effluent of paper factories, including PFOS, which has a very stringent environmental quality standard (0.65 ng/l annual average value for surface water). It was suspected that the increased levels were caused by the use of recycled paper. Based on the results found in this study, (other) PFAS have been detected in the paper pulp samples. A clear link with PFOS is currently still missing (unclear presence of precursors).

5.1.4 Recycling samples

The recycling samples concern recycled carpet and textile fluff. The PFAS-target concentrations in these samples are relatively low, except for the carpet fluff samples, which originates from a carpet recycling factory. This sample has a relatively high PFAS-target concentration and EOF concentration. The sample also has been subjected to the TOP analysis. These results show lower PFCA concentrations (mainly PFOA), and therefore do not completely confirm the PFAS-target data.

5.1.5 Waste samples

The waste samples concern sludge from wastewater treatment plants. In all sewage sludge samples, PFAS have been detected, in the order of magnitude of 100 µg/kg. EOF concentrations are in the same order of magnitude.

5.2 Observations per type of industry

5.2.1 Textile, carpet and leather

In the category textile, carpet and leather, high concentrations of PFAS-target compounds and EOF have been detected in awning cloth. In these types of samples concentrations of more than 25 ppb (µg/kg) PFOA have been detected. However, the detected concentrations in carpets and leather (1 sample) are relatively low. Outdoor clothing has not been tested as considerable data are already available in the literature.

The detected concentrations of organic fluorine are especially high in fabric and leather treatment sprays and polishes (up to 0.5 g/kg). Usually in these sprays the PFAS-target compounds are not detected or can explain less than 1% of the EOF. Using the TOP-analysis it has been confirmed that this might be explained by the presence of PFAS-precursors.

In all dust samples at the textile, carpet and leather factories significant amounts of PFAS-target compounds are being detected. Also in dust, PFAS-precursors have been detected by the TOP-analysis

The PFAS-target and EOF concentrations in the recycled carpet samples are relatively low (used for lining ponds, drainage tubes and horse stables), except for the carpet fluff sample, which was obtained directly at a recycling facility.

The sewage sludge close to several carpet factories shows higher concentrations of PFAS-target than other sewage sludge samples.

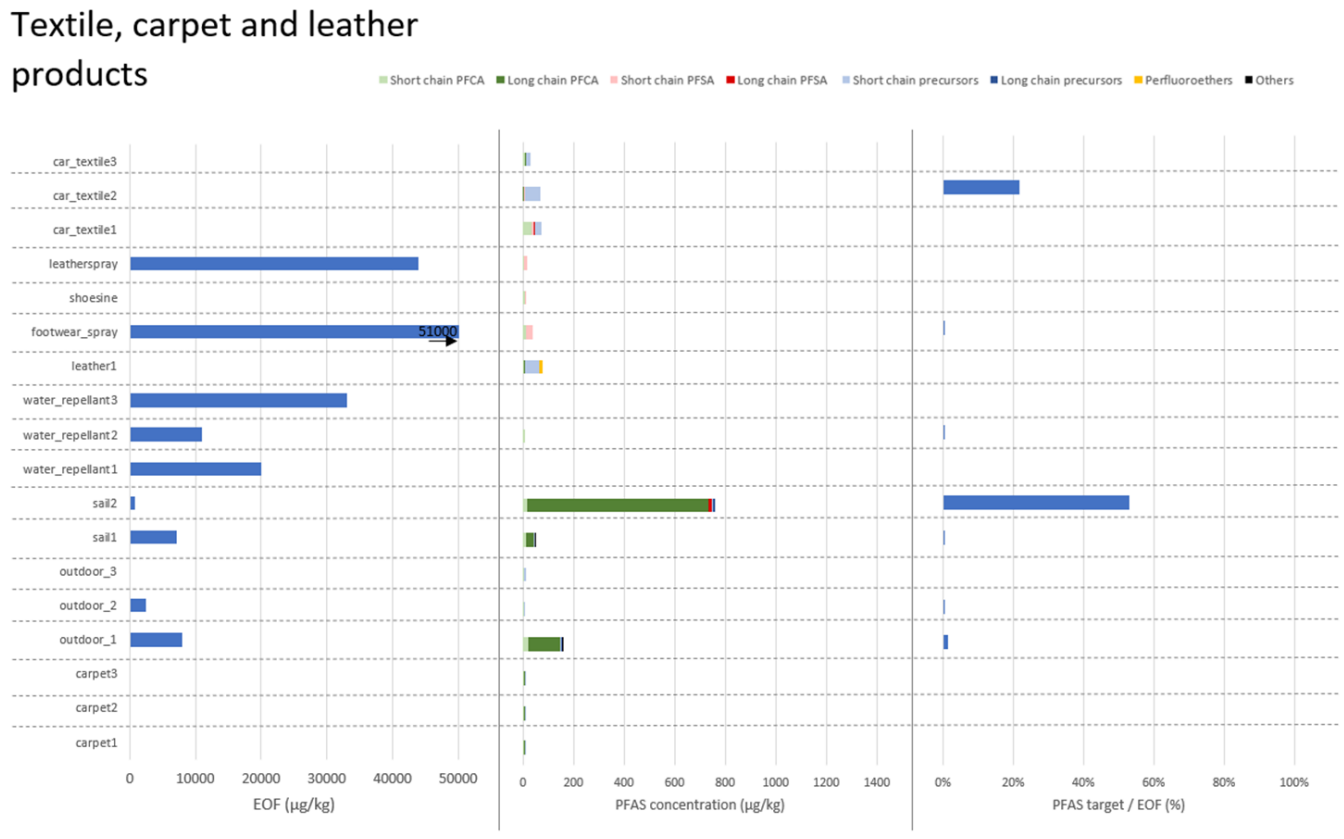

Figure 11 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for textile, carpet and leather products

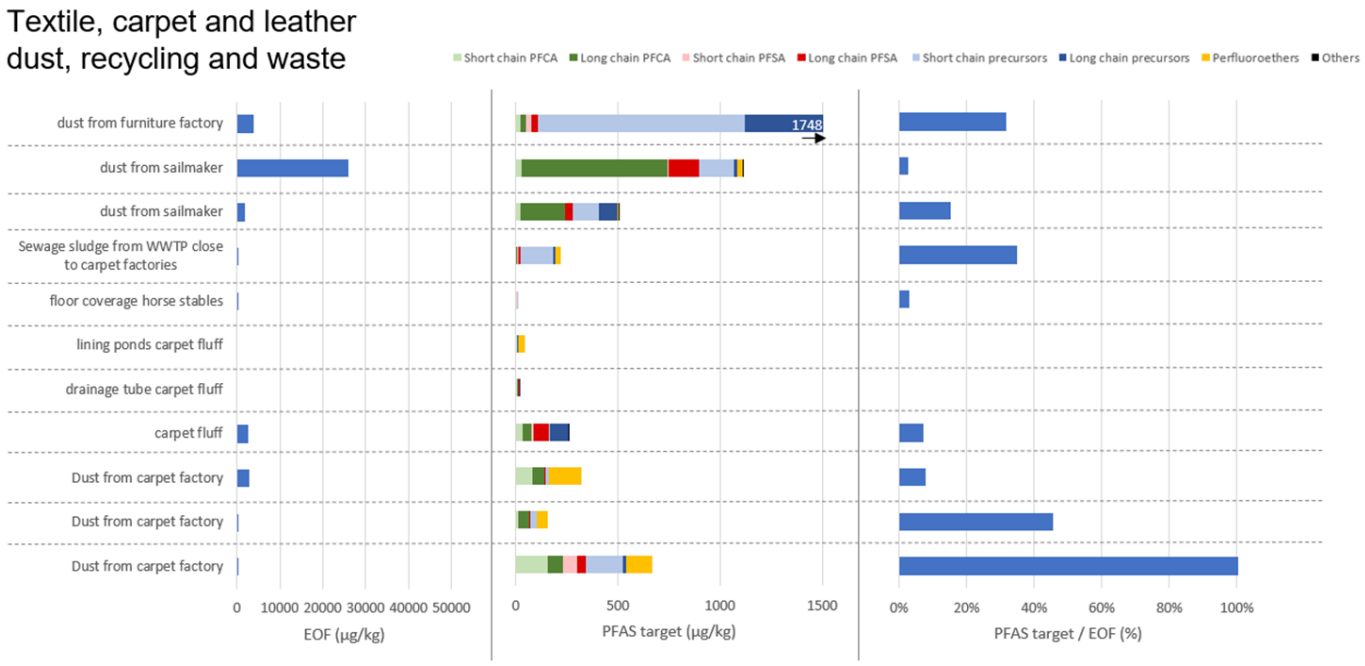

Figure 12 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for textile, carpet and leather industry dust, recycling and waste samples

5.2.2 Paper

In the general papers tested (paper tablecloth, glossy paper, newspaper), low PFAS-target concentrations and EOF have been detected. No food contact materials were tested, since these are being tested in a study that is currently being performed by NVWA.

In another study concerning the presence of PFAS in effluents from different industries, the paper industry was highlighted as a possible source, elevated concentrations of PFSA's and PFCA's have been detected in 3 out of 4 tested wastewaters from paper factories (Jans and Berbee, 2020). In this study it was suggested that recycled paper might contribute to the PFAS load in the wastewater. Therefore, we tested 8 paper pulp samples from 3 different paper factories (not necessarily being the same as in the study of Jans and Berbee). These concerned paper pulp from the recycled paper input and some samples halfway the paper production process. Also an input of alternative sources has been tested, which concerned recycled food contact materials and other materials.

In the tested pulp samples several PFAS-target compounds are detected (including several precursors like fluorotelomersulfonates). The major contributor to the sum-PFAS-target is PFPrA (perfluoropropanoic acid, C3 PFCA), concentrations up to 1300 µg/kg PFPrA have been detected. This is a very short chain PFCA which usually just falls outside the general PFAS-target suite (which starts at C4). Also the paper samples of the fireworks contain PFPrA.

