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Synopsis

Per- and polyfluoroalkyl substances (PFASs) in food contact materials

The term 'food contact materials' describes packaging materials which are used for food and consumer items, such as pans, dishes and baking aids. Per- and polyfluoroalkyl substances (PFASs) are used in the construction of these materials because of their grease repelling qualities. It appears that some of the PFASs used in paper and cardboard can end up in food; this also includes substances whose use in food contact material is not allowed. At present there is not enough information about these PFASs to make reliable exposure estimations and risk assessments. That is the conclusion of RIVM research.

RIVM suspects that these PFASs have ended up in food contact materials because they are present as impurities in the substances used to treat paper and cardboard (the starting materials). Various PFASs can also degrade into these substances. In addition, it is not known whether impurities and degradation products of PFASs are released from food contact materials composed of silicone or rubber.

It is recommended that an investigation be conducted into whether the amount of PFASs that can come from food contact materials in food is harmful to health. In this research, the focus should be placed on the substances contained in paper and cardboard food contact material that might end up in food. This mainly concerns PFASs such as PFOA.

The regulation on the authorisation of food contact materials is complex and only partially harmonised. RIVM recommends that attention be given wherever possible to uniform, harmonised regulation of the authorisation of substances in food contact materials within Europe. RIVM also recommends reviewing old PFAS assessments, because in recent years new information about the harmfulness of these compounds has become available.

Keywords: food contact material, fluorine compound, migration, toxicity, legislation, impurities, degradation, PFCA, FT, PFOA, GenX, PFAS

Publiekssamenvatting

Per- en polyfluoroalkyl verbindingen (PFAS’en) in voedselcontactmaterialen

Voedselcontactmaterialen zijn verpakkingsmaterialen voor levensmiddelen en gebruiksartikelen zoals pannen, servies en bakvormen. PFAS’en worden in deze materialen gebruikt omdat ze vet afstoten. Het blijkt dat sommige PFAS’en die in papier en karton zitten, in voedsel terecht kunnen komen. Hier zitten ook stoffen bij die niet in het voedselcontactmateriaal mogen worden gebruikt. Op dit moment is er onvoldoende informatie beschikbaar om van deze PFAS’en een betrouwbare blootstellingsschatting en risicobeoordeling te maken. Dit blijkt uit onderzoek van het RIVM.

Het RIVM vermoedt dat deze PFAS’en in voedselcontactmaterialen zijn terechtgekomen omdat ze als onzuiverheid in de stoffen zitten waarmee papier en karton wordt behandeld (uitgangsstof). Ook kunnen diverse PFAS’en afbreken tot deze stoffen. Daarnaast is niet bekend of onzuiverheden en afbraakproducten van PFAS’en vrijkomen uit voedselcontactmaterialen van siliconen of rubber.

Aanbevolen wordt te onderzoeken of de hoeveelheid PFAS’en die uit voedselcontactmaterialen in voeding kan komen, schadelijk is voor de gezondheid. In dit onderzoek moet de nadruk liggen op de stoffen in papier en karton en wat daaruit in voedsel kan terechtkomen. Het gaat dan vooral om PFAS’en zoals PFOA.

De regulering van de toelating van voedselcontactmaterialen is complex georganiseerd, en slechts gedeeltelijk geharmoniseerd. Het RIVM raadt aan waar mogelijk aandacht te schenken aan uniforme, geharmoniseerde regulering van de toelating van stoffen in voedselcontactmaterialen in Europa. Het RIVM raadt ook aan om oude beoordelingen van PFAS’en opnieuw te bekijken, omdat de afgelopen jaren nieuwe informatie over de schadelijkheid van deze verbindingen beschikbaar is gekomen.

Kernwoorden: voedselcontactmateriaal, fluorverbinding, migratie, toxiciteit, wetgeving, onzuiverheden, degradatie, PFCA, FT, PFOA, GenX, PFAS

Contents

Summary

The aim of present report is to provide an overview of the available information on per- and polyfluoroalkyl substances (PFASs) in Food Contact Materials (FCMs) in view of potential health risks. PFASs are man-made substances that generally consist of a highly fluorinated carbon chain. PFASs include the well-known PFOA (perfluorooctanoic acid), PFOS (perfluorooctanesulfonic acid) and HFPO-DA (hexafluoropropylene oxide dimer acid, also referred to as GenX). PFASs are being used in FCMs because of their grease and water repellent properties.
Here, the PFASs that are allowed for use in FCMs according to various legal frameworks are listed (see Table 2). This information is compared to the actual presence of PFASs in, and the migration of PFASs out of FCMs. An overview of the toxicity of several PFASs, and information on an approach to assess the risk of the cumulative exposure to a mixture of PFASs is described.

Main findings are:

- A small number of studies indicate that considerable fractions of perfluoroalkyl carboxylic acids (PFCAs) and fluorotelomers (FTs) can migrate from paper and board FCMs into food simulants, especially when the food (simulant) contains alcohol. Several FTs are allowed for use in paper and board, whereas application of PFCAs is not allowed. Some PFCAs in FCMs may be Non-Intentionally Added Substances (NIAS). These NIAS are for example impurities in the technical-grade food contact substances (used to treat the paper and board), the presence of degradation products, contamination via e.g. recycled material or non-compliance with the legislation. Given the toxicological relevance of PFCAs and their ability to migrate from paper and board, further research to enable assessment of their potential health risk and the possible need for further regulatory measures is recommended. The same holds for possible precursors of PFCAs (e.g. FTs and polyfluoroalkyl phosphates (PAPs)).
- There is some information available on migration of PFASs from coatings and paper and board FCM. Information on migration of PFASs from other FCMs is mostly lacking. In particular, there is practically no information available on the presence and migration properties of PFAS impurities. Further research in these areas is recommended.
- Based on their embedding in FCMs and the available information on migration and content of PFASs, highest priority for human health risk assessment of PFASs in FCMs should be on its use in paper and board. Coatings are considered to be of lesser concern, because of the negligible release of PFASs from polymers. The priority for rubber and silicones is unclear because of lacking information and unknown strength of the incorporation of the PFASs into the polymeric matrix.
- Based on the limited data available, a very rough and probably conservative estimation of the cumulative exposure from paper and board FCM can be made for PFOA, PFOS and several other

PFCAs. The exposure to PFOA equivalents can exceed the health based guidance value for PFOA. This estimate is highly uncertain and merely indicates the need for further data.

- High total organic fluoride concentrations can occur in FCMs, which are often expected due to the presence of polymeric PFAS. While the polymeric PFAS are generally considered to be inert, attention should be paid to which and in which extent their degradation products are formed at end-of-life (e.g. after incineration or environmental breakdown). The persistency and potential effects of these degradation products may result in unwanted environmental effects.
- To enable risk assessment, more migration data for all PFASs (including impurities and contaminants) and FCMs is needed. In some cases, also more toxicity data may be required. Risk assessment of PFASs should preferably be performed for the cumulative and aggregated exposure to PFASs. A start to combine the toxicity of a number of PFCAs has been made via Relative Potency Factors (RPFs). Further development of RPFs for other PFASs like FTs and alternatives of PFOA and HFPO-DA, like ADONA, is considered necessary to enable risk assessment of the mixture of PFASs.
- Within the EU, the presence of substances in plastic FCMs are uniformly regulated. However, the non-plastic FCMs are regulated at a national level, and the various national legislations are inconsistent and complex. This is a general finding relating to all substances used in non-plastic FCMs, including PFASs.
- Several PFASs have been allowed in FCMs for decades. The underlying information on risk assessment in these dossiers requires actualization, for example to consider progressing scientific insights with regard to bioaccumulation.
- Like is the case for plastic FCMs, harmonised legislation within the EU for other FCMs would allow for a more effective regulation of PFASs (and other substances) in FCMs. It would also simplify adherence to the regulation and facilitate efficient enforcement. Alternatively, a substance specific regulation on PFASs in FCMs could be developed, similar to the regulations considering e.g. bisphenol-A and epoxy derivatives.
- Little attention is paid to NIAS and their potential health risks in current national and EU legislations on FCMs.
- Regulatory measures that are under development under REACH may impact the use of PFASs in FCMs once these measures will be adopted and are put into force.

List of abbreviations

ADONA	3H-perfluoro-3-[(3-methoxy-propoxy)propanoic acid], ammonium salt
AOF	adsorbable organic fluorine
APFO	perfluorooctanoic acid ammonium salt
CLP	classification and labelling
diPAP	disubstituted polyfluoroalkyl phosphate (see also PAP)
FASA	Perfluoroalkane sulfonamide
FCM	food contact material
FP	fluoropolymer
FRD-902	ammonium-2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoate
FRD-903	2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)-propanoic acid
FT	Fluorotelomer (substance)
FTA	fluorotelomer acrylate
FTCA	fluorotelomer (saturated) carboxylic acid
FTOH	fluorotelomer alcohol
FTS	fluorotelomer sulfonate
FTUCA	fluorotelomer unsaturated carboxylic acid
GC	gas chromatography
HBGV	health-based guidance value
HFC	hydrofluorocarbon
HFE	hydrofluoroether
HFO	hydrofluoroolefin
HFPO-DA	hexafluoropropylene oxide dimer acid
HPCL	high-pressure liquid chromatography
monoPAP	monosubstituted polyfluoroalkyl phosphate (see also PAP)
MS	mass spectrometry
NOAEL	no observed adverse effect level
PAF	perfluoroalkanoyl fluoride
PAP	per/polyfluoroalkyl phosphoric acid ester, per/polyfluoroalkyl phosphate, fluorotelomer phosphate
PASF	perfluoroalkane sulfonylfluoride
PBT	persistent, bioaccumulative and toxic
PFAA	perfluoroalkyl acids
PFAE	per/polyfluoroalkyl ether
PFAI	perfluoroalkyl iodides
PFAL	perfluoroalkyl aldehyde
PFBA	perfluorobutanoic acid
PFBS	perfluorobutane sulfonic acid
PFC	perfluorocarbon
PFCA	perfluorocarboxylic acid
PFDA	perfluorodecanoic acid
PFDoDA	perfluorododecanoic acid
PFDPA	perfluorodecane phosphonic acid
PFDS	perfluorodecane sulfonic acid
PFECA	per/polyfluoroether carboxylic acid
PFESA	per/polyfluoroether sulfonic acid
PFHpA	perfluoroheptanoic acid
PFHpDA	perfluoroheptadecanoic acid
PFHpS	perfluoroheptane sulfonic acid