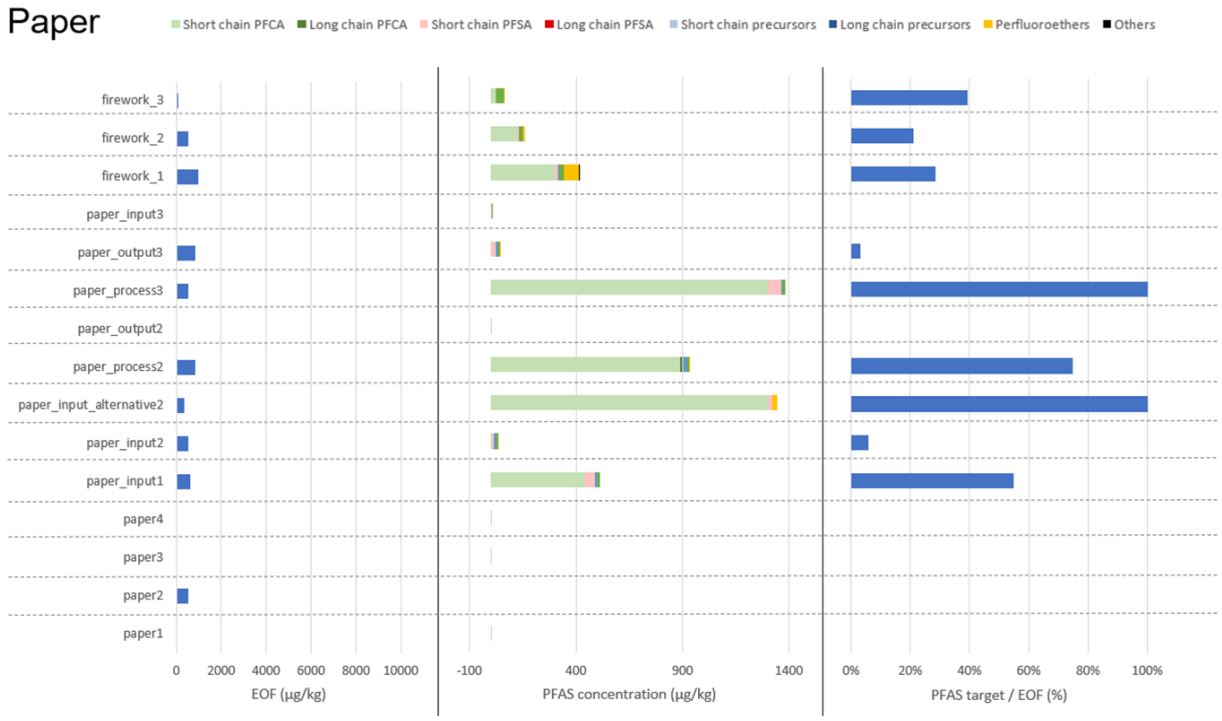

Figure 13 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for paper samples

5.2.3 Food related

In the category food related, PTFE coated pans, PTFE baking mats and silicone baking forms have been tested. The detected PFAS-target concentrations in the pans and silicone baking forms are relatively low. In the extract of the baking mats, which are used in ovens and on barbecues, PFAS have been detected, mainly PFOA. Since this comes into contact with food, the detected concentrations are of concern (in one of the samples the PFOA-concentration is > 25 µg/kg).

5.2.4 Cleaning agents

In the cleaning agents, relatively low concentrations of PFAS-target have been detected. The EOF concentrations might indicate the presence of PFAS-precursors. This has been tested using the TOP analysis for two of the samples. No additional PFAS showed up in the TOP analysis, indicating that no PFAS-precursors are present. Since the cleaning agents are water based, the elevated EOF levels might be caused by inorganic fluorine (mixing of the agent with the extraction solution), or other PFAS are present which are not detected using the target and TOP analysis.

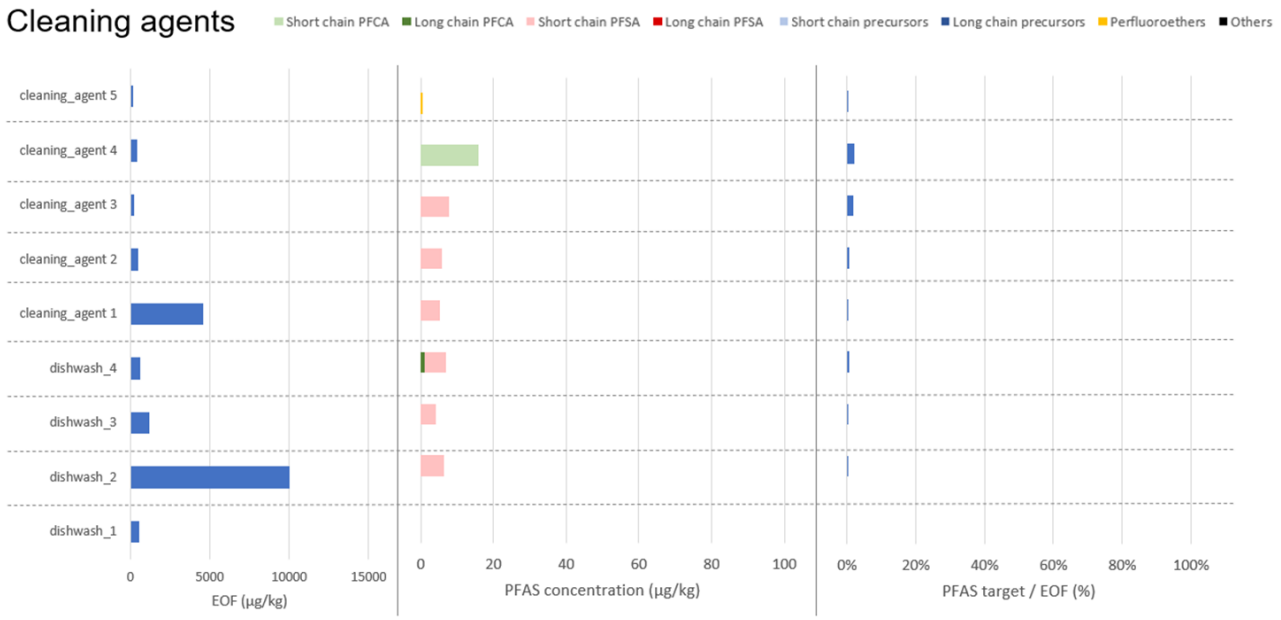

Figure 14 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for cleaning agent samples

5.2.5 Coatings and polishes

Several polishing materials contain either a high PFAS-target concentration or high EOF concentrations. Especially the floor polishes show high concentrations. For one of them, the EOF concentration is for 100% caused by 6:2 (di)PaP compounds. Another floor protector shows a very high EOF level, which is confirmed by the TOP-analysis, which shows that 64% of the EOF can be explained after TOP-oxidation (mainly C4-C6 compounds). In both of these floor protector products short chain PFAS-precursors are being used.

One of the anti-fog treatments shows very high levels of EOF, which do not show up in significant levels in the PFAS-target analysis, nor in the TOP-analysis. This might be caused by inorganic fluorine (the anti-fog treatment is water-based), but might also be caused by PFAS which are not detected using the TOP-analysis. In an indicative PFAS-analysis in an earlier stage of this project FDEA was detected (2-perfluorodecyl ethanoic acid) which is an indication for other PFAS being present.

Other products like paints, carwash treatment and shower cabin protector also show increased levels of PFAS-target or EOF, however, these were not tested using the TOP-analysis.

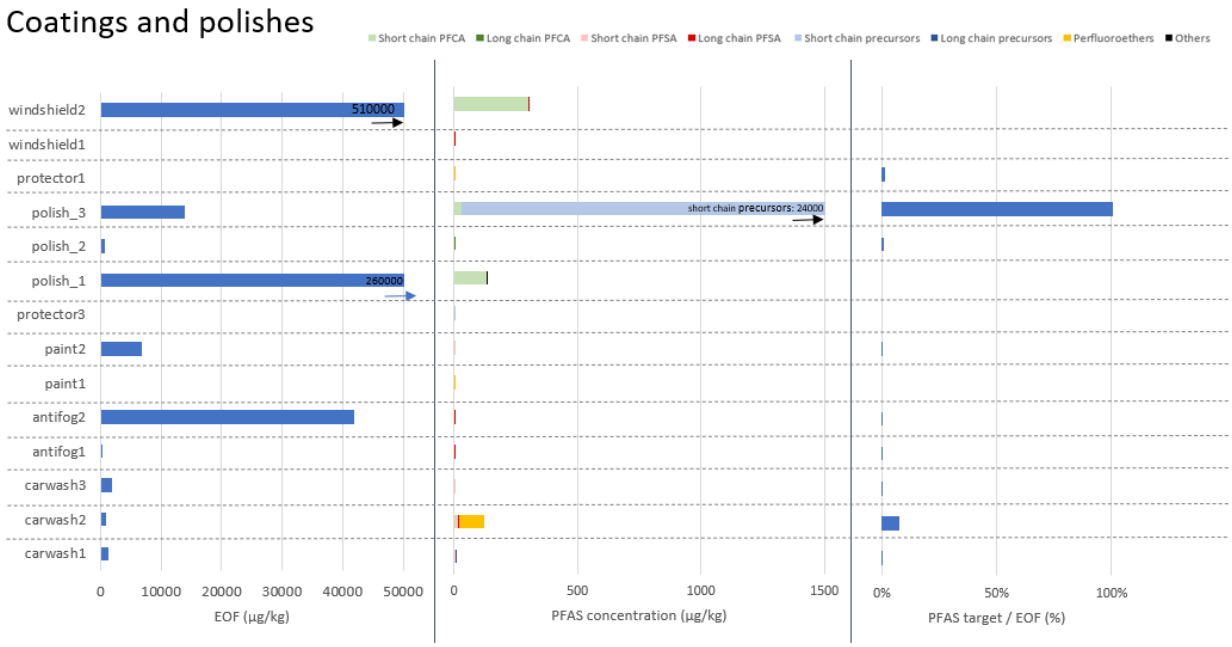

Figure 15 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for coatings and polishes

5.2.6 Other industries and miscellaneous

Fluoropolymers and elastomers

In the fluorpolymer and elastomer industry, several dust samples were collected at factories where PTFE type of products were processed into end-products. The presence of several PFAS-target compounds indicate that at industries where fluoropolymers are processed PFAS-target compounds are present (e.g as impurity in fluoropolymers), however in the end-products PTFE, ETFE and FEP relatively low concentrations of PFAS-target were detected.

In some other articles, the label indicates that PTFE is present in the product, e.g. one of the lubricants (lubricant1). In this sample PFAS-target compounds have been detected (several PFCAs), but no additional PFAS have been detected using the TOP-analysis. It is possible that these PFCAs originate from the PTFE used in the product.

Extremely high EOF has been detected in the three fluorinated rubbers tested (Viton/FKM rubber rings). It is expected that parts of the rubber dissolve into the methanol used for extraction, hence also the fluoroelastomer itself is being detected in the EOF analysis. In the extract of the rubber rings PFAS-target compounds have been detected up to 300 µg/kg, the PFAS detected are mainly PFCA's, with a high level of PFOA in one of them (190 µg/kg).