PFHxA	perfluorohexanoic acid
PFHxDA	perfluorohexadecanoic acid
PFHxPA	perfluorohexane phosphonic acid
PFHxS	perfluorohexane sulfonic acid
PFNA	perfluorononanoic acid
PFOA	perfluorooctanoic acid
PFODA	perfluorooctadecanoic acid
PFOPA	perfluorooctane phosphonic acid
PFOS	perfluorooctane sulfonic acid
PFOSA	perfluorooctane sulfonamide
PFPA	Perfluorophosphonic acid
PFPE	perfluoropolyether
PFPeA	perfluoropentanoic acid
PFPeDA	perfluoropentadecanoic acid
PFPeS	perfluoropentane sulfonic acid
PFPIA	perfluorophosphinate / Perfluoroalkyl phosphinic acid
PFSA	Perfluoroalkyl sulfonic acid
PFSIA	Perfluoroalkyl sulfinic acid
PFTeDA	perfluorotetradecanoic acid
PFTriDA	perfluorotridecanoic acid
PFUnDA	perfluoroundecanoic acid
PIGE	particle-induced gamma ray emission
PPA	polymer production aid
SML	specific migration limit
SVHC	substance of very high concern
TOF	total organic fluorine
vPvB	very persistent and very bioaccumulative

1 Introduction

Per- and polyfluoroalkyl substances (PFASs) are man-made substances that generally consist of a highly fluorinated carbon chain, including the well-known PFOA (perfluorooctanoic acid), PFOS (perfluorooctane sulfonic acid) and HFPO-DA (also referred to as GenX). PFASs have been used for decades and are amongst others applied in food contact materials (FCMs), water-repellent fabrics, waxes, fire-fighting foams and in electronics manufacturing.

1.1 Aim and outline of the research

The aim of present report is to provide an overview of the available information on PFASs in Food Contact Materials (FCMs). To that end, a comparison is made between PFASs that are legally allowed in FCMs to findings described in scientific literature on the presence of PFASs in FCMs and migration out of FCMs. Also, information on the toxicity of PFASs is considered.
Legal frameworks on FCMs - both national and European - use positive lists. If a substance (or its monomer) is on this list, it is allowed for use in FCMs. In most cases, specific restrictions or specifications apply that need to be taken into account. Here, an overview is provided of the different legislations that are applicable to PFASs in FCMs in the Netherlands and the EU (Chapter 3), and the PFASs that are allowed in FCMs according to the corresponding positive lists (Chapter 5). When available, further details from the legislation and dossiers on these substances like restrictions, specifications, migration and toxicity are also provided.
Additional information on the actual presence of PFASs in FCMs (Chapter 6) and migration out of FCMs (Chapter 7) is provided by compilation of experimental findings described in scientific literature. This is shown by an overview of the existing information of the concentration of specific PFASs in various types of FCMs. In a few cases both total organic fluoride (TOF) is determined as well as a number of individual PFASs. These results are discussed in view of the knowledge of the PFASs allowed in FCMs.
Furthermore, the following aspects are briefly addressed:

- the functionality of PFASs in FCMs (Chapter 1)
- the terminology of PFASs and their arrangement into different groups (Chapter 2)
- developments related to PFASs within REACH (Chapter 3)
- analytical methods for individual PFASs and TOF determination (Chapter 4)
- the toxicity and health based guidance values of a few well-known PFASs (PFOA, PFOS, HFPO-DA (i.e. GenX) and ADONA) (Chapter 8)
- the relative toxic potency of a set of 20 PFASs (Chapter 8)
- incineration and persistency of PFASs (Chapter 9)

In Chapter 10, issues related to the subjects above are discussed in view of their relevance for potential risks.

1.2 Why are PFASs used in food contact materials?

Per- and polyfluorinated substances are used in food contact materials mainly because of their fat repellent properties (Borg and Ivarsson, 2017; Trier *et al.*, 2017). Combined with their chemical and thermal stability, PFASs are ideal to be used in non-stick coatings on cookware, in rubbers and silicones used in items which have to endure high temperatures, like silicon baking aids, and in paper and board that are used in contact with fatty food, like in fast food containers, microwave popcorn bags, pizza boxes, butter paper, wrappers for bread and candy. Although the reason for using PFASs in the various food contact materials is usually the same, its technical function and migration potential varies with the type of material and the PFAS used.

1.2.1 *Technical functionality of different PFASs*

In coatings in kitchenware FCMs, PFASs are mainly used as monomers, forming the core chain of fluoropolymers at curing. For instance, the coating of frying pans is often a polytetrafluoroethylene (PTFE, also known as Teflon) produced by polymerization of the PFAS tetrafluoroethylene. The polymers formed have high molecular weights (>100,000 D) and contain negligible residual quantities of monomers and oligomers, and therefore migration of the monomer or related oligomers is very low (Henry *et al.*, 2018) (see Section 2.1 for further explanation on monomers and oligomers). Because PTFE is poorly water soluble, the polymerization is conducted in an emulsion in water. This requires the presence of an emulsifier, such as the surfactant PFOA and HFPO-DA (Wang *et al.*, 2013). At the end of the polymerization reaction, the aqueous part of the mixture, including most of the surfactant, is removed. So, besides the use as monomers, some PFASs are used as 'polymerisation production aid' in coatings. Production aids are substances that are added to the polymer, but not with the intention to be present or have a technological function in the final product. The surfactant does not become part of the polymer chain and does not convey specific properties to the polymer. As the fluoropolymers are applied (on metal surfaces) under high temperature (>> 100^{o}C), residual low molecular weight PFASs will further be removed by evaporation. Other PFASs like HFPO-DA (partly) decompose during heating to other PFAS substances (HFPO-DA decomposes to E1 (Beekman *et al.*, 2016)).

Coatings for paper and board are different from fluoropolymer coatings used in kitchenware. The PFASs used in paper are not acting as monomers or production aids, but as additives. In contrast to production aids, additives are substances that are intended to be present in the final FCM product, as they have physical or chemical functions in it, i.e. in the case of PFASs in paper and board, to repel water and oil. PFASs can be added to the pulp ('internal sizing') or alternatively are used as a surface treatment on the paper or board ('external sizing') (Trier *et al.*, 2017). The PFASs used in paper and board are surfactants (Trier *et al.*, 2011). Their polar heads bind to the cellulose fibres in the pulp or paper, and the hydrophobic tails form side chains of the cellulose fibre (see also Figure 8). The bonding can be due to strong electrostatic bonds like between anions and cations, but more often the PFASs are bound covalently to the hydroxyl group of the cellulose (chemisorption) by

forming ester bonds when the paper is heated and dried. These ester bonds are however reversible, as they can break due to hydrolysis. This particularly occurs if paper is in contact with alcohol or warm water, and this can lead to migration of PFAS from the paper and board into the food (Trier *et al.*, 2017).
Some additives in paper and board are ‘polymeric additives’, which refers to molecules that contain some repeating units but the substance is not added to polymerize any further, rather it will be present in the material as additive. By the production of polymeric additives, usually some other PFASs (precursors or by-products) are also present as impurities in the final product (see Chapter 7). These other PFASs can also arise from degradation of the polymeric substance, which occurs in time.

2 Terminology and structure groups of PFASs

This chapter describes which substances are considered to be PFAS, followed by a description of the terminology and structure groups of PFASs. These groups are used in further discussions.

2.1 Definition of PFAS

A single, globally harmonized system for PFAS classification has not (yet) been defined, resulting in lack of recognition of important distinctions between PFASs. Perfluorinated and polyfluorinated alkyl substances (PFASs) are generally defined as substances that contain one or more perfluoroalkyl moieties, $–C_nF_{2n+1}$. The fluorinated alkyl moiety of PFAS substances may contain linear, branched or cyclic carbon chains. A large number of fluorinated substances with diverse properties meet this requirement. In addition, compounds not having a $–C_nF_{2n+1}$ moiety, but which are produced from PFASs or which may degrade to compounds containing the moiety are generally also considered as PFASs (e.g. several polymers). In their 2018 database on PFASs, OECD considered substances as PFASs when containing a perfluoroalkyl moiety with three or more carbons (i.e. $-C_nF_{2n}–$, n ≥ 3) or a perfluoroalkylether moiety with two or more carbons (i.e. $-C_nF_{2n}OC_mF_{2m}–$, n and m ≥ 1) (OECD, 2018). OECD adapted the definition to include substances with both ends of the perfluoroalkyl moiety connected to a functional group[1] and cyclic analogues of linear PFASs[2]. In the Sections below the PFAS classifications of Buck *et al.* (2011), OECD (2013; 2015; 2018) and KemI (2015) are described and compared. **In this report we will consider the presence of PFASs in food contact materials, where PFASs are defined according to OECD 2018, i.e. substances with a perfluoroalkyl moiety ($–C_nF_{2n}–$ with n ≥ 3) or a perfluoroalkylether moiety ($–C_nF_{2n}OC_mF_{2m}–$ with n and m ≥ 1), and substances that may degrade to form them (precursors).** PFASs are divided into two sub-groups: non-polymeric and polymeric PFASs (see Figure 1 and Figure 2). Both of these groups are relevant for FCM. It is assumed that PFASs present in coatings and plastics mainly comprise of polymeric PFASs. However, non-polymeric PFASs may be present as residues of fabrication or as degradation products from polymers, or as applied in the production of water- and grease-repelling paper and (card)board.

2.2 Non-polymer PFASs

Within the non-polymeric PFAS group, ten groups are defined by Buck (2011) (Figure 1):

- (Aliphatic) perfluorocarbons (PFCs);
- Perfluoroalkyl acids (PFAAs);
- Perfluoroalkane sulfonyl fluorides (PASFs);
- Perfluoroalkane sulfonamides (FASAs);
- Perfluoroalkyl iodides (PFAIs);

[1] E.g. perfluoroalkyl dicarboxylic acids: HOOC-C_nF_{2n}-COOH.
[2] E.g. undecafluorocyclohexanesulfonic acid potassium salt, CAS No. 3107-18-4.

- Perfluoroalkyl aldehydes (PFALs) and aldehyde hydrates (PFAL·H_2Os);
- Perfluoroalkanoyl fluorides (PAFs);
- Perfluoroalkane sulfonamido derivatives;
- Fluorotelomer (FT)-based compounds, incl. semifluorinated n-alkanes (SFAs) and alkenes (SFAenes);
- Per- and polyfluoroalkyl ether carboxylic acids (PFECAs)[3].

OECD (2013; 2015) merged several of these groups (see Figure 1) and concluded that four main groups are involved:

- Perfluoroalkyl acids (PFAAs);
- PASFs and compounds derived from PASFs, i.e. the FASAs and perfluoroalkane sulfonamido derivatives as mentioned by Buck;
- PFAIs, fluorotelomer iodides (FTIs) and fluorotelomer (FT)-based compounds (including the PFALs, SFAs and SFAenes);
- Per- and polyfluoroalkyl ether-based compounds (PFAEs).

In 2018 OECD rearranged their PFAS groups. The PFAA group was also considered to cover per- and polyfluoroalkylether acids (Figure 1, red dashed box), i.e. those acids with the acidic functional group(s) directly connecting to a per- or polyfluoroalkylether chain. Other PFAEs and all PASF and PFAI related substances were designated as PFAA precursors (Figure 1, yellow dashed box). Several new groups (Figure 1, blue dashed box) were included containing highly fluorinated substances which were considered to match the definition of PFASs. However, (some) PFASs in these new groups do not have surfactant properties and are no precursors of PFAAs. Substances that were considered PFAS by OECD (2018), due to changing the definition of a perfluoroalkyl moiety from $-C_nF_{2n+1}$ to $–C_nF_{2n}–$, are substances with both ends of the perfluoroalkyl moiety connected to a functional group (e.g., perfluoroalkyl dicarboxylic acids, HOOC-C_nF_{2n}-COOH), cyclic analogues of linear PFASs, e.g. $C_6F_{11}SO_3K$ (CAS no. 3107-18-4), and substances with one of the fluorine atoms in the alkyl chain substituted by another atom, e.g. F-53B (CAS no 73606-19-6).
(Linear) PFCs (C_nF_{2n+2}), also known for their ozone depletion when gaseous, are considered as PFAS by Buck (2011) because “Those PFCs that contain a $–C_nF_{2n+1}$ moiety are, by definition, members of the PFAS family”. However, Buck *et al.* also state that PFCs are chemically very stable substances, and it is uncertain whether any of them can actually degrade in the environment to give functionalized PFASs. OECD (2013; 2015) did not seem to consider PFCs as PFASs, because PFCs “….contain only carbon and fluorine and have properties and functionalities different from those of PFASs”. However, in 2018, OECD included PFC in their overview of PFAS groups. KemI (2015) includes branched and/or cyclic PFCs in their survey because they may degrade to per- or polyfluorinated surfactants.
The PFAAs are generally subdivided into four groups of surfactants:

- Perfluoroalkyl carboxylic acids (PFCAs, e.g. perfluorooctanoic acid (PFOA));
- Perfluoroalkane sulfonic acids (PFSAs, e.g. perfluorooctane sulfonic acid (PFOS));

[3] Abbreviation not mentioned by Buck (2011), but by OECD (2015).

- Perfluoroalkyl phosphonic acids (PFPAs);
- Perfluoroalkyl phosphinic acids (PFPIAs);

In addition Buck (2011) also identifies perfluoroalkane sulfinic acids (PFSIAs) as a separate PFAA group, which is an intermediate in the production of (PFSA). In 2018, OECD included the per- and polyfluoroalkylether acids in the PFAA group.
The PAFs are not mentioned by OECD (2013; 2015; 2018). They are intermediates in the manufacturing (by electrochemical fluorination) of PFCAs.
The (sub)group of per- and polyfluoroalkyl ether carboxylic acids (PFECAs) has, according to Buck (2011), a terminal $–COO^-$ group in common. This group is attached to one or both ends of the fluorinated ether chain, and can have an H^+ or NH_4^+ counter ion. OECD (2013; 2015) included the PFECAs in a more general ether-based group. This group was abbreviated by OECD 2013 as PFPE, suggesting that it contains degradation products of perfluoropolyethers. As the abbreviation PFPE was already designated to perfluoropolyethers by Buck and OECD 2015 and 2018 (2015; 2018), this document will abbreviate the per- and polyfluoroalkyl ethers to PFAEs. According to OECD 2015 (2015) the PFAEs can be subdivided into carboxylic acids (PFECAs, e.g. HFPO-DA (i.e. GenX) and ADONA) and sulfonic acids (PFESAs). As already mentioned, OECD (2018) added the per- and polyfluoroalkyl ether acids (PFECAs and PFESAs) to the PFAA group and considered per- and polyfluoroalkyl ether based substances as PFAA precursors. It has been suggested that PFECAs and PFESAs have similar properties, including persistence, to PFCAs and PFSAs with longer carbon chains (Gomis *et al.*, 2015).
KemI mentioned the group of perfluoroalkyl ethers (PFECAs), in which several short perfluoroalkyl chains are linked to each other by ether bridges. If there are several (>2, (OECD, 1993)) monomers bound to at least one other monomer by ether bridges in the same chain, the substance is considered a polymer: perfluoropolyether (PFPE, see below). However, others consider >2 and <10 repeating units as oligomers and in case of about >10 repeating units as polymers (Klaerner and Padmanabhan, 2001).

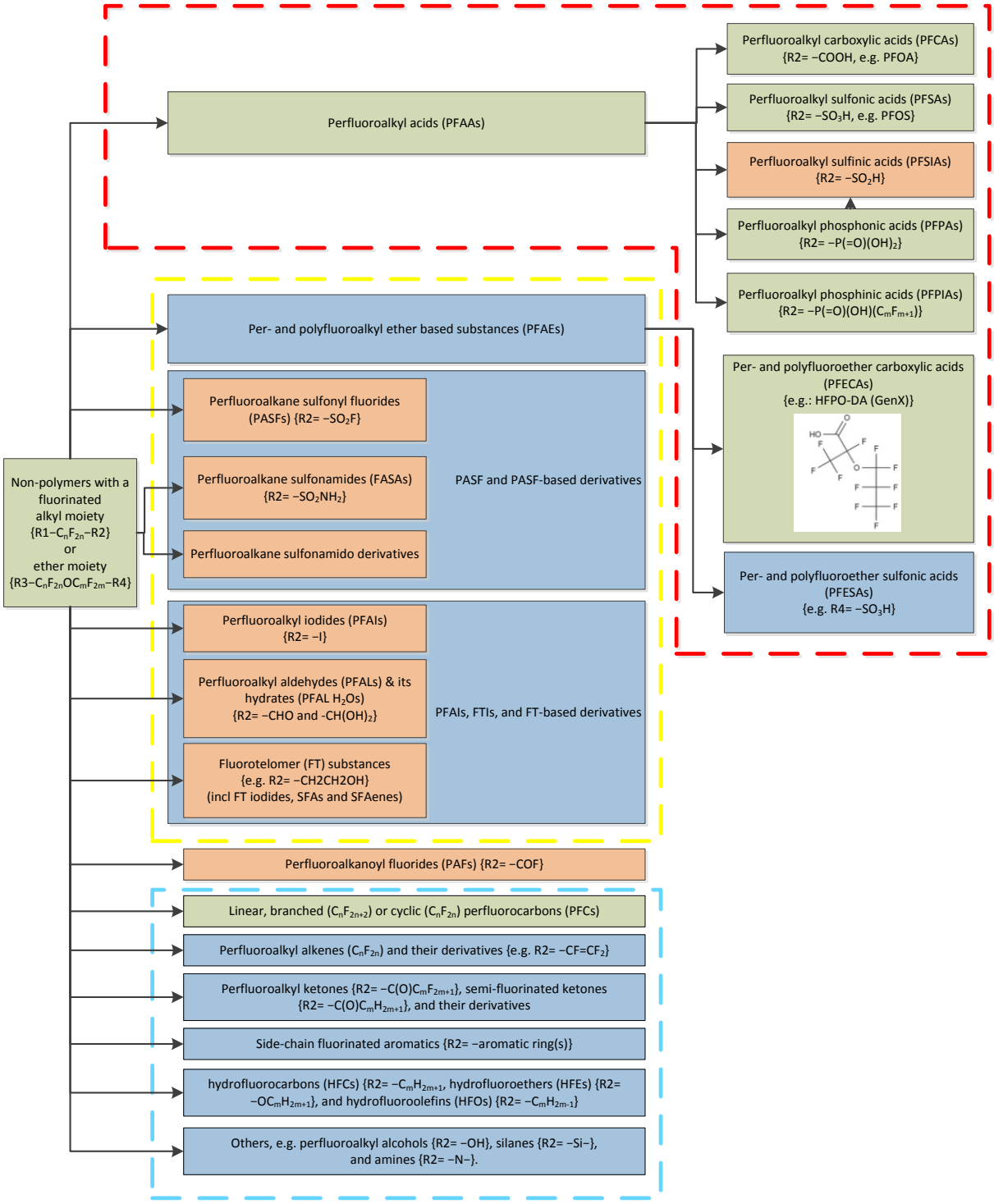


Figure 1: Groups of non-polymer per- and polyfluoroalkyl substances (PFASs) based on OECD (2013; 2015; 2018) and Buck et al. (2011). Green: groups are described by OECD and Buck et al. Orange and blue: group described only by Buck et al. (2011) or only by OECD (2013; 2015; 2018) respectively. Dashed blue boxes contain groups that are regarded by OECD (2018) as other high fluorinated substances matching the definition of PFASs. Dashed yellow and red boxes include groups which are designated by OECD (2018) as PFAA precursors and PFAAs respectively. In Buck et al. (2011) and OECD (OECD, 2013; 2015)

the definition of a PFAS containing a –CnF2n+1 moiety, required R1 and R3 to be a fluorine atom. In OECD (2018) R1 and R3 can be any single atom or a larger moiety (e.g. –COOH, see text).