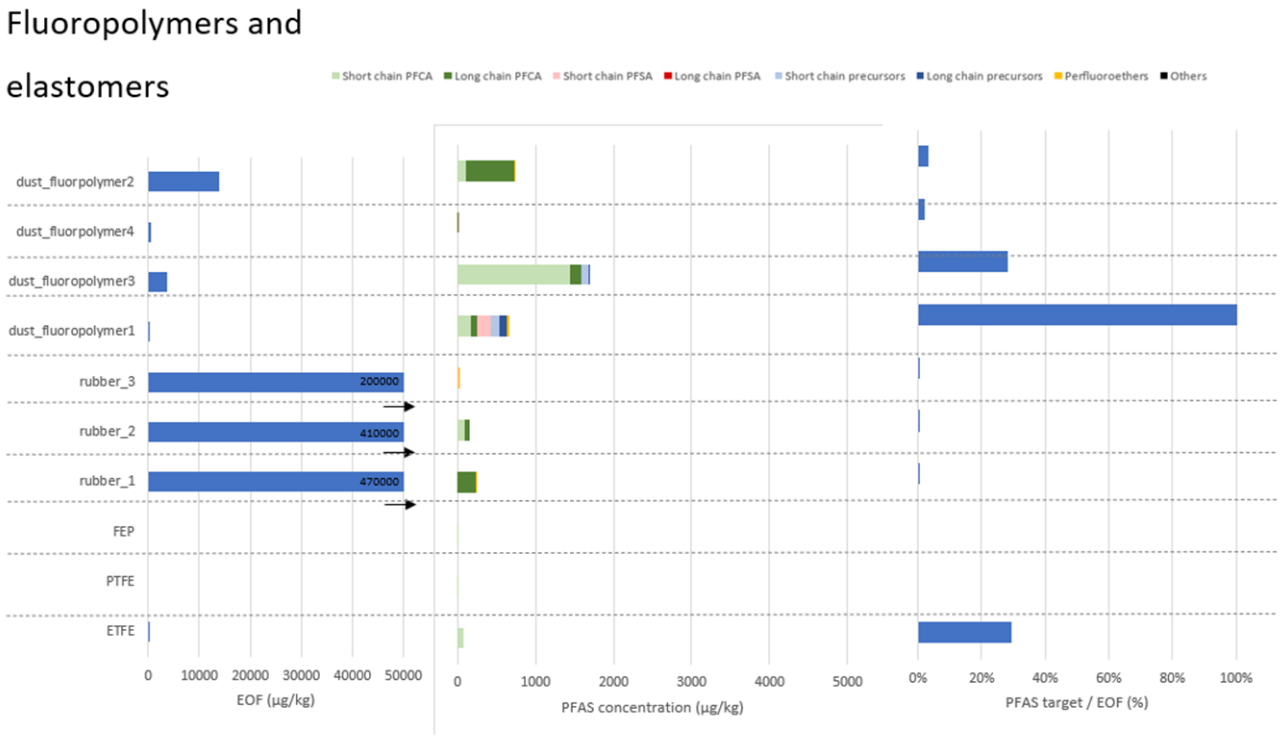

Figure 16 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for fluoropolymer industry and elastomers

Rubber, plastic and electronic industries

These industries were screened via dust sampling. Although the presence of PFAS in dust is only an indication for the use of PFAS, elevated concentrations were measured, mainly in the plastic industry and electronics.

Miscellaneous

In the category “miscellaneous” other types of samples have been analysed. This concerns several types of lubricants, car lacquer, windshield treatment, adhesives and sealants, rubber tires, pesticides, cosmetics, artificial grass and firework.

In most of the samples some PFAS-was detected. Low concentrations were detected in rubber tires and artificial grass (<10 µg/kg).

High concentrations were detected in car paint, windshield treatment and in protector used for joints between tiles. The presence of PFAS in the windshield treatment and joint protector was confirmed by TOP-analysis.

The PFAS-target concentrations detected in pesticides are very low. One of the samples shows high EOF, which is caused by the pesticide itself containing a fluorine (not necessarily being PFAS). The paper of the fireworks contains PFAS, mainly PFPrA (similar to the paper pulp samples).

Another interesting category is make-up. The detected concentrations in both mascara samples are moderate (20-40 µg/kg), but make-up has direct contact to the skin and could therefore be very relevant. In the study of Colles et al., 2020, the use of cosmetics was identified as a possible exposure route for human, based on the levels of PFAS in blood.

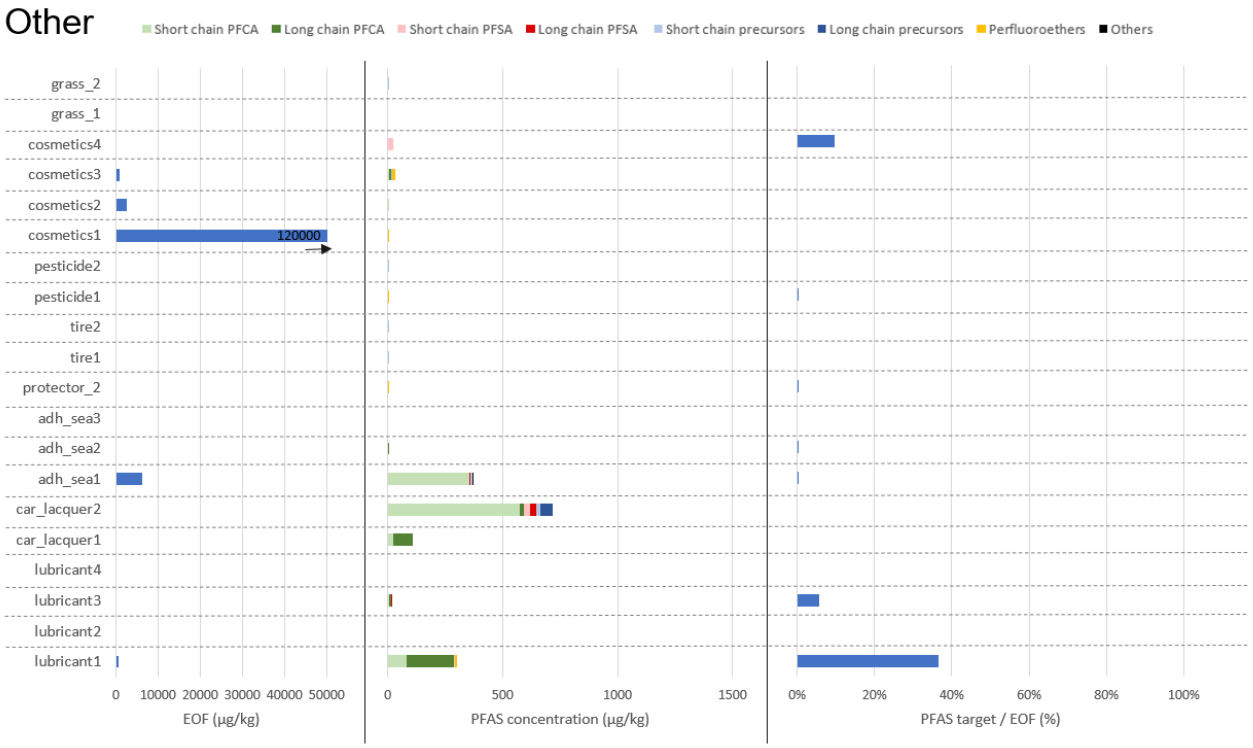

Figure 17 Results EOF, PFAS target analysis and percentage EOF found with PFAS target for other industries and miscellaneous

5.2.7 End of life industries

In sewage sludge, PFAS are being detected (PFAS-target; 100-200 µg/kg). In general, the EOF has the same order of magnitude, a large part of the EOF can be explained by the PFAS-target compounds. The TOP analysis for one of the sludge samples shows that PFAS-precursors are present.

5.3 Identifying the most relevant sources

The goal of this study is to gain insight in the presence of PFAS in products, production and recycling processes and waste, and to identify significant exposure routes and release pathways of PFAS in products and waste to humans and the environment. The study focusses on the identification of the most relevant sources of PFAS and does not aim to be fully comprehensive.

Detected concentrations in certain products do not mean that these products are important exposure or release pathways. Some of the products are very specific, and are not used on a regular basis (e.g. joint protector in bathrooms). This does not alter the fact that high concentrations in a short period of time also contribute to the general PFAS load in the Netherlands.

Based on the results gathered in this study an estimation of the load of PFAS in certain industries has been made. Although many samples have been analysed, it must be stressed that this evaluation only gives an indication, since it is based on a small amount of samples per industry.

5.3.1 Quantifying the load of PFAS (indicatively)

For determining the quantities (total load) of PFAS that are used or sold in the Netherlands, data from the CBS (Centraal Bureau voor de Statistiek, Netherlands Statistics) database on the total amount of tonnes products/year are combined with the measured concentrations of PFAS in the product categories specified.

CBS data on tonnes product

The CBS makes use of different databases. One of these is the ‘ProdCom’ database. This database shows the sales of several product categories in the Netherlands. This database was used for the outdoor and sail samples, the detergents, water repellent products, paper, leather and carpets. Sales numbers can give a good indication of the use of products. The datasets from the CBS are not in all cases complete. For some years and some product categories, the data were missing (noted as “0”). The missing data have not been taken into account in average calculations.

Another database is the ‘goederensoort naar land’ database. This shows the import and export of product categories for the Netherlands. This database has been consulted, but after analysis the import of all relevant product categories (in kg) is lower than the export, which results in a negative net use and is therefore not useful for the purpose of this research.

The data for the pesticide use is a total from all pesticide use in the Netherlands in kilograms. The kilograms are ‘kilograms of active ingredient’, calculated to tonnes.

The total of sewage sludge is based on the on the yearly production of sewage sludge by wastewater treatment plants in the Netherlands (van Voorthuizen et al., 2019). The total amount of sludge is 1,4 million tonnes. After consulting the authors of the article, this sludge has a percentage of 23% dry weight on average (personal communication of the author).

Categories and starting points for calculation

Using the CBS data and literature, the PFAS load of the following categories can be estimated: outdoor and sail samples, the detergents, water repellent products, paper, leather, carpets, pesticides and sewage sludge. The other categories have been estimated based on other literature sources. An overview of the quantities and calculations is given in Appendix C.

It must be noted that the samples detergent category are quite diverse. Also the content of the selected CBS categories are not completely clear. Considering the uncertainties that come from the data analysis and the diversity that exist between detergents and samples, the numbers found in the category should be interpreted carefully.

For the water repellent products (shoeshine and related products), the product categories contain a wider variety of products. However, because the water repellent properties are needed for these applications, the analysed products will still be considered relevant for this calculation.

For the paper samples, total numbers of all paper categories were calculated and used for the calculation. Because there are no numbers of paper pulp, the total amount of paper per year is also used for this category. Therefore the assumption is that the amount of paper that is sold in a year, will all be recycled. This will probably lead to overestimation of the results, but will give an indication of the amounts that may be considered.

For the pesticide, the whole product (consumer product) and not the active ingredient was analysed (the active ingredient is reported in the statistical data), which may lead to an underestimation of the PFAS load because the active product is usually diluted in the pesticide end-product and may therefore lead to an underestimation. The analysed products are consumer products because professional products were not available for this project because of strict regulations. The database only considers professional use. Therefore, the PFAS concentrations can differ from the given numbers. It is not clear whether this leads to over- or underestimation of the total load. However, it is known that pesticides often contain fluorine as an active group in the molecule. This might, or might not, be a PFAS (based on the definition of PFAS).

5.3.2 Calculation and results

The calculation was possible for a selection of (mainly) product categories. In the table below, the CBS data on the total amount of material/product in tonnes were used to calculate the amount of PFAS and EOF that were possibly used. Per category an average is taken from the analyses that were performed in this study. Because of the small number of samples per category, this can only be a rough estimation of the possible content per category. Also the CBS data categories are not very clearly specified. As a result of these two uncertainties only a rough indication of the total kilograms in the order of magnitude per category can be given. See Table 3.