2.3 Polymeric PFASs

The polymers considered as PFAS are those: 1) whose synthesis involves the incorporation of one or more PFASs as monomers. In this case, there is some potential (theoretical or demonstrated) for the degradation of the polymer, during or after its useful lifetime, to lead to release of PFASs to the environment; or 2) whose manufacture requires the use of a PFAS as a production aid (Buck *et al.*, 2011).
Within the polymeric PFAS group Buck *et al.* (2011), OECD (2013; 2015; 2018) and KemI (2015) all differentiate between (Figure 2):

- Fluoropolymers: fluorinated polymers consisting of carbon-only backbone with fluorines directly attached to this backbone, formed from the following monomers or other starting substances e.g.:
  - Polytetrafluoroethylene (PTFE);
  - Polyvinylidene fluoride (PVDF);
  - Fluorinated ethylene propylene (FEP);
  - Perfluoroalkoxyl polymer or (PFA);
  - etc.;
- Side-chain fluorinated polymers: fluorinated polymers consisting of variable compositions of non-fluorinated carbon backbones with polyfluoroalkyl (and possibly perfluoroalkyl) side chains. The fluorinated side-chains, including PASF- and fluorotelomer-based derivatives, are potential precursors of PFAAs (Liu and Mejia Avendano, 2013). The following three side-chain polymers are mentioned;
  - Fluorinated (meth)acrylate polymers;
  - Fluorinated urethane polymers;
  - Fluorinated oxetane polymers.
- Perfluoropolyethers (PFPEs): fluorinated polymers consisting of backbones containing carbon and oxygen with fluorines directly attached to carbon. They are not made from PFAAs or their potential precursors; and PFAAs or their potential precursors are not involved in the manufacturing of perfluoropolyethers.

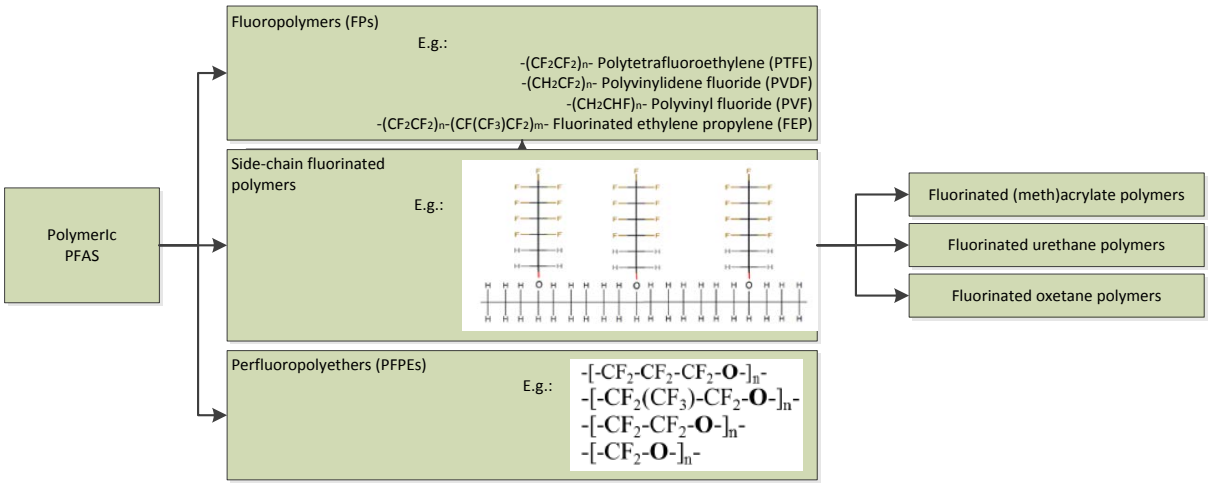


Figure 2: General classification of polymer per- and polyfluoroalkyl substances (PFASs), based on OECD (2013; 2015; 2018) and Buck et al. (2011). Green: groups are described in both references. However, OECD 2018 does not mention the side-chain fluorinated polymers as a separate group.

3 Overview regulatory developments and requirements for PFASs

3.1 Overview of legal requirements for substances in FCMs

Food contact materials are used for packaging of food but also include utensils such as cutlery, plates and cups as well as machinery used in a food factory or at home (e.g. a coffee machine). Materials in contact with food are for example plastics, coatings, paper and board, rubbers, and silicons, but other materials are used as well. Because substances can migrate from materials into the food, food contact materials have to meet certain requirements, which are laid down in European and national legislation. On the European level, all food contact materials are regulated under the Framework Regulation (Reg. EC No. 1935/2004 (EC, 2004)), in which general requirements are established. These include the requirement of Good Manufacturing Practice (GMP) and that materials must be sufficiently inert to preclude contamination of the food that could endanger human health.
In addition to the Framework Regulation, a number of specific directives are established on the European level that are either on specific materials or on specific substances. There is no such substance specific regulation for PFAS. The specific directives regulate the authorisation of (individual, or groups of) substances used in the food contact materials. Moreover, they restrict their migration, either by setting maximum use levels, but most often in the form of so-called “specific migration limits” or SML’s. In the European legislation, restrictive lists of authorized substances (‘positive lists’) have only been specified for monomers and additives used in plastics (EU, 2011), and substances used in regenerated cellulose (EC, 2007). The positive list for plastic FCM includes several specific PFASs (see Chapter 5). There are no PFASs included in the positive list for regenerated cellulose.

For all other materials, national legislation is applicable, which may or may not include positive lists of substances. In the Netherlands, FCMs are regulated by the “Regulation on Packaging and Consumer Articles of the Commodities Act” (Warenwetregeling Verpakkingen en Gebruiksartikelen (WVG, 2014)). The Dutch Regulation includes positive lists for: paper and paperboard, coatings, rubber, metals, wood and cork, and textiles, and it even includes an additional list for plastics, i.e. for substances used as “aids to polymerisation” and “polymerisation production aids”; both groups of substances are not specifically regulated in Reg. EU (No.) 10/2011. In the Dutch regulation, PFASs are only included on positive lists for ‘paper and board’, ‘coatings’ and ‘rubber’ (see Chapter 5).

To set acceptable limits for substances, a risk assessment is performed to check if no adverse health effects will occur. For this, the manufacturer must submit a migration and toxicity dossier with data on the substance, according to the “EFSA Note for Guidance for petitioners” (EFSA, 2008c). In case of no or very limited migration, only genotoxicity studies are needed; no further toxicological data is required. If migration however exceeds 0.05 mg/kg, additional toxicity data for derivation of

an Acceptable Daily Intake (ADI) are required. Alternatively, if absence of potential for accumulation in man is demonstrated, and the migration is below 5 mg/kg, then besides the genotoxicity data, only an additional subchronic (90-day) repeated dose study in rodents is sufficient for the listing of the substance.
For substances listed or specifically regulated in European legislation, risk assessment is performed by EFSA in what is now called the CEP Panel (Panel on Food Contact Materials, Enzymes, and Processing aids). For national registration, the risk assessment is performed by the "Commission G4".

Besides European and Dutch FCM-legislation, the principle of Mutual Recognition applies. This means that products that have been lawfully manufactured or placed on the market in another Member State of the European Union (or in a State which is party to a customs Union Treaty or Free Trade Area Treaty), and which meets requirements that provide a level of protection at least equivalent to the level of the pursued national requirements, are allowed to be placed on the Dutch market as well. For instance, products that are manufactured with substances listed in the BfR Recommendations[4] and complying to its restrictions, can be legally on the market in Germany and could be recognised as legally on the market in the Netherlands as well.

In Chapter 5, an overview is provided on the PFASs that are allowed in materials and articles intended to come into contact with food.

3.2 Developments related to PFASs within REACH

3.2.1 *General overview*

This section provides an overview of the regulatory actions for PFASs in the context of European REACH and CLP Regulation. REACH comprises Registration, Evaluation, Authorization and Restriction of Chemicals, and CLP stands for Classification, Labelling and Packaging of substances and mixtures. Table 1 provides an overview of the ongoing regulatory actions on perfluorinated carbons under the REACH Regulation (EC No 1907/2006).

[4] https://BfR.ble.de/kse/faces/DBEmpfehlung_en.jsp

Table 1: Regulatory actions under the REACH Regulations (dd. February 2018)[5].

Subgroup	PFASs group (includes precursors where relevant)	RMOA	Assessment	CLH / SVHC	Restriction
Long-chain PFCAs	PFOA	√	√	√ / √	√
	PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTDA	√	√	√ / √	Under preparation*
	C_{11}-C_{14}-PFCAs	√	√	- / √	Under preparation
	PFHxS	√	√	- / √	Under preparation*
	PFHpA (C_7-PFCA)		On-going (including testing)		
Short-chain PFCAs	PFHxA (C_6-PFCA)	√*	√*	Withdrawn*	Under preparation*
	PFBA (C_4-PFCA)	√*			
Short-chain PFSAs	PFBS (C_4-PFSA)	√*		On-going*	
Perfluoroether carboxylic acids (PFECA)	ADONA HFPO-DA (GenX, FRD-902) 2 other related subst.	√*	On-going On-going*	On-going*	
Perfluoropolyethers (PFPEs)	TFEE-5		On-going		

* Adapted by authors to account for most recent developments (December 2018).

Restrictions and Authorizations are laid down in Annex XVII and Annex XIV of the REACH Regulation respectively. Restrictions under REACH can be proposed for substances or groups of substances for which there is an overarching risk for society at a European scale due to its production or the use. A Restriction on the use of a particular substance in articles applies both on articles produced within the EU and articles produced outside the EU that are imported. Without further notice, Restrictions of substances do apply to substances used in FCM materials[6]. For

[5] As presented in *Appendix II, Overview of current and past REACH and CLP regulatory activities on PFASs* that is extracted from the RiME-I 2018 Concept paper – common needs and options for regulatory risk management of short-chain PFASs from the PFAS working group; discussed at the Risk Management Expert Meeting , RiME-1/2018, Uppsala, Sweden, 13-14 February 2018.

[6] Proposals for Restrictions for substances contain a Part I that includes the scope of the proposal and the resulting hazard and risk assessment, and a Part II that contains a socio-economic analysis of the impact of the

substances on the Annex XIV, the Authorization regime does not apply to certain specific uses of a substance for which there is other legislation available that already sufficiently controls its possible risks. This is partly the case for food contact materials. FCM substances that are on Annex XIV due to their human health hazards are exempt from authorisation because EU food contact material legislation specifically controls health risks. However, when these substances are on Annex XIV due to their environmental hazards, these substance do have to be authorised for use in FCM. This includes substances with PBT or vPvB properties, as set out by REACH Annex XIII, or substances which, according to scientific evidence, give rise to an equivalent level of environmental concern when this involves environmental health effects. For PFAS it is therefore expected that, once these substances are included in Annex XIV, authorization will have to be requested for their use in FCM materials. Authorization is required when the concentration of the PFAS is above 0.1 w/w% (most cases) within the mixture and does not apply to PFAS in imported articles.
Further, already the SVHC identification through Candidate Listing (which precedes Annex XIV listing) will influence the communication on hazard properties of these substances. This is also the case when their particular use and hazard concern is outside the scope of Authorization. When a substance used in FCMs is identified as SVHC, substance suppliers shall inform their recipients with the appropriate information to use or work safely with the SVHC substance according to article 32. In addition, suppliers of articles, containing the substance above 0.1 w/w %[7] or 0.3 w/w %[8], need to inform the recipient of the article sufficiently in order to use the article safely according to article 33. Furthermore, producers and importers of articles containing SVHC substances above these concentration levels will have to notify ECHA of their presence in articles according to article 7(2). However, as authorisation is only applicable above 0.1 w/w % and PFASs are often present and toxicologically relevant at concentrations below 0.1 w/w %, restriction is considered a better regulatory measure as it allows setting much lower concentration limits.

3.2.2 *Perfluor Restrictions*

PFAS substances with a harmonized classification as reproduction toxicant under CLP (Repro Cat. 1B[9]) are restricted under REACH according to entry 30 of Annex XVII. This entry regulates the supply to the general public of reproductive toxicants. These substances may thus not be provided to the general public on their own, as constituents of other substances, or, in mixtures in a concentration of 0.3% and above.

To date, there is only one restriction formulated specifically for perfluor compounds: entry 68 of Annex XVII for Perfluorooctanoic acid (PFOA;

proposed Restriction. These proposals are open for public consultation. During this public consultation phase, all societal actors are invited to comment on the proposal or add information that could shed a new light on the proposal. When a proposal may be in potential conflict with the FCM regulation, this may be flagged by i.e. the responsible authorities. The RMOA phase (Risk management option analysis) that precedes the development of proposals for further regulatory measures under REACH is a first moment where these types of interferences or possibilities of alternative regulatory options are identified. When there is sufficient ground that safe use is possible for FCM, a derogation for use in these materials can be included in a restriction.

[7] SVHCs according to articles 57(a-b) and 57(d-f)

[8] SVHCs according to article 57(c)

[9] Annex VI CLP Regulation EC/1272/2008

EC 206-397-9) and its salts. Entry 68 is special in a way that it not only refers to a specific substance and its salts (PFOA), but also to substances that may lead to similar biodegradation products[10]:

- *Any related substance (including its salts and polymers) having a linear or branched perfluoroheptyl group with the formula C_7F_{15}- directly attached to another carbon atom, as one of the structural elements.*
- *Any related substance (including its salts and polymers) having a linear or branched perfluorooctyl group with the formula C_8F_{17}- as one of the structural elements.*

Entry 68 will apply from 4 July 2020 onwards to the manufacturing and placing on the market of these substances on their own, or their use in other substances, mixtures or articles to a concentration equal to or above 25 ppb of PFOA including its salts or 1000 ppb of one or a combination of PFOA-related substances. Hence, from 2020 onwards, these manufacturing and placing on the market activities are no longer allowed. Several uses are excluded from the restriction or will enter into force only later. **Because restrictions apply to food contact materials and food contact materials are not explicitly excluded from this entry, the limits specified (e.g. 25 ppb of PFOA including its salts or 1000 ppb of one or a combination of PFOA-related substances) also apply to food contact materials.** Similar types of restrictions are currently in the making for PFNA, PFDA, C_{11}-C_{14}-PFCAs, and PFHxS, and are being discussed for the short-chain perfluorinated carbon PFHxA. As a first step, their PBT and vPvB properties are being assessed. It is expected that for those substances and groups of substances for which PBT or vPvB can be concluded, Candidate listing will precede any further regulatory follow-up measures. It is also in the line of expectation that in parallel to SVHC identification, further proposals for restriction will be submitted in the upcoming years leading to the further regulation of both long and short chain perfluorinated carbons.

3.3 POP-regulation

PFOS is regulated as a persistent organic pollutants (POPs) under the European Regulation (EC) No 850/2004. This Regulation prohibits, phases out or restricts the production, placing on the market and use of POP substances subject to the Stockholm Convention on persistent organic pollutants (POPs) and/or the 1998 Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Persistent Organic Pollutants. Restrictions laid down in the POP Regulation also apply to the use of these substances in FCM, unless specified differently. The POP Regulation also applies to waste treatment, recovery, recycling, reclamation or re-use of POPs. PFOS was added to the Annex B of the Stockholm Convention in 2009 and subsequently added to Annex I of the European Regulation (EC) No 850/2004 (List of substances subject to prohibitions). The amendment to this Regulation in 2010 (EU, 2010) lays down a prohibition of PFOS to 0.001 w/w% or below in substances or preparations and to 0.1 w/w% in articles and 1 µg/m^2 on coated

[10] Excluded from entry 68 are: C_8F_{17}-X, where X = F, Cl, Br, and C_8F_{17}-C(=O)OH, C_8F_{17}-C(=O)O-X′ or C_8F_{17}-CF_2-X′ (where X′ = any group, including salts).

articles. This implies that for FCM, PFOS is prohibited in FCM at concentrations above 0.1 w/w%, or 1 µg/m^2 in case PFOS is applied as a surface coating. Several derogations apply to this prohibition. The regulation does not contain specific derogations for PFOS in FCMs.

4 Analytical methods

This chapter provides a short overview of generally used quantitative detection methods (Trier *et al.*, 2017; Trojanowicz and Koc, 2013). PFASs can be measured as individual PFAS or as the non-specific total amount of fluorine.

The following paragraphs show that only a limited number of PFASs can be measured analytically and these measurement are technically challenging. TOF measurements do not allow identification on individual PFAS and may also comprise other fluor-containing substances.

4.1 Total fluorine

Several non-specific methods exist which can screen for total fluorine, which can also comprise other substances that contain fluoride than PFAS. These include sliding spark, droplet test, ^{19}F NMR (Moody *et al.*, 2001), Raman spectroscopy, Particle-Induced Gamma Ray Emission (PIGE) spectroscopy and combustion of the total organofluorine followed by measurement of fluoride by electrode or ion chromatography. Only the combustion ion chromatography (CIC) and PIGE methods seem sufficiently quantitative to be used as a confirmatory enforcement or compliance method for detection of the total amount of fluorine (TOF).
In the CIC method, the inorganic fluoride content of a sample is measured by ion chromatography in a liquid extract and the total (organic plus inorganic) fluoride content is determined by ion chromatography after extraction and combustion (mineralization) of the sample (Miyake *et al.*, 2007) or after combustion of the entire (solid) sample (Trier *et al.*, 2017). The organic fluorine content is determined by subtraction of the inorganic fluoride from the total fluoride content. A drawback of this method is that if the inorganic fluoride concentration largely exceeds the organic fluorine concentration (as is typically the case in groundwater and drinking water), then the two large number are subtracted from each other, which results in a low precision in the determination of organic fluorine.
To improve the method for aqueous samples an adsorbable organic fluorine (AOF) method was developed (Fritsche and Hüttenhain, 1994; Wagner *et al.*, 2013). The organofluorines and inorganic fluoride were extracted from the water by adsorption onto a solid sorbent, followed by removal of the inorganic fluoride. Subsequently, the total organic fluorine was determined.
In the PIGE (Ritter *et al.*, 2017) method, a solid sample is bombarded with a beam of protons. The resulting gamma rays are measured. Two gamma rays characteristic of the decay of the ^{19}F nucleus are integrated and the total number of counts in these two peaks was proportional to the total fluorine concentration.