Table 3 Indicative masses of PFAS and EOF in products in the Netherlands estimated. Masses are calculated as the multiplication of tonnes product with average content analysed. Numbers with a higher degree of uncertainty are given in grey and italic

Description	Samples used for calculation	Total tonnes of material (average per year)	Potential total kg PFAS-target (order of magnitude)	Potential total kg EOF (order of magnitude)	Comments
Awning (sunscreens, tents, sails)	outdoor1 sail1 sail2	7000	1-10	10-100	No clothing included
Carpet	carpet1 to 3	194000	1-10	10-100	
Leather (excluding clothes and artificial leather)	leather	4250	0.1-1	0.1-1	Based on 1 product
Water and stain repellents and polishes	water_repellent1 to 3 footwear_spray1 footwear_cream_2 footwear_leather_3	18750	0.1-1	100-1,000	Not all categories of CBS database and our products do align. Totals are considered as indicative for the other categories
Paper	paper1 paper2 paper3 paper4	925000	1-10	100-1,000	
Processed paper recycling	paper_input1 paper_input2 paper_input_alternative2 paper_process2 paper_output2 paper_process3 paper_output3 paper_input3	925000	100-1,000	100-1,000	Same source as 'paper' samples. This is assuming all paper will be recycled
Cleaning agents	All 'dishwash' and 'cleaning agent' samples	*58000 (assumed 10% is relevant)*	*0.1-1*	*100-1,000*	Assumption; 10% of soaps concerns dishwashing and cleaning agents. Not clear what causes the EOF (likely inorganic fluorine)
Fluoropolymers	ETFE PTFE FEP	5000	0.1-1	1-10	Based on 10% of total of fluorpolymers is relevant for NL

Description	Samples used for calculation	Total tonnes of material (average per year)	Potential total kg PFAS-target (order of magnitude)	Potential total kg EOF (order of magnitude)	Comments
Fluoroelastomers	fluor_rubber1 fluor_rubber2 fluor_rubber3	500	0.1-1	*100-1,000*	EOF of elastomers is indicative because it dissolves during extraction. Assumed 20% of fluoroelastomers is relevant for NL
Pesticides	pesticide1 and 2	6000	0.01-0.1	*10-100*	Based on active ingredients – possible underestimation
Fireworks (paper)	firework1 firework2 firework3	16000	0.1-1	0.1-1	Annual use; ~16 mln kg (Deltares/TNO, 2018) Assuming 10% of sold fireworks is paper
Sewage sludge WWTPs	sewage _sludge 1 to 4	320000	10-100	10-100	

Note: no usable data were found on coatings, polishes or special lubricants. These may also be important sources.

Some numbers in the table are given in italic and grey. These numbers show a high uncertainty, which is explained below.

Based on the estimated amounts of EOF and the contribution of PFAS in EOF, it can be concluded that 4 categories represent a possible high load of EOF (>100 kg/year) and thus likely PFAS. These are water- and stain repellents, paper and possibly cleaning agents and fluoroelastomers.

In the recycled paper processes this high load of EOF can directly be appointed to target compounds PFAS. In the repellents and cleaning agents, there is a large difference between the target analysis and EOF analysis, which can be caused by PFAS-precursors. The presence of PFAS-precursors is confirmed by the TOP-analysis for the repellents, but not for the cleaning agents. Therefore, it is not clear whether the EOF measured in cleaning agents is caused by PFAS or by other fluorine-containing compounds. The same accounts for the fluoroelastomers, likely a part of the fluoroelastomers has dissolved during extraction, and the fluoroelastomer molecules themselves are being detected by the analytical technology. Nevertheless, the fluoroelastomers show significant concentrations of PFAS-target compounds in the analyses.

The textile categories (awning and carpets) show moderate loads (10-100 kg/year) for awning and carpets, and low loads for leather (<10 kg/year). The water repellents are closely related to the textile categories and are often used for these type of products. The water- and stain repellents are the products that end up in the environment the most, because of weathering from the products they are used on, and in case of the use of sprays.

For the paper industry, PFAS do circulate in this industry in the recycling process and in the wastewater (and sludge) coming from this industry. The PFAS could originate from recycled food-contact materials and other oil- and water repellent applications, e.g. papers that are imported since oil and greaseproof paper is not produced in the Netherlands (Hekster et al. 2002).

Wastewater sludge shows a moderate load (10-100 kg/year). Wastewater and wastewater sludge are the endpoint of different sources, the load found in wastewater sludge can be considered as an indication of the PFAS use in the Netherlands. The load is in line with those of the other product categories, showing that no highly contributing source has been missed in the process.

The amounts of organofluorine load in pesticides can be overestimated (estimated at 10-100 kg/year), since the numbers are based on active ingredient instead of finished product. However, in most highly selective pesticides and herbicides the fluorinated group of the pesticides is quite short, and it depends on the definition of PFAS whether these compounds are highlighted as PFAS. Many of the highly selective pesticides and herbicides that are used currently are based on fluorinated building blocks (website Unimatec, 2019). Because pesticides are applied on crops they end up directly in the environment and, via the food chain, in human bodies.

Remarkably, fluoropolymers like PTFE, ETFE are not identified as a big source of PFAS (<10 kg/year). The analyses were aimed at the extractable amount of PFAS, not at the (inert) fluoropolymers themselves.

Cleaning agents are the products that also end up in the environment and in our wastewater easily, and moreover humans are most exposed to this product group via various ways, directly and indirectly. Although it is not certain that PFAS (precursors) are present in these types of products, some PFAS-target compounds have been found in this investigation and the high EOF concentrations indicate that there might be other PFAS present. Also the study of Jans and Berbee (2020) indicated the presence of PFAS at a production site of cleaning agents. At this moment, it is not clear to which extent the EOF is caused by other compounds. Since this is a relevant category for the environment and the amount of samples in this investigation was limited, this should be investigated into more detail.

Summarizing the loads of PFAS in the different product categories, combining the calculated quantities with the estimation whether the EOF is caused by PFAS or by other compounds (e.g. inorganic fluorine, organofluorine not being PFAS, dissolution of the samples), the relevance of the categories for release into the environment are estimated to be:

Highly relevant (>100 kg/year):
- Water and stain repellent products, including treated textile, carpets and leather.
- Paper recycling.

Moderately relevant (10-100 kg/year):
- Cleaning agents.
- Fluoroelastomer products.
- Pesticides.

Low relevance (<10 kg/year):
- Fluoropolymer products (PTFE etc).
- Fireworks.

Sewage sludge is also estimated to contain a relevant amount of PFAS, but is an endpoint for release into the environment.

5.3.3 Expected relevance for release into the environment

In Table 1 (Chapter 3) the investigated industries have been summarized. For most industries, samples have been taken and analysed, and based on the results, in the previous paragraph, an estimation has been made of the amount of PFAS that is being released into the environment. In Table 4, a summary of the estimated load and the expected relevance for release into the environment has been made for all categories and industries summarized in the beginning of this investigation (including the industries not sampled, and the industries for which it was not possible to estimate the load based on use data).

Not all industries and categories have been investigated in this study. It was not possible to include all industries because of the limited amount of samples and difficulty to obtain samples. Also a few known uses, like the use of AFFF has not been tested, since it is known that the current C6-based fluorine containing foams contain significant amounts of PFAS precursors.

For the industries which are lacking data on quantities used, the expected relevance has been estimated based on expert judgement.

Based on the information in the table, in addition to the list in the previous paragraph, AFFF has been added to the list with high relevance. Several industries like coatings and polishes, galvanic industry, automotive, semiconductors and healthcare and hospitals are also estimated to have moderate relevance and might be a source of PFAS to the environment, however, this is not based on actual numbers within this investigation.

Table 4 Expected relevance for release into the environment of the different industries.

Description industry	Total estimated load (kg/year)	Expected relevance for release into the environment	Comments
Textile, leather and carpet industry (including water and stain repellents)	>100	**High**	Mainly water and stain repellent applications. This can also include clothing. PFAS end up in dust and recycled materials.
Paper (including recycling)	>100	**High**	Highest PFAS load in recycled paper
Food related	See fluoropolymers	**Low (relevant for human)**	Low relevance for the environment but possible exposure for human
Cleaning agents	10-100	**Moderate**	Possible load present, cause elevated EOF not clear
Coatings and polishes	Unknown	**Moderate**	High concentrations in stain and water repellent applications. Total load unknown, some included in category water and stain repellents). Relevance estimated to be moderate
AFFF	Estimated to be >100 kg/year	**High**	Not tested in this investigation. Relevance for release into the environment is high due to direct application into the environment and known use of PFAS-precursors
Other industries			
- Fluorpolymer/-chemical industry (PTFE etc)	<10 kg	**Low**	Based on this investigation
- Aerospace/aircrafts	Unknown	**Low-moderate**	Not investigated in this research. Likely use in hydraulic fluids
- Galvanic industry	Unknown	**Moderate**	Not investigated in this research. Known use
- Automotive	Unknown	**Moderate**	PFAS present in several applications, sprays, textiles, windshield and car wash/wax/paint
- Rubber including fluoroelastomers	10-100	**Moderate**	Fluoroelastomers might indicate a moderate significant source to the environment. In other rubber samples (tyres) no significant concentrations detected. Dust indicates some PFAS use.
- Plastic	Unknown	**Low**	Dust indicates some PFAS use. Amount unknown. Expected to be low based on likely use of polymers (solids)
- Electronics	Unknown	**Low-moderate**	Dust indicates some PFAS use. Amount unknown. Likely use of polymers (solids, less leaching to the environment), and indication of use of non-polymers in electronics industry (possible source to the environment)

Description industry	Total estimated load (kg/year)	Expected relevance for release into the environment	Comments
- Energy	Unknown	**Unknown**	Not tested
- Oil and gas	Unknown	**Unknown**	Not tested
- Semiconductors	Unknown	**Moderate**	Not tested, but known use
- Healthcare and hospitals	Unknown	**Moderate**	Not tested, likely used for stain and water repellence
Miscellaneous			
- Pesticides/herbicides	10-100	**Moderate**	Pesticides/herbicides often contain organofluorine. Whether this is PFAS depends on the definition of PFAS
- Cosmetics	Unknown	**Low (relevant for human exposure)**	The measured concentrations in cosmetics are relatively low, but might be relevant for human exposure
- Artificial grass	Unknown	**Low**	Low concentrations detected
- Fireworks	<10	**Low**	Significant amounts detected but the total load is relatively low
End of life industries	10-100	**Moderate**	Sewage sludge is a sink for PFAS

5.4 Other Observations

Cheap versus expensive materials
As indicated in paragraph 2.2, PFAS are considered to be expensive materials (Kissa, 2001). However, low amounts of PFAS can be sufficient for a certain application, and the lower concentrations might compensate the higher price, as well as the longer lifespan for the materials.

In this research, also a couple of very cheap materials, from discount stores has been tested (amongst others a water repellent for textiles, barbecue cleaner, cleaning agent and anti-fog for cars). All these samples show elevated levels of PFAS-target or EOF, despite the fact PFAS is considered an expensive material. The elevated level of EOF in the water repellent for textiles has been confirmed by the TOP analysis.

All the other materials obtained during this study are generally obtainable. Expensive materials like perfluoroether greases have not been analysed, but are certainly being used in certain industries.

Ingredient list
Whenever available, the ingredient list of the product has been studied. Fluorinated compounds were not mentioned on any of the products, except for one of the pesticides. It is assumed that for most products, PFAS-compounds are included into the category <5% anionic surfactants, which was mentioned at almost all liquid products.

Certainly the category <5% anionic surfactants does not mean that in all cases this would be PFAS. However, a combination of the category <5% anionic surfactants and an oil, grease and water repellence gives a high probability for PFAS.

Concentrations of PFOS + PFOA + PFHxS + PFNA

Recently, the European Food and Safety Authority (EFSA) has published new tolerable weekly intake numbers (TWI) for the sum of 4 PFAS, being PFOS, PFOA, PFHxS and PFNA. The TWI is set at a (very low) value of 4.4 ng/kg bw/day. For the analyses in this project, the PFOS+PFOA+PFHxS+PFNA concentrations have been calculated in the products (see Appendix A). These values are generally high (> 25 µg/kg) in several (12) dust samples, and also in the recycled carpet fluff, outdoor textiles (fabric sun screen and sail), in a baking mat (direct contact with food) and fluorinated rubber.

6 CONCLUSIONS AND RECOMMENDATIONS

The goal of this study is to gain insight in the presence of PFAS in products, production and recycling processes and waste, and to identify significant exposure routes and release pathways of PFAS in products and waste to humans and the environment. For obvious reasons this study had to be limited to a manageable portion of these categories, and does therefore not aim to be comprehensive, but as representable as reasonably possible.

The selected samples focus at (consumer) products. Known industrial PFAS-production processes and locations with PFAS-sources have not been investigated (e.g. PTFE-production, fire-training areas).

6.1 Conclusions

Relevant categories for release of PFAS to the environment

The relevance of a product category is estimated based upon the PFAS load that might end up in the environment. The contribution and relevance of a product or waste category for the release of PFAS in the environment does not only depend on the concentration present in the samples, but also on the amount of product used and the type of use. The estimated loads are very rough but give a good impression of the relative contribution to the environment. The most relevant categories in order of relevance are expected to be:

Highly relevant (>100 kg/year):
- Water and stain repellent products, including treated textile, carpets and leather.
- Paper recycling.

Moderately relevant (10-100 kg/year):
- Sewage sludge.
- Cleaning agents.
- Fluoroelastomer products.
- Pesticides.

Low relevance (<10 kg/year):
- Fluoropolymer products.
- Firework.

Coatings and polishes also show high loads of PFAS, but no estimate could be made of the relevance compared with the other categories as a result of failing or unsuitable data on amounts used in the Netherlands.

In some cases, products with a low relevance for the environment (looking at kgs/year) can cause a direct exposure for PFAS to human. E.g. in PTFE-baking mats, PFAS-target compounds have been detected, some even above the current limit for PFOA in products according to the POP-regulation (25 µg/kg). Also in make-up some PFAS has been detected. The amounts are relatively low when comparing to the other tested products, but can be a concern considering the direct use of the product on the skin or possible uptake via food.

Dust samples have been taken in several industries, some offices and households, and indicate the widespread use of PFAS. In dust, the concentrations are often relatively high, several hundreds to several thousand µg/kg. The origin of dust is usually not clear, and can also be explained by wear of clothing, carpets, but in several industries higher levels have been measured than in household and office dust. Together with sewage sludge this is considered to be caused by wear and waste of PFAS containing products.

All samples tested were selected based on the exposure routes and release pathways to human and the environment. Some industries have not been tested, e.g. chrome plating, in this industry it is known that PFAS are being used, and it concerns a closed process. Another type of important use of PFAS is AFFF (fluorine firefighting foams), which can also be directly released to the environment. Currently phasing out of AFFF (containing fluorinated compounds) is taking place or considered.

Conclusions concerning the types of PFAS present

For most product samples, the PFAS-target concentrations explain less than 10% of the EOF detected. This indicates the presence of PFAS-precursors, which has been confirmed by the TOP-analysis for several samples. Water and greaseproof treatments, for textiles, carpets, leather, tiles (floor polish), facades and glass often contain high amounts of PFAS-precursors. This can also be seen in e.g. the textile, carpet and leather industry, where the awning and leather contains significant amounts of PFAS.

In dust (and also in sewage sludge, process and recycling samples), a higher fraction of the EOF can be explained by PFAS-target compounds, which shows that dust acts as a sink for PFAS and likely, the PFAS-precursors have partly been transformed into detectable compounds. Summarizing; in products relatively more PFAS-precursors are expected to be present, which transform in the environment into (a larger fraction of) detectable PFAS-compounds, which are detected in dust and sludge.

In the paper industry, although low concentrations have been measured in the general paper samples tested, high concentrations of PFAS have been detected in the paper of fireworks and in the recycled paper pulp samples. Especially the concentration of PFPrA (C3 PFCA) is very high, which could be a degradation product of short chain PFAS-precursors used for paper. PFPrA has been detected in the paper of fireworks as well as the recycled paper pulp.

In the fluoropolymer and -elastomers industry, the fluorelastomers pop out. Significant PFAS-target concentrations have been measured in fluoroelastomers, about 10 times higher than in fluoropolymers.

The findings of this study are roughly in line with what can be expected based on the properties of PFAS. The categories that were found to contain the highest volumes of PFAS are water and stain repellent sprays or the materials that they are treated with (including some type of polishes and coatings). A as result of the widespread use, high amounts of PFAS are also found in sewage sludge and dust from industry, offices and even households. The amounts of PFAS in fluorpolymers like PTFE and Teflon, car tires, silicon baking forms and artificial grass are found to be relatively low.

6.2 Recommendations and discussions

Recommendations for additional investigations

Paper
The detection of PFPrA in several paper and paper pulp samples indicates the use of PFAS-precursors in these types of material. It would be recommended to further investigate the origin of these compounds, whether these types of paper are imported or that the precursors are added in the production process in the Netherlands. As far as information is currently available, PFAS are not intentionally added in the paper production process in the Netherlands. However, if these compounds originate from PFAS-precursors or fluorinated polymers, the registration of the compounds is often mentioned as confidential business information or not registered at all (e.g. polymers in REACH).

Cleaning agents
The high EOF detected in cleaning agents does not necessarily mean that PFAS are present, although some PFAS-target compounds have been detected. Since cleaning agents represent a high volume use, and elevated concentrations were measured at a cleaning agent production plant (Jans en Berbee, 2020), it would be recommended to investigate the high EOF in further detail.

Pesticides
For the pesticides, which are directly used in the environment, it would be recommended to check in further detail which pesticides might be defined as PFAS (this depends on the definition of PFAS).

Fluoroelastomers
Fluoroelastomers are polymers based on fluorinated compounds. Since relatively high concentrations of PFAS-target compounds have been measured, it would be recommended to check the fluoroelastomers production process on the use and emission of PFAS, and how to reduce the remaining PFAS in the product (e.g. content of PFOA).

Gaps in the exposure pathways and sampling programs

The analysis in this investigation focusses on solids and related streams such as dust and sludge. Wastewaters are being analysed in the study of Rijkswaterstaat. The distribution of PFAS does not only take place through water, products or dust, but also through air. The release of PFAS via the air is significant, as the elevated background concentrations in Dutch top soils and the effects of the emissions of DuPont/Chemours have proven. Sampling of the air at production locations is not foreseen in the three investigations that currently are being conducted (Rijkswaterstaat, NVWA, current investigation), but might be interesting at certain locations where PFAS are being used.

Constraints in the analyses

Technical limitations of the analyses limit the range of PFAS that can be detected but this does not mean that these undetected PFAS are not present or are harmless:

- EOF relies on an extraction of the PFAS from the materials. It is possible that not all PFAS are being extracted during this step.
- The EOF analysis is not specific for the group of fluorinated organic surfactants that have been the main focal point of environmental and health concerns. EOF is a measure for all extractable organofluorines including, for example, non-ionic fluorohydrocarbons. Because of the potentially very diverse group of compounds covered by the EOF analysis, levels cannot be translated into environmental or health impact without further knowledge of the composition of the samples.
- Volatile PFAS (e.g. FTOHs) could volatilize during the analytical procedure and therefore the concentrations of volatile PFAS can be underestimated.
- PFAS polymers like PTFE are not being extracted (intentionally, since the project focusses at the PFAS monomers).
- The PFAS analysis focuses on a certain set of PFAS. Not all PFAS are being detected with this analysis.
- The analysis focusses at C4-C18 PFAS compounds. Also, a C3-PFCA has been added to the list (PFPrA). Smaller PFAS compounds (C1-C2) are not included. The presence of PFPrA in several samples (mainly paper) shows the significance of these short chain compounds.
- Due to all the different steps in analysis, the uncertainty of the measurements can be higher than for regular analyses, and re-analysis can result in different values.

Limitations in extent of the sampling program

This investigation is limited to 129 samples. Although that is a relatively large number, there are multiple types of application of PFAS. The choice was made to select at least 2-4 samples per product or waste stream. This implicates that not all products, processes or streams could be sampled. Sampling selection was based on the literature research from phase 1. Within the 2-4 samples per category, the samples were chosen to be as representative as possible, for example by selecting products from different price categories or by selecting products which are expected to contain less or more PFAS than another product in the same category. Although this selection has been executed with great care, it is possible that some product categories will not be represented completely. However, the goal of the current project is to get an indication of which product categories contain PFAS and the relative importance of these product categories, and this goal can be achieved with the current setup.

Monitoring of other media

The presence of PFAS in human blood is a good indicator for the exposure of PFAS to humans, and in the end is the best indicator for overall exposure to PFAS and the related risks. Data on the presence of PFAS in blood in the Netherlands is scarce, and usually focused on only a few legacy PFAS. Since there currently is a significant changeout of PFAS, we strongly recommend expanding this to a larger range of PFAS and analytical approaches including total organic fluorine, to be able to monitor the effect of shifting PFAS usage on human exposure (within the HMB4EU study a larger set of PFAS is being monitored). The same accounts for PFAS in food, it is recommended to expand the analytical scope from just a few target-PFAS to a wider scope including total organic fluorine.

With increasing data this will facilitate linking the levels in blood, and thus the exposure of humans, to the use of PFAS in products. PFAS profiling of both products, pathways (water, dust etc.) and blood will enable an even better identification of the major PFAS uses and risks.