4.2 PFAS-specific methods

Determination of individual PFAS can be subdivided into targeted and screening analysis. In target analyses, the compound(s) of interest is known, while in a screening analysis the aim is to determine which

PFASs are present in the sample without predefining set of PFASs (Trier *et al.*, 2017).
In target analyses, the compound of interest is known, and a pure analytical reference compound (a “standard”) should be available. Target analyses are used in confirmatory research, i.e. to determine the concentration of a particular (predefined) set of PFASs in a sample. This information may, for example, be used to perform risk assessments or to determine whether or not a sample complies with present limit values. PFASs are typically quantified by methods coupling liquid or gas chromatography (LC or GC) to a mass spectrometric (MS) detector. The purpose of the chromatography is to separate the mix of PFASs in a sample from each other, so they ideally arrive one at a time at the MS detector.
Analytical and isotopically-labelled standards can be bought for a limited number of PFASs. For these PFASs it is possible to make target methods, typically using liquid chromatography methods (e.g. (U)HPLC-MS/MS, LC-(ESI-)MS/MS), whereas volatile PFASs have been mostly analysed by gas chromatography (GC) MS methods (Trier *et al.*, 2017; Trojanowicz and Koc, 2013).
When screening for PFASs, all peaks in the sample need to be identified. The PFASs for which a standard is available are relatively easy to identify (using target analysis). However, identifying PFASs without a standard is a time-consuming and iterative process (Trier *et al.*, 2017, and references therein). To limit the number of possible chemical structures, it is therefore advantageous prior to the chemical analyses to assemble as much information as possible about the substances which are likely to be present in the sample.
A first step in a non-target analysis is typically to screen for likely or suspected target contaminants called “semi-target” analysis. The search is often done by setting the detector to look only at signals specific to the analyte of interest. By identifying some of the peaks, the remaining number of unknown peaks can be reduced.
After identification, peaks can be quantified. If standards are unavailable, the PFASs can at best be semi-quantified. This is typically done by quantifying the PFAS using the calibration curve for a known and structurally similar PFAS compound (e.g. Gebbink *et al.*, 2017; Gobelius *et al.*, 2018).

5 PFASs allowed in FCMs

In this chapter an overview is provided on the PFASs which are allowed in materials and articles intended to come into contact with food. Several regulations on FCMs were searched on PFASs, as defined by OECD (see Chapter 2). In addition (fluorinated) monomers, which can be used to produce the fluorinated polymers described in Chapter 2 are also identified. The obtained PFASs and monomers are listed in Table 2. Note that, although not listed, salts of authorised acids, phenols or alcohols are allowed to be used as well.

In total 33 PFASs have been identified on positive lists, which are mentioned in the various legislations. The reviewed legislations are summarized in Appendix A. Several of these entries comprise groups of PFASs, e.g. substances with fluorinated carbon chains of varying length. For 31 of these entries a CAS number is available, or at least one of the substances in a group could be identified with a CAS number.

Plastic and coatings regulations contain 21 PFAS entries, combining all legislations. The main application is their use in coatings. In Reg. 10/2011, the use of PFASs as additive or polymerisation production aid is restricted to repeated use articles that are sintered or processed at high temperatures, which seems to limit their use more or less to coatings. The use of PFASs as monomers is authorised without such restrictions, and therefore allows their use in non-coating plastics as well. They are used for example to produce release agents used in polyethylene.
Paper and (card)board regulations mention 12 PFAS entries, and rubber and silicone FCM regulations contain five and one entries, respectively. The sum of the number of entries for each FCM regulation adds up to more than 33, because several PFASs were mentioned in multiple legislations.

Of the 33 entries, 14 PFASs are assumed to be used as an additive or polymer production aid. Fourteen PFASs are indicated as monomers or other starting substances, and 5 entries consider polymers. The regulations of 10 out of the 33 PFASs entries report a specific migration limit (SML).

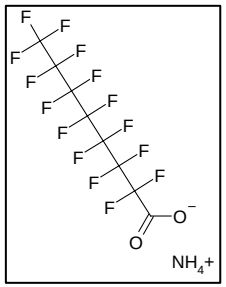
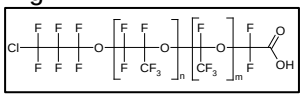
5.1 PFASs in FCMs allowed according to regulations

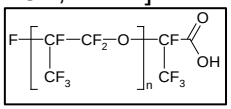
The PFASs that are allowed in FCMs according to positive lists are presented in Table 2. The table contains the following information:

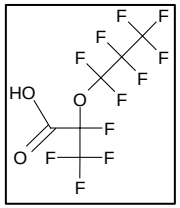
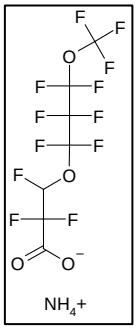
- Column 1:
  - CAS No: the Chemical Abstracts Service (CAS) registry number
  - FCM substance No: the unique identification number of the substance
  - Ref. No: the EEC packaging material reference number
- Column 2:
  - (Substance Name): the chemical name

  - (Subgroup, group): Subgroup and group (Figure 1 and 2) containing the substance. (Sub)groups are not indicated for monomers. Attribution to a (sub)group has been performed by expert judgement of the authors.
- Column 3: (Use as additive or polymer production aid (PPA) (yes/no)): an indication if the substance can be used as additive or polymer production aid (yes) or if the substance is not authorised to be used as additive or polymer production aid (no). If the substance is only authorised as PPA it is indicated (yes) and in the specifications the use is restricted to PPA.
- Column 4: (Use as monomer or other starting substance (yes/no)): an indication if the substance can be used as monomer or other starting substance (yes) or if the substance is not authorised to be used as monomer or other starting substance (no).
- Column 5: polymer (yes/no).
- Column 6: (SML [mg/kg]): the Specific Migration Limit, i.e. the maximum permitted amount of a given substance released from a material or article into food or food simulants. ND is not determined.
- Column 7: (Restrictions and specifications): contains other restrictions than the specific migration limit specifically mentioned and it contains specifications related to the substance. In case detailed specifications are set out a reference to Table 4 is included.
- Column 8: (Migration and toxicity) as available in the indicated dossier.
- Column 9: (Source) legislation that mentions the substance. The following abbreviations have been used to indicate legislation of countries:
  - EU: European Union
  - NL: the Netherlands
  - DE: Germany
  - BE: Belgium
  - ES: Spain
  - IT: Italy
  - Fr: France

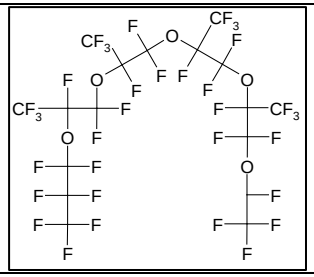
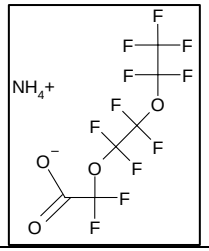
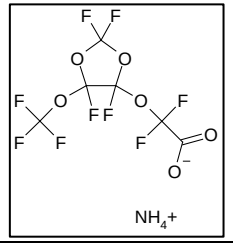
Table 2: Overview of PFASs allowed in FCMs according to regulations. Note that in some legislations (e.g. EU 10/2011) single substances and monomers are regulated, while others aim to regulate single substances and polymers. Therefore, both the monomers as the polymers constructed from the monomers may be present in the table below.

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
3825-26-1 [468] (71960)	Perfluorooctanoic acid (PFOA), ammonium salt [PFCA, PFAA] 	Yes (PPA: emulsi-fier/dis-persion agent)	No	No	NA	EU: Only to be used in repeated use articles, sintered at high temperatures (maximum addition level 0.5%) NL and DE coatings: as emulsifier only in coatings on kitchen utensils for cooking, baking, roasting etc. ES: Only to be used in repeated use articles, sintered at high temperatures.	Migration (EFSA, 2005) : The calculated worst case migration was 0.017 mg/kg food, (sample thickness 0.6 cm, 6 dm^2/kg food). Specific migration was not determined. Toxicity: see overview Section 8.1.1	EU 10/2011 [plastics] NL X.11.2.1.1.b. 2 and X.11.2.2.b [coatings] DE LI.2.1.4.1 [coatings] ES Parte B lista 1 [coatings]
329238-24-6 [854] (71943)	Perfluoro acetic acid, α-substituted with the copolymer of perfluoro-1,2-propylene glycol and perfluoro-1,1-ethylene glycol, terminated with chlorohexa-fluoropropyloxy groups E.g.  where n ranges from 1 to 4 and m from 0 to 2. [PFECA, PFAE]	Yes (PPA: emulsi-fier and as a disper-sing agent)	No	No	NA	EU, NL: Only to be used in concentrations up to 0.5 % w/w in the polymerisation of fluoropolymers that are processed at temperatures at or above 340 °C and are intended for use in repeated use articles.	Migration (EFSA, 2010): The polymer sheet was extracted using solvents and neither the substance nor any substance derived impurities were detected at the detection limit of 11 µg/kg of solvent. It is assumed that the migration of the substance during the service life will not be greater than about 1 µg/kg food. Toxicity (EFSA, 2010): not genotoxic.	EU 10/2011 [plastics]

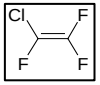
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
51798-33-5 [860] (71980)	Perfluoro[2-(poly(n-propoxy))propanoic acid] [PFECA, PFAE] 	Yes (PPA during emul-sion polymeri sation of fluoro-poly-mers	No	No	NA	EU: Only to be used in the polymerisation of fluoropolymers that are processed at temperatures at or above 265 °C and are intended for use in repeated use articles NL: - Only for manufacturing of PTFE for use as top layer for cooking, baking and roasting utensils, used at max 230 °C, and - Impurity HFPO <5 µg/kg in the mixture; (hexafluoropropylene oxide, CAS 428-59-1), and - QMA (T) = 0.01 mg/6 dm^2 for sum of perfluoropropoxy-1,2,2,2-tetrafluoroethane and poly-perfluoropropoxy-1,2,2,2-tetrafluoroethane (Mw<1000 Da).	The substance is manufactured from the monomer hexafluoropropylene oxide (HFPO) and has a molecular weight distribution between 600 and 12000 Da. The highest fraction of oligomers with molecular weights below 1000 Da is 13.5%.The substance completely decarboxylates during the production process to form a defined heat stable product. Decarboxylation starts at temperatures above 150°C (EFSA, 2009). Migration (EFSA, 2009): calculation migration from 2 to 26 µg/kg in food for the fraction below 1000 Da Toxicity of HFPO (=starting substance of polymer) (EFSA, 2009): not genotoxic.	EU 10/2011 [plastics] NL X.11.2.1.1.b.2 [coatings]
13252-13-6 [861] (71990)	Perfluoro[2-(n-propoxy)propanoic acid] or 2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)-propanoic acid Code names: HFPO-DA; GenX; FRD-	Yes	No	No	NA	EU: Only to be used in the polymerisation of fluoropolymers that are processed at temperatures at or above 265 °C and are intended for use in repeated use articles. NL: - Only for manufacturing of	Migration (EFSA, 2009): The substance completely decarboxylates under the high temperatures of the production process to a heat stable and volatile product (E1). The residual content of this decarboxylation product was determined and the worst case migration was	EU 10/2011 [plastics] NL X.11.2.1.1.b.2 [coatings]

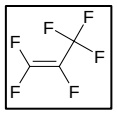
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
	903 [PFECA, PFAE] 					PTFE for use as top layer for cooking, baking and roasting utensils, used at max 230 °C and - QMA (T) = 0.01 mg/6 dm^2, for sum of perfluoropropoxy-1,2,2,2-tetrafluoroethane and poly-perfluoropropoxy-1,2,2,2-tetrafluoroethane with MW<1000 Da	calculated using the total mass transfer assumption. For a number of typical polymer applications, these calculation indicated migration to be **5 µg/kg** in food or less occurring with the first use and followed by zero exposure over the remaining service life of the food contact articles. Toxicity: See Section 8.1.3.	
958445-44-8 [896] (71958) NB: CAS No. of the acid is 919005-14-4	3H-perfluoro-3-[(3-methoxy-propoxy)propanoic acid], ammonium salt or ADONA [PFECA, PFAE] 	Yes (emulsi-fier)	No	No	NA	EU: Only to be used in the polymerisation of fluoropolymers when: *processed at temperatures higher than 280 °C for at least 10 minutes, *processed at temperatures higher than 190 °C up to 30 % w/w for use in blends with polyoxymethylene polymers and intended for repeated use articles.	Migration (EFSA, 2011a): Residual concentration of the substance was not detectable in final sintered perfluoropolymer materials at a detection limit of 0.02 mg/kg. Worst case migration was calculated to be **below 1 µg/kg** food when considering total mass transfer on first use. For an unsintered fluoropolymer micropowder a residual concentration of 3.3 mg/kg of the substance was determined. Such an unsintered material would only be used for polyoxymethylene (POM) blends at a maximum use level of 30%. Worst case	EU 10/2011 [plastics]

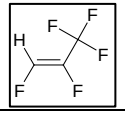
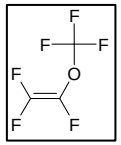
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/ kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
							migration was calculated to be **31 µg/kg food** when assuming that the entire residual amount present in the micropowder before being blended into POM polymer transfers during the first application into food. However, in view of the volatility of the substance and when considering compounding and extrusion conditions of approx. 200°C, effective removal of the substance and negligible migration potential is expected from such a POM polymer blend. Toxicity: See Section 8.1.4	
37486-69-4 [903] -	2H-perfluoro-[(5,8,11,14-tetramethyl)-tetraetheleneglycol ethyl propyl ether] [PFAE]	Yes (PPA)	No	No	NA	EU: Only to be used as a polymer production aid in the polymerisation of fluoropolymers intended for: (a) repeated and single use materials and articles when sintered or processed (non-sintered) at temperatures at or above 360 °C for at least 10 minutes or at higher temperatures for equivalent shorter times; (b) repeated use materials and articles when processed (non-sintered) at temperatures from	Migration (EFSA, 2012): Non-sintered > 300°C: 13 µg/kg food (based on 100% release from end product); 1 µg/kg food (based on modelling with conservative assumptions) At >360 °C in sintered or processed (non-sintered) articles: not detected. Toxicity (EFSA, 2012): not genotoxic.	EU 10/2011 [plastics]

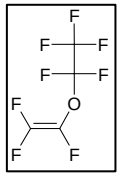
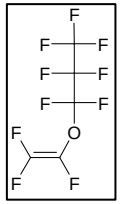
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
						300 °C and up to 360 °C for at least 10 minutes.		
908020-52-0 [926] (71955)	Perfluoro[(2-ethyloxy-ethoxy)acetic acid], ammonium salt [PFECA, PFAE] 	Yes (emulsi-fier)	No	No	NA	EU: Only to be used in the polymerisation of fluoropolymers that are processed at temperatures higher than 300 °C for at least 10 minutes.	Migration (EFSA, 2011b): Using 3% acetic acid, 10% ethanol and 95% ethanol as food simulants (4 hours at reflux temperature) migration of the substance was not detectable at detection limits of 2 ppb in the aqueous food simulants and 6.5 ppb in 95% ethanol. Toxicity (EFSA, 2011b): not genotoxic.	EU 10/2011 [plastics]
1190931-27-1 [1045]	Perfluoro[acetic acid, 2-[(5-methoxy-1,3-dioxolan-4-yl)oxy]], ammonium salt [PFECA, PFAE] 	Yes (emulsi-fier/dis-persion agent)	No	No	NA	EU: Only to be used as a polymer production aid during the manufacture of fluoropolymers under high temperature conditions of at least 370 °C.	Migration (EFSA, 2014): Residual content of the substance in a polytetrafluoroethylene (PTFE) film with a thickness of 60 µm manufactured at a low processing temperature (370 °C) and time (2-7 min) was not detectable, at a detection limit of 0.06 µg/g. This corresponds to a maximum migration of **0.4 µg/kg food.**	EU 10/2011 [plastics]

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/ kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
							Two degradation products were not detectable in the polymer at detection limits of 0.045 mg/6 dm² and 0.001 mg/6 dm², respectively, which is equivalent to 45 µg/kg and 1 µg/kg food. Due to the high detection limit for the major decomposition product, a migration test was carried on the same test film and using 3 % acetic acid and olive oil as food simulants at contact conditions of 4.5 hours at 100 °C and 3 hours at 175 °C, respectively. In both cases the decomposition product was not detectable at 0.4 µg/6 dm² corresponding to **0.4 µg/kg food**. Toxicity (EFSA, 2014): not genotoxic.	
75-38-7 [132] (26140)	Vinylidene fluoride or 1,1-difluoroethene monomer of e.g. PVDF 	No	Yes	No	5 (EU) 1 (NL rubber)	FR: NOT on positive list LMS = ND (LD = 0.05 mg/kg) Section B : monomers and other starting substances can be used until 31 December 1998. These substances can no longer be used at present: the modification or deletion of this list is currently under study.	Migration data (EC, 2005): not available Toxicity data (EC, 2005): List 3; SML= 5 mg/kg of food. Many inhalation studies, 1-year oral rat study, carcinogenicity studies by inhalation in mice and rats negative, mutagenicity	EU 10/2011 [plastics] NL III.4.2.1 [rubber] NL X.3.a [coatings] ESCO_FR_ Arrêté du

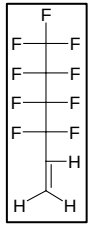
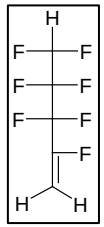
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA#$	Monomer or other starting substance$	Poly-mer$	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
						IT: The restrictions are under revision. Elastomer, monomers for special elastomers.	studies negative, reproduction study negative.	9/11/1994 (FR) [rubber] ESCO_IT_ D.M. 21/3/73 [rubber]
79-38-9 [148] (14650)	Chlorotrifluoroethylene Monomer of e.g. polychlorotrifluoroethylene 	No	Yes	No	ND (EU) 0.05 (NL rubber)	NL rubber: only together with vinylidene fluoride. FR: not on positive list, Qm = 5 mg/kg. Section B : monomers and other starting substances can be used until 31 December 1998. These substances can no longer be used at present: the modification or deletion of this list is currently under study.	SCF (SCF, 2001b); QMA = 0.5 mg/6 dm². The residual content QM = 0.05 mg/6 dm^2, assuming 100% migration, is raised 10-fold as it has been demonstrated that more than 95% of migrants appear in the headspace because of the extreme volatility of the additive. Toxicity (SCF, 2001): Not genotoxic.	EU 10/2011 [plastics] NL III.4.2.1 [rubber] ESCO_FR_ Arrêté du 9/11/1994 (FR) [rubber]
116-14-3 [281] (25120)	Tetrafluoroethylene Monomer of e.g. PTFE 	No	Yes	No	0.05 (EU, NL rubber)	NL rubber: only together with vinylidene fluoride and hexafluoropropene; molecular weight of the elastomer at least 100,000 NL and DE coatings: copolymer with perfluoroalkyl (C_1-C_3) vinyl ether and/or (max 5% of) hexafluoropropene; only for coating of kitchen utensils for cooking, baking, roasting etc. The melting viscosity of these copolymers at 372 °C, must be	Migration data (EC, 2005): not available Toxicity data (EC, 2005): Not genotoxic.	EU 10/2011 [plastics] NL III.4.2.1 [rubber] NL X.11.2.1.1.b +c [coatings] DE LI.2.1.1.2 [coatings] ESCO_ FR_

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
						greater than 10^3 Pa.s; their melting point must be no less than 305 °C. NL coating: copolymers containing hexafluoropropene can be used up to 140 °C, other copolymers up to 230 °C. FR: not on positive list SML = ND (LOD = 0.05 mg/kg). Section B : monomers and other starting substances can be used until 31 December 1998. These substances can no longer be used at present: the modification or deletion of this list is currently under study. IT: Elastomer, monomers for special elastomers. The restrictions are under revision.		Arrêté du 9/11/1994 (FR) [rubber] ESCO_IT_ D.M. 21/3/73 [rubber]
116-15-4 [282] (18430)	Hexafluoropropylene Monomer (with tetra-fluoroethylene) of e.g. FEP 	No	Yes	No	ND (EU, FR) 0.05 (NL rubber)	NL rubber: only together with vinylidene fluoride; molecular weight of the elastomer at least 70,000. NL coatings and DE coatings: see 281. FR: Section A : permitted monomers and starting substances.	SCF (2001a) No migration data available; substance is a gas. Toxicity (SCF, 2001a): Genotoxic substance.	EU 10/2011 [plastics] NL III.4.2.1 [rubber] ESCO _FR_ Arrêté du 9/11/1994 (FR) [rubber]