7 LITERATURE

- Barzen-Hanson, K. A., et al. (2017). "Discovery of 40 Classes of Per- and Polyfluoroalkyl Substances in Historical Aqueous Film-Forming Foams (AFFFs) and AFFF-Impacted Groundwater." Environ Sci Technol 51(4): 2047-2057.
- Bentum, E. van, Pancras, T., Slenders, H., Enden, B. van der (2017). “Luchtdepositie onderzoek PFOA en HFPO-DA (GenX) Dordrecht en omgeving. Onderzoek naar de invloed van luchtemissies op de kwaliteit van grond en grondwater. Expertisecentrum PFAS, mei 2017.
- Bentum, E. van, Pancras, T., Slenders, H., Linden, P. van der (2018a) “Verkennend onderzoek naar PFOA en GenX in het milieu in Helmond. Onderzoek naar het voorkomen van PFAS in grond, grondwater, waterbodem en oppervlaktewater, Expertisecentrum PFAS, 23 oktober 2018.
- Bentum, E. van, Pancras, T., Slenders, H., (2018b) Aanvullende luchtdepostieonderzoek PFOA en HPFO-DA (GenX) Dordrecht en omgeving. Aanvullend onderzoek naar de invloed van luchtemissies op de kwaliteit van grond en grondwater. Expertisecentrum PFAS, 26 maart 2018.
- Bentum, E. van, H. Slenders (2019). Onderzoek naar de aanwezigheid van PFOA in de gemeente Molenlanden, Fase 4. Onderzoek naar concentraties PFOA in de grond, grondwater en irrigatiewater in de gemeente Molenlanden. Expertisecentrum PFAS, 3 oktober 2019.
- Blom, C., Hanssen, L., (2015). “Nordic Working Papers: Analysis of per- and polyfluorinated substances in articles.” Nordic Council of Ministers, NA2015:911.
- Bokkers, B.G. H., B. van de Ven, P. Janssen, W. Bil, F. van Broekhuizen, M. Zeilmaker, A.G. Oomen (2018). Per- and polyfluoroalkyl substances (PFASs) in food contact materials. RIVM letter report 2018-0181.
- Bruinen de Bruin, Y., Zweers, P., Bakker, J., Beekman, M. (2009). “Estimation of emissions and exposures to PFOS used in industry. A PFOS use inventory in metal plating and fire fighting”. RIVM report 601780002/2009
- CBS (Centraal Bureau voor de Statistiek) databases, https://opendata.cbs.nl/, consulted in January 2021.
- Chen, F., Eriksson, U., Aro, R., Yeung, L., Wang, T., Kallenborn, R. & Kärrman, A. (2016) “Screening of per- and polyfluoroalkyl substances (PFASs) and total organic fluorine in wastewater effluent from Nordic countries”
- Deltares/TNO (2018). Emissieschattingen diffuse bronnen, emissieregistratie. Afsteken vuurwerk, June 2018.
- European Commision DG Environment and European Chemicals Agency (2020) “The use of PFAS and fluorine-free alternatives in fire-fighting foam”, document reference 41288-WOD-XX-XX-RP-OP-0009_a_PO3, Final report (issue 3)
- European Commision DG Environment and European Chemicals Agency (2020) “The use of PFAS and fluorine-free alternatives in textiles, upholstery, carpets, leather and apparel”, document reference 42054-WOOD-XX-XX-RP-OP-0004_S4_P01.3, Final report V3, 22/10/2020.
- EU (2017). Commission Regulation 2017/1000 of 13 June 2017 amending Annex XVII to Regulation (EC) No 1907/20016 of the European Parliament and of the Council concerning REACH as regards PFOA, its salts and PFOA-related substances. Official Journal of the European Union L150/14.
- EU (2019). Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants (recast). Official Journal of the European Union L169/45.
- Eriksson, U. & Kärrman, A. (2015). “World-wide indoor exposure to polyfluoralkyl phosphate esters (PAPs) and other PFASs in household dust”
- Gardiner, J. (2015). “Fluoropolymers: Origin, Production and Industrial and Commercial Applications. Aust. J. Chem. 68, 13-22.
- Glüge, J, M. Scheringer, I.T. Cousins, J.C. DeWitt, G. Goldenman, D. Herzke, R. Lohmann, C.A. Ng, X. Trier, Z. Wang (2020). An overview of the uses of per- and polyfluoroalkyl substances (PFAS). Environmental Science: Processes & Impacts, 12: 2345-2373.
- Glynn, A., Berger, U., Bignert., A., Ullah, S., Aune, M., Lignell, S. & Darnerud, P. O. (2012), “Perfluorinated Alkyl Acids in Blood Serum from Primiparous Women in Sweden: Serial Sampling during Pregnancy and Nursing, And Temporal Trends 1996−2010”
- Glynn, A., J. Benskin, S. lignell, I. Gyllenhammar, M. Aune, T. Cantillana, P.O. Darnerud, O. Sandblom (2015). “Temporal trends of perfluoroalkyl substances in pooled serum samples from first-time mothers in Uppsala 1997-2014. Report to the Swedish EPA, contract no. 215124, 7 September 2015.
- Groot, de, G.M., Bakker, J., Luit, R.J. (2013). “Ketenanalyse impregneermiddelen”. RIVM briefrapport 300003002/2013.

- Hage, K., T. Pancras, R. Vis (2018). “Aanwezigheid van PFAS in Nederland. Deelrapport A - PFAS in grond en oppervlaktewater”. Expertisecentrum PFAS, d.d. 1 June 2018.
- Harrad, S., Wemken, N., Drage, D. S., Abdallah, M. A. E. & Coggins, A. M. (2019), “Perfluoroalkyl substances in drinking water, indoor air and dust from Ireland: Implications for Human exposure”
- Haug, L. S., Huber, S., Schlabach, M., Becher, G. & Thomsen, C. (2011), “investigation on per- and polyfluorinated compounds in paired samples of house dust and indoor air from Norwegian homes”
- Hekster, F.M., Voogt, P. de, Pijnenburg, A.M.C.M., Laane R.W.P.M. (2002). “Perfluoroalkylated substances – Aquatic environmental assessment.” Report RIKZ/2002.043, 1 July 2002.
- Hertzke, D., Olsson, E., Posner, S. (2012). “Perfluoroalkyl and polyfluoroalkyl substances (PFAS) in consumer products in Norway – A pilot study.” Chemosphere 88: 980-987.
- Holmquist, H., et al. (2016). "Properties, performance and associated hazards of state-of-the-art durable water repellent (DWR) chemistry for textile finishing." Environ Int 91: 251-264.
- ICIS2000 'fluoroelastomers become more complex' d.d. 13 November 2000
- IL&T (2019). Vervolgonderzoeken afvalstromen Chemours. Inspectie Leefomgeving en Transport. d.d. 26 augustus 2019.
- ITRC (2017). History and use of per- and polyfluoroalkyl substances (PFAS).
- Jans, A.C.H., R.P.M. Berbee (2020). Bronnen van PFAS voor het Nederlandse oppervlaktewater. RWS Lelystad, d.d. 14 July 2020.
- KEMI, S. C. A. (2015). Report-7-15-Occurrence-and-use-of-highly-fluorinated-substances-and-alternatives. https://www.kemi.se/global/rapporter/2015/report-7-15-occurrence-and-use-of-highly-fluorinated-substances-and-alternatives.pdf.
- Kissa, E. (2001). Fluorinated Surfactants and Repellent. NY, Marcel Dekker, Inc.
- Kort, M.J. de, C.J. de Jong, J.E.E. Ng-A-Tham, J.K. Verhoeven, P.E. Boon, A.J. Verschoor (2019). Verspreiding van GenX-stoffen in het milieu. Metingen in Nederland 2013-2018. RIVM Rapport 2019-0083.
- Kotthoff, M., Müller, J., Jürling, H., Schlummer, M., Fiedler, D. (2015). "Perfluoroalkyl and polyfluoroalkyl substances in consumer products." Environ Sci Pollut Res(22): 14546-14559.
- Lambert, Y.J., D.A.M. Plume (2008). United States Patent Application. No: US2008/0090955 A1. Polyethylene composition for artificial turf.
- Nilsson, H., et al. (2010). "A time trend study of significantly elevated perfluorocarboxylate levels in humans after using fluorinated ski wax." Environ Sci Technol 44(6): 2150-2155.
- OECD (2018). "Comprehensive global database of per-and polyfluoroalkyl substances (PFASs)."
- Pancras, T, E. van Bentum, H. Slenders (2018a). “Aanwezigheid van PFAS in Nederland. Deelrapport B - Onderzoek van PFAS op potentiële risicolocaties”. Expertisecentrum PFAS, d.d. 1 June 2018.
- Pancras, T, E. van Bentum (2018b). “Aanwezigheid van PFAS in Nederland. Deelrapport C – Diffuse belasting van PFOS en PFOA in de bovengrond”. Expertisecentrum PFAS, d.d. 1 June 2018.
- PlasticsEurope (2017). “Socio-economic analysis of the European fluoropolymer Industry – executive summary.
- Roelandse, A., Timmer, H. (2017). “Het effect van de industriële lozing van Chemours op de aanwezigheid van FRD-903 in oevergrondwater. Oasen, 1 May 2017.
- UNEP-POPs (2017). “Guidance on best available techniques and best environmental practices for the use of PFOS and related chemicals listed under the Stockholm Convention on Persistent Organic Pollutants. Updated January 2017.
- UNEP-POPs (2011). “Report of the Persistent Organic Pollutants Review Committee of the work of its sixth meeting. Addendum. Guidance on alternatives to perfluorooctane sulfonic acid and its derivatives. UNEP/POPS/POPRC.6/13/Add.3/Rev.1, 8 November 2011.
- Vestergren, R, G. Thorsén, M.A. Nguyen (2019). Identifiering av hittills okända PFAS i konsumentprodukter med HRMS. Kemi PFAS network meeting on 9 May 2019. https://www.kemi.se/download/18.7729c7771744357921040e8/1599136351617/pfas-natverksmote-9-maj-2019.pdf
- Voorthuizen, E. van, R. van Esch, B. Reitsma, T. Leeuwerke (2019). Tot over de oren in de slibcrisis. Verwerking zuiveringsslib vraagt om duurzame oplossingen. H2O, nummer 10, oktober 2019.
- Wang, Z., J.M. Boucher, M. Scheringer, I.T. Cousins, K. Hungerbuhler (2017). Toward a Comprehensive Global Emission Inventory of C4-C10 Perfluoroalkanesulfonic Acids (PFSAs) and Related Precursors: Focus on the Life Cycle of C8-Based Products and Ongoing Industrial Transition. Environmental Science and Technology, 51:8, 4482-4493
- Wang, Z., I.T. Cousins, M. Scheringer, R.C. Buck, K. Hungerbuhler (2014). Global emission inventories for C4-C14 perfluoroalkyl carboxylic acid (PFCA) homologues from 1951 to 2030, Part I: production and emissions from quantifiable sources. Environment Int. 70:62-75.

- Weiss, J., Franke, V. & Gustafsson, Å. (2019), “Screening of Polyfluoralkyl substances (PFASs), including fluorinated organophosphates in Skellefteå municipality.”
- Wintersen, A., J. Spijker, P van Breemen, H van Wijnen, RIVM “Achtergrondwaarden perfluoralkylstoffen (PFAS) in de Nederlandse landbodem”, RIVM-briefrapport 2020-0100.
- Wu, X. M., Bennett, D. H., Calafat, A. M., Kato, K., Strynar, M., Andersen, E., Moran, R. E., Tancredi, D. J., Tulve, N. S. & Hertz-Picciotto, I. (2015), “Serum concentrations of perfluorinated compounds (PFC) among selected populations of children and adults in California”
- Yeung, L. & Mabury, S. A. (2016), “Are humans exposed to increasing amounts of unidentified organofluorine?” Environmental Chemistry 13(1).
- Zeilmaker, M. et al. (2016). “Risicoschatting emissie PFOA voor omwonenden. Locatie: DuPont/Chemours, Dordrecht, Nederland. RIVM Briefrapport 2016-0049.