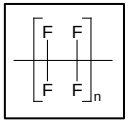
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
						IT: Under revision, Elastomer, monomers for special elastomers The restrictions are under revision.		ESCO_IT_ D.M. 21/3/73 [rubber]
2252-83-7	1-Hydro pentafluoro propene or 1,2,3,3,3-Pentafluoropropene [perfluoroalkyl alkene] 	No	Yes	No	NA	IT: Restrictions are under revision. Elastomer, monomers for special elastomers.	The substance has been evaluated by Italian authority and these evaluations are not publically available.	ESCO_IT_ D.M. 21/3/73 [rubber]
1187-93-5 [391] (22932)	Perfluoromethyl perfluorovinyl ether (PFMVE) 	No	Yes	No	0.05 (EU, ES, NL)	EU: Only to be used in: *anti-stick coatings; *fluoro- and perfluoropolymers intended for repeated use applications where the contact ratio is 1 dm^2 surface in contact with at least 150 kg food. ES: Only to be used for anti-stick coatings. NL: Only for manufacturing of PTFE for use as top layer for cooking, baking and roasting utensils, used at max 230 °C.	Migration (EFSA, 2004): Test sample terpolymer containing 7% PFMVE as comonomer. Migration: <LOD (in 15% ethanol and olive oil) No residual PFMVE was detected at a detection limit of 14 µg/6 dm^2 (is < 14 µg/kg of food) Toxicity (EFSA, 2004): Not genotoxic.	EU 10/2011 [plastics] ES Parte B_list1 [coatings] NL X.11.2.1.1.b [coatings] DE LI.2.1.1.2 [coatings] In NL & DE: listed as perfluoroalkyl (C_1C_3)vinyl-ether

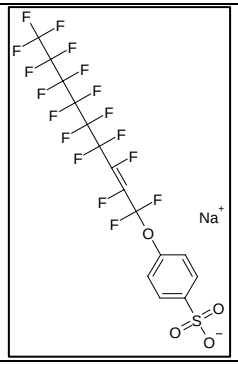
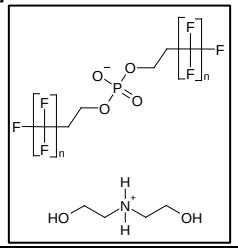
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
10493-43-3	Perfluoroethyl perfluorovinyl ether (PFEVE) 	No	Yes	No	- [11]	NL: Only for manufacturing of PTFE for use as top layer for cooking, baking and roasting utensils, used at max 230 °C.	See PFMVE and PFPVE and footnote	NL X.11.2.1.1.b [coatings] DE LI.2.1.1.2 [coatings] In NL & DE: listed as perfluoroalkyl (C_1C_3)vinyl-ether
1623-05-8 [423] (22937)	Perfluoropropyl perfluorovinyl ether (PFPVE) 	No	Yes	No	0.05 (EU, NL)	NL: Only for manufacturing of PTFE for use as top layer for cooking, baking and roasting utensils, used at max 230 °C.	EU (non-published SDS, 1994): The major application of PFPVE is in dispersions for coatings to obtain houseware cooking utensils, used at high temperatures. Extraction from 100% Teflon*PFA coating with water at 121°C under 2 atm for 7h, was close to the LOD of 0.003 mg/dm^2 (total fluorine). Assuming all fluorine is coming only from PFPVE: 0.05 mg/kg food, but part of the monomer has disappeared by volatilisation. Advice is to set an QM expressed in mg/dm^2.	EU 10/2011 [plastics] NL X.11.2.1.1.b [coatings] DE LI.2.1.1.2 [coatings] In NL & DE: listed as perfluoroalkyl (C_1C_3)vinyl-ether

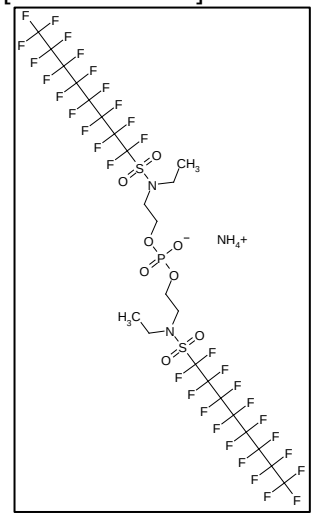
[11] For substances listed in NL, the SMLs of monomers in 10/2011 apply. However, no EU-SML has been derived for PFEVE as only PFMVE and PFPVE are listed in 10/2011, with an SML of 0.05 mg/kg.

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
							PFPVE, tested as a gas, was not mutagenic in the required test systems *in vitro*.	
19430-93-4 [973] (22931)	(Perfluorobutyl)-ethylene [HFO] 	No	Yes	No	NA	EU: Only to be used as a co-monomer up to 0.1 % w/w in the polymerisation of fluoropolymers, sintered at high temperatures.	Migration (EFSA, 2011c): Residual conc. in polymer PTFE powder (not yet sintered): 31 µg/kg Residual conc. in the finished material (sintered): <5 µg/kg (LOD) Estimated migration of oligomers and reaction products will not exceed the range of 1-10 µg/kg food. Toxicity (EFSA, 2011c): Not genotoxic.	EU 10/2011 [plastics]
1547-26-8 [1063]	2,3,3,4,4,5,5-Heptafluoro-1-pentene [HFO-like] 	No[12]	Yes[13]	No	NA	EU: Only to be used together with tetrafluoroethylene and/or ethylene co-monomers to manufacture fluorocopolymers for application as polymer production aid at up to 0.2 % w/w of the food contact material, and when the low-molecular mass fraction below 1 500 Da in the fluorocopolymer does not exceed 30 mg/kg.	Migration (EFSA, 2016): Worst case migration from finished articles: substance: 0.02 µg/kg food sum of the impurities: < 0.02 µg/kg food oligomers < 1,500 Da: 1.5 µg/kg food. The ground fluorocopolymer was extracted by solvent reflux for 4h with isooctane, 95% ethanol, 10% ethanol and 3% acetic acid, resp.	EU 10/2011 [plastics]

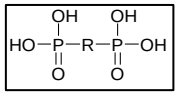
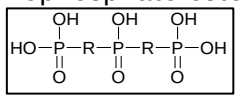
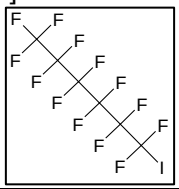
[12] Classification according to EU 10/2011. However, the substance is allowed to be used as a monomer in the manufacture of a polymer production aid. So, it is not a monomer as defined in the Guideline (EU, 2014), that 'monomers […] that are the building blocks of the polymer'.

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
							Toxicity (EFSA, 2016): Not genotoxic.	
9002-84-0 (81160)	Polytetrafluoro-ethylene (PTFE),  [PTFE/FP]	No	No	Yes	NA	NL and DE: melting point at least 320 °C, melting viscosity at 380 °C at least 50 Pa.s. FR: viscosity > 50 Pa.s at 380°C.	Toxicity is determined by monomer, see CAS 116-14-3.	NL X.11.2.1.1.a [coatings] DE LI.2.1.1.1 [coatings] ESCO_FR_Arrêté du 25/11/1992 [sillicones]
25067-11-2	***Copolymer*** *of tetrafluorethene and hexafluoropropene* or Poly(hexafluoro-propene-co-tetrafluoroethene) [copolymers of FP]	No	No	Yes	NA	NL and DE: The copolymer must be compliant with following specifications: - melting point: ≥ 305 °C - melting viscosity at 372°C: ≥ 10^3 Pa.s - hexafluoropropene: max 5% w/w in copolymer	Toxicity is determined by monomers, see CAS 116-14-3, 10493-43-3 and 116-15-4	NL X.11.2.1.1.c [coatings] DE LI.2.1.1.2 [coatings]
e.g. 59536-17-3	Perfluoro-alkenyl-oxy-benzene sulfonic acid [FT] Example:	Yes, emulsi-fying agent	No	No	NA	DE coatings: as emulsifier only in coatings on kitchen utensils for cooking, baking, roasting etc. Max. 0.005 mg/dm^2.	The substance has been evaluated by German authority and these evaluations are not publically available	DE LI.2.1.4.1 [coatings]

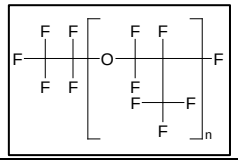
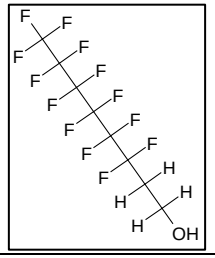
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
								
E.g. (bis): 65530-64-5	Perfluoroalkyl(C_6-C_{16}) phosphates of bis(2-hydroxyethyl)amine or Diethanolamine salts of mono- and bis(1H,1H,2H,2H-perfluoroalkyl(C_8-C_{18}) phosphates ◇ [mono- and di-PAP, FT] 	Yes	No	No	1 (NL)	NL paper/board: no more than 1%.	NL paper&board (1971): Nontox data: extraction: 0.9 mg/5 dm^2 ≈ exposure: 0.9 mg/pers/d Toxicity data: oral LD_{50} (rat &rabbit); 1-2 weeks oral rat 90-day oral rat, & dog ADI: 1 mg/pers/day.	NL X.6.I [coatings] NL II.1.2.2.r [paper/board] BE_bijlage 4.III.3.1.8 [paper/board]

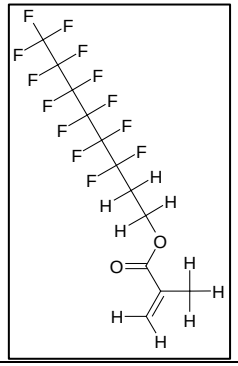