Websites accessed 2019/2020

- 3M, 2019.
  - http://multimedia.3m.com/mws/media/624307O/3m-fluorosurfactants-for-paints-and-coating.pdf
  - http://multimedia.3m.com/mws/media/774720O/3m-flame-retardant-additive-fr-2025.pdf
  - http://multimedia.3m.com/mws/media/750888O/3m-stain-resistant-additives-and-sealers-brochure.pdf
  - https://www.3m.com/3M/en_US/company-us/all-3m-products/~/All-3M-Products/Chemicals/Fluorochemicals
- Unimatec, 2019. http://www.unimatec-europe.com/index.php?id=67
- Daikin, 2019. https://www.daikinchemicals.com/solutions/products/mold-release-agents.html
- ChemicalWatch, 2019. https://chemicalwatch.com/85665/international-ski-federation-to-ban-pfass-in-ski-waxes#overlay-strip
- Ecocenter, 2019. https://www.ecocenter.org/toxic-forever-chemicals-infest-artificial-turf
- Canonvannederland, 2020. https://www.canonvannederland.nl/nl/overijssel/salland/zwartewaterland/de-tapijtindustrie-

APPENDIX A RESULTS EOF AND PFAS-TARGET ANALYSES

Appendix A. Results EOF and PFAS-target analyses

Type	Sample name		PFPrA	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUdA	PFDoA	PFTrDA	PFTeDA	PFHxDA	PFODA	PFBS	PFPeS	PFHxS	PFHpS	PFOS	PFDS	FOSA	FOSAA	N-MeFOSA	N-EtFOSA	N-MeFOSAA	N-EtFOSAA	4:2 FTS	6:2 FTS	8:2 FTS	10:2 FTS	8:2 diPAP	6:6 PFPI	6:8 PFPI	8:8 PFPI	6:2 PAP + 6:2 DiPAP	FHUEA	FOUEA	FDUEA	GENX	NADONA	9CI3ONS	11ClPF3OUdS	sum PFAS target	Extractable organic fluorine	PFOA	PFOS+PFOA+PFNA+PFHxS	PFAS target/Org. F (%)	TOP-analysis	Remarks
			µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	µg/kg	%			
Product	carpet1	most water and dirt resistant	2,7	1,3	0,51	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1,9	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	6,2	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	13	<100	<0,5	0	-	
Product	carpet2	polyester carpet	3,3	1,4	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	2	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	8	<100	<0,5	0	-	
Product	carpet3	long pile carpet	1,8	1,2	<0,5	0,92	0,64	1,1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1,4	<10	0,93	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	3,1	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	11	<100	1,1	1	-	
Dust	dust_carpet1	dust from carpet factory	<5	13	23	150	110	76	<2,5	<2,5	<2,5	<2,5	<2,5	<2,5	<10	48	22	8,3	<2,5	33	<2,5	<2,5	<5	<2,5	3,6	<2,5	<2,5	<2,5	<2,5	180	6,2	3,7	2,6	<5	<5	<5	<1000	<2,5	<2,5	<2,5	130	<2,5	<2,5	<2,5	809	360	76	117	154%		
Dust	dust_carpet2	dust from carpet factory	<5	7,4	2,7	3,3	<2,5	46	<2,5	<2,5	<2,5	<2,5	<2,5	<2,5	8,8	<10	<2,5	<2,5	<2,5	<2,5	6,3	<2,5	<2,5	<5	<2,5	<2,5	<2,5	<2,5	<2,5	33	<2,5	<2,5	<2,5	<5	<5	<5	<1000	<2,5	<2,5	<2,5	52	<2,5	<2,5	<2,5	160	240	46	52	45%		
Dust	dust_carpet3	dust from carpet factory	<5	79	<2,5	4,7	3	52	<2,5	<2,5	<2,5	<2,5	<2,5	<2,5	6,2	<10	<2,5	<2,5	<2,5	<2,5	4,1	<2,5	<5	<2,5	<2,5	<2,5	<2,5	<2,5	<2,5	19	<2,5	<2,5	<2,5	<5	<5	<5	<250	<2,5	<2,5	<2,5	160	<2,5	<2,5	<2,5	328	2900	52	52	8%	X	
Recycling	carpetfluff1	carpet fluff	3,8	15	1,7	16	9,4	40	1,7	1,7	0,59	1,2	<0,5	<0,5	<0,5	<10	10	0,71	2,4	3,1	66	<0,5	2,5	3,6	1,2	<0,5	28	3,5	<0,5	7,2	26	22	2,1	<1	<1	<1	<250	1,3	1,1	<0,5	<0,5	<0,5	<0,5	<0,5	272	2600	40	110	7%	X	
Recycling	carpetfluff2	drainage tube	1,6	1,3	3,3	<0,5	<0,5	0,69	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	3,3	<0,5	2,1	<0,5	<0,5	<1	<0,5	<0,5	0,73	<0,5	<0,5	2,4	<0,5	<0,5	0,56	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	16	<500	0,69	6	-		
Recycling	carpetfluff3	lining ponds	3,9	0,93	<0,5	<0,5	<0,5	1,2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	1	<0,5	<0,5	<0,5	<0,5	2,1	<0,5	<1	1,9	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0,9	<0,5	<0,5	1,8	<1	<1	<1	<250	<0,5	<0,5	<0,5	31	<0,5	<0,5	<0,5	44	<500	2,2	4	-		
Recycling	carpetfluff4	floor coverage horse stables	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	7,4	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	1,4	<0,5	<0,5	<0,5	<1	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	9	200	<0,5	0	3%		
Waste	sewage sludge1	Sewage sludge from WWTP close to carpet factori	1,2	1,2	<0,5	2,6	0,56	4,3	<0,5	0,84	<0,5	<0,5	<0,5	1,2	<10	5,2	<0,5	0,51	<0,5	5,9	<0,5	<0,5	3,3	<0,5	<0,5	<0,5	<0,5	0,64	7,1	1,7	<0,5	0,75	4,2	5,1	1,9	<250	<0,5	<0,5	<0,5	23	<0,5	<0,5	<0,5	71	140	4,3	11	35%	X	
																																																0			
Product	outdoor_1	fabric sun screen	4,7	8,4	2,3	20	5,3	74	5,1	31	2	11	0,72	2,3	0,89	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	2,3	1,9	1,9	<0,5	<1	<1	<1	<250	<0,5	2,1	1,5	<0,5	<0,5	<0,5	<0,5	177	8000	74	79	2%	X	
Product	outdoor_2	fabric outdoor furniture	<1	3,2	0,53	2,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	11	2500	<0,5	0	0%		
Product	outdoor_3	umbrella	<1	2,7	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	0,68	<0,5	<0,5	6,7	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	10	<500	<0,5	1	-		
Dust	dust_textile1	dust from sailmaker	2,5	3,6	5,3	20	12	84	18	18	10	14	22	26	23	<10	1,2	<0,5	1	<0,5	38	<0,5	1,3	6,8	1,2	0,58	2,4	8	<0,5	6,4	5,5	65	<0,5	<1	<1	<1	<250	1,5	1	<0,5	5,3	<0,5	<0,5	<0,5	<0,5	404	1800	84	141	15%	X	
Dust	dust_textile2	dust from sailmaker	2,7	4,5	4,8	31	15	180	75	18	56	110	72	93	110	<10	0,91	<0,5	3,7	0,65	150	<0,5	1	3,6	0,59	3,1	<0,5	2,6	<0,5	5,4	5,2	3,6	<0,5	<1	<1	<1	<250	1,3	14	1,7	24	<0,5	<0,5	<0,5	<0,5	993	26000	180	409	3%	X	
Product	sail1	sail sample	3,8	2,1	1,2	8,4	3	11	2,3	3,4	1,7	2,4	2,6	3,3	3,8	<10	0,77	<0,5	0,64	<0,5	1,4	<0,5	<0,5	1,3	<0,5	<0,5	<0,5	<0,5	<0,5	1,6	<0,5	0,98	<0,5	<1	<1	<1	<250	1,1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	57	7200	11	15	1%		
Product	sail2	sail sample	2,1	2,1	1,3	18	11	410	7	110	2,6	70	3,9	70	41	<10	<0,5	0,84	0,63	<0,5	13	<0,5	7,2	1,6	<0,5	<0,5	<0,5	<0,5	<0,5	3,2	0,69	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	776	880	410	431	60%		
Product	water repellant1	cheap water repellant	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	20000	<0,5	0	0%	X	
Product	water_repellant2	textile spray general	1,1	1,9	1,5	3,8	1,6	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	10	11000	<0,5	0	0%		
Product	water_repellant3	general water repellant	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	33000	<0,5	0	0%		
																																														0			
Product	leather1	leather sample from factory	1	0,5	<0,5	<0,5	3	0,85	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	0,76	1,3	0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	1,6	<0,5	<0,5	<0,5	0,57	2,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	14	<0,5	<0,5	<0,5	27	<100	0,85	1			
Dust	dust_leather	dust from furniture factory	21	2,7	<0,5	6,4	1,2	19	1,3	1,4	<0,5	1,2	0,63	0,6	0,96	<10	26	<0,5	1	<0,5	32	0,69	<0,5	450	0,53	4,3	1,3	0,6	110	0,8	9,6	3,1	1,6	2,4	<1	<1	<1	1000	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1753	3800	19	53	32%	X	
Product	footwear_spray	shoe spray from shoe repair shop	2,9	9,1	<0,5	9	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	26	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	47	51000	<0,5	0	0%	X	
Product	shoeshine	shoeshine from shoe repair shop	<1	8,4	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	2,2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	11	<100	<0,5	0		Partially dissolved	
Product	leatherspray	leather spray	1,4	2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	12	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	15	44000	<0,5	0	0%		
																																														0			
Product	pan1	PTFE coated	2,3	3,4	<1	<1	<1	<1	<1	<1	<1	<1	<20	<1	<1	<1	<1	<1	<1	<1	1,2	<1	<1	<2	<1	<1	<1	<1	1,6	<1	<1	<1	<2	<2	<2	<2	<500	<1	<1	<1	<1	<1	<1	<1	<1	9	2100	<1	1	0%		
Product	pan2	eTFE coated	<2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<20	<1	<1	<1	<1	<1	<1	<1	<2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<2	<2	<2	<2	<500	<1	<1	<1	<1	<1	<1	<1	<1	0	600	<1	0	0%		
Product	pan3	PTFE coated	<2	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	<20	<1	<1	<1	<1	<1	1	<1	<1	<2	<1	<1	<1	<1	<1	<1	<1	<1	<2	<2	<2	<2	<500	<1	<1	<1	<1	<1	<1	<1	<1	1	<100	<1	1	0%		
Product	baking_mat1	cheap baking mat	<1	<0,5	<0,5	<0,5	<0,5	4,7	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	2,4	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1,8	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	11	640	4,7	5	1%		
Product	baking_mat2	expensive	<1	<0,5	<0,5	<0,5	<0,5	12	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	2,2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	14	940	12	12	1%		
Product	baking_mat3	expensive	<1	<0,5	<0,5	<0,5	<0,5	34	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	3,1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	37	330	34	34	8%		
																																													0			
Product	silicon cupcake	cake	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	<100	<0,5	0	-		
Product	silicon baking mat	baking mat	<1	0,62	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1	<100	<0,5	0	-		
Product	silicon cake	cupcake	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	<100	<0,5	0	-		
																																													0			
Product	paper1	paper table cloth	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	0,66	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1	<100	<0,5	0	-		
Product	paper2	paper table cloth	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	520	<0,5	0	0%		
Product	paper3	glossy paper	<1	2,2	<0,5	1,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	4	<100	<0,5	0	-		
Product	paper4	newspaper	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	0,66	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1	<100	<0,5	0	-		
Process	paper_input1	paper recycling samples input	440	<0,5	<0,5	2,4	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	44	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	1,8	3	9,8	0,64	<0,5	10	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	515	640	2,4	2	55%	X	
Process	paper_input2	paper recycling samples input	2,9	<0,5	<0,5	12	<0,5	2,2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	1,2	<0,5	5,7	1,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1,2	<0,5	14	0,53	<0,5	4,3	<1	<1	<1	<250	<0,5	<0,5	<0,5	0,83	<0,5	<0,5	<0,5	46	550	2,2	2	6%		
Process	paper_input_alternat	paper recycling samples alternative streams input	1300	<0,5	<0,5	2	<0,5	0,72	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	21	0,6	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	0,72	<0,5	19	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	19	<0,5	<0,5	<0,5	1344	340	0,72	1	270%		
Process	paper_process2	paper recycling during process	890	<0,5	<0,5	6,2	<0,5	3,6	2,1	<0,5	<0,5	<0,5	<0,5	<0,5	<10	<0,5	<0,5	3,9	0,73	<0,5	<0,5	<0,5	<1	1,1	<0,5	<1	<0,5	<0,5	3,6	<0,5	22	1,3	<0,5	4,1	<1	<1	<1	<250	<0,5	<0,5	<0,5	1,3	<0,5	<0,5	<0,5	940	860	3,6	6	75%		
Process	paper_output2	paper recycling output	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	4,8	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	5	<100	<0,5	0	-		
Process	paper_process3	paper recycling during process	1300	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	63	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	1,1	1,3	4,9	0,77	<0,5	1,3	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	1384	550	<0,5	0	172%		
Process	paper_output3	paper recycling output	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	16	6,7	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	1,5	1,6	5,2	<0,5	<0,5	7,2	<1	<1	<1	<250	<0,5	<0,5	<0,5	1	<0,5	<0,5	<0,5	41	850	<0,5	0	3%		
Process	paper_input3	paper recycling samples input	<1	<0,5	<0,5	0,67	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	2,3	1,2	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	0,64	0,99	<0,5	<0,5	<0,5	2,8	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	9	<100	<0,5	0	-		
																																													0			
Product	dishwash_1	cubes all-in-one	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	0	590	<0,5	0	0%		Partially dissolved
Product	dishwash_2	cubes all-in-one	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	6,3	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	6	10000	<0,5	0	0%	X	Partially dissolved
Product	dishwash_3	rinsing aid	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	4,3	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	4	1200	<0,5	0	0%		Partially dissolved
Product	dishwash_4	rinsing aid	<1	<0,5	<0,5	<0,5	1,2	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	5,7	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	7	630	1,2	1	1%		Partially dissolved
Product	cleaning_agent 1	sprayer, cheap cleaning agent	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	5,3	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	5	4600	<0,5	0	0%	X	Partially dissolved
Product	cleaning_agent 2	bbq cleaner	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<10	5,9	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<1	<1	<1	<250	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	6	470	<0,5	0	1%		Partially dissolved
Product	cleaning_agent 3	abbrasive	<1	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5	<0,5

APPENDIX B RESULTS TOP-ANALYSES

Appendix B. Results TOP analyses

Before TOP

Barcode	Description	PFBA µg/kg	PFPeA µg/kg	PFHxA µg/kg	PFHpA µg/kg	PFOA µg/kg	PFNA µg/kg	PFDA µg/kg	PFUnDA µg/kg	PFDoA µg/kg	PFBS µg/kg	PFHxS µg/kg	PFOS µg/kg	PFDS µg/kg	Sum Target µg/kg	Sum F µg/kg
IAC20-01854.003	carpetfluff1	15	1,7	16	9,4	40	1,7	1,7	0,59	1,2	10	2,4	66	< 0,5	272	186
IAC20-01854.004	water_repellant1	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	0	0
IAC20-01854.006	Footwear_spray	9,1	< 0,5	9	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	26	< 0,5	< 0,5	< 0,5	47	32
IAC20-01854.016	antifog2	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	0,72	< 0,5	2	1
IAC20-01854.021	lubricant1	33	13	10	9,7	13	7,2	15	15	40	2,8	< 0,5	< 0,5	< 0,5	310	212
IAC20-02617.007	Windshield2	51	230	1,6	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	6,6	< 0,5	308	211
IAC20-02617.008	Floor_polish1	90	16	49	4,7	0,78	< 0,5	0,55	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	188	129
IAC20-02620.004	Outdoor1	8,4	2,3	20	5,3	74	5,1	31	2	11	< 0,5	< 0,5	< 0,5	< 0,5	177	121
IAC20-02620.009	Adh_Sea1	170	34	97	32	0,97	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	0,52	< 0,5	468	320
IAC20-06664.007	Dust_carpet3	79	< 2,5	4,7	3	52	< 2,5	< 2,5	< 2,5	< 2,5	< 2,5	< 2,5	< 2,5	4,1	328	224
IAC20-06664.011	Dust_leather	2,7	< 0,5	6,4	1,2	19	1,3	1,4	< 0,5	1,2	26	1	32	0,69	1753	1199
IAC20-06664.013	Dust_textile2	4,5	4,8	31	15	180	75	18	56	110	0,91	3,7	150	< 0,5	993	679
IAC20-06664.017	Dust_fluopolymer3	69	68	4,4	3,8	100	2,9	1,8	3,9	1,7	1,1	0,76	5,5	< 0,5	1681	1150
IAC20-06664.023	sewage_sludge1	1,2	< 0,5	2,6	0,56	4,3	< 0,5	0,84	< 0,5	< 0,5	5,2	0,51	5,9	< 0,5	71	49
IAC20-09763.001	fluor rubber1	< 0,5	< 0,5	56	3,2	190	4,3	28	2,3	1	< 0,5	< 0,5	< 0,5	< 0,5	302	206
IAC20-09763.009	Dishwash2	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	6,3	< 0,5	< 0,5	< 0,5	6	4
IAC20-09763.014	Cleaning agent4	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	5,3	< 0,5	< 0,5	< 0,5	5	4
IAC20-09763.022	Dust electronic4	< 0,5	< 0,5	0,91	< 0,5	4,4	< 0,5	< 0,5	< 0,5	< 0,5	34	< 0,5	< 0,5	< 0,5	212	145
IAC20-10368.004	cosmetics4	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	< 0,5	24	< 0,5	< 0,5	< 0,5	78	53
IAC20-10475.001	paper_input1	< 0,5	< 0,5	1,9	< 0,5	2,4	< 0,5	< 0,5	< 0,5	< 0,5	44	< 0,5	< 0,5	< 0,5	2115	1446

After TOP

Barcode	Description	PFBA µg/kg	PFPeA µg/kg	PFHxA µg/kg	PFHpA µg/kg	PFOA µg/kg	PFNA µg/kg	PFDA µg/kg	PFUnDA µg/kg	PFDoA µg/kg	PFBS µg/kg	PFHxS µg/kg	PFOS µg/kg	PFDS µg/kg	Sum Target µg/kg	Sum F µg/kg
IAC20-01854.003	carpetfluff1	490	71	69	49	470	< 5	< 5	< 5	< 5	38	< 5	32	< 5	1219	834
IAC20-01854.004	water_repellant1	736	4825	880	13	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	6454	4414
IAC20-01854.006	Footwear_spray	1379	312	70	< 5	< 5	< 5	< 5	< 5	< 5	797	< 5	< 5	< 5	2558	1750
IAC20-01854.016	antifog2	< 50	< 20	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0	0
IAC20-01854.021	lubricant1	59	59	32	< 5	< 5	26	< 5	< 5	< 5	< 5	< 5	< 5	< 5	176	120
IAC20-02617.007	Windshield2	2470	6148	4372	843	< 5	< 5	< 5	< 5	< 5	91	< 5	< 5	< 5	13924	9524
IAC20-02617.008	Floor_polish1	72978	146096	20609	3864	142	100	< 5	< 5	< 5	< 5	< 5	< 5	< 5	243789	166752
IAC20-02620.004	Outdoor1	< 50	620	784	220	101	631	275	< 5	< 5	< 5	< 5	< 5	< 5	2630	1799
IAC20-02620.009	Adh_Sea1	3800	9724	2685	117	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	16326	11167
IAC20-06664.007	Dust_carpet3	990	174	< 10	< 5	< 5	< 5	< 5	< 5	< 5	301	< 5	< 5	< 5	1465	1002
IAC20-06664.011	Dust_leather	591	188	68	52	134	16	12	< 5	< 5	38	< 5	< 5	< 5	1098	751
IAC20-06664.013	Dust_textile2	2368	4357	2725	2621	2841	551	447	< 5	< 5	< 5	< 5	< 5	< 5	15910	10882
IAC20-06664.017	Dust_fluopolymer3	< 50	176	22	< 5	21	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	218	149
IAC20-06664.023	sewage_sludge1	< 50	56	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	56	39
IAC20-09763.001	fluor rubber1	56	151	120	35	176	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	539	368
IAC20-09763.009	Dishwash2	< 50	< 20	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0	0
IAC20-09763.014	Cleaning agent4	< 50	< 20	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0	0
IAC20-09763.022	Dust electronic4	< 50	33	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	33	22
IAC20-10368.004	cosmetics4	< 50	< 20	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0	0
IAC20-10475.001	paper_input1	< 50	< 20	< 10	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	0	0

APPENDIX C CBS DATA

Appendix C. CBS data

Database: 'Verkoop Industriële Producten' (ProdCom)

			2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019	Average
Productcategorie	**Productcode ProdCom**		mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg	mln kg
Zonnewering	13922210								2		5	3	3	4	3
Tenten en Zeilen	13922230 13922250								.	.	.	3	4	3	3
Zepen	20412020 20412030 20412050 20412090 20412120 20412150 20412180 20412240 20412250 20412260 20412270								452	324	595	599	804	716	582
Poetsmiddelen	20414330 20414370 20414383 20414389 20414400								13	13	29	20	.	.	19
Papier	171 en onderliggend								800	942	851	957	985	1010	924
Leder (excl kleding en kunstleder)	15113200 15113300 15113130 15113150 15113230 15113250 15113330 15113350 15115100 15115200 15115230 15115960								4	3	6	4	0	0	3
Tapijten	13931100 13931200 13931300 13931930 13931990								178	196	218	184	218	.	199

Database: 'Gebruik gewasbeschermingsmiddelen in de landbouw; gewas en toepassing'

Toepassingsgroepen: 'Totaal gewasbeschermingsmiddelen'

			2012	2016	Average
Productcategorie	**Specification**		mln kg	mln kg	mln kg
Totaal gewassen/teeltsectoren	Gebruik werkzame stof | totaal		5,88	5,67	5,78

COLOPHON

PFAS IN PRODUCTS AND WASTE STREAMS IN THE NETHERLANDS

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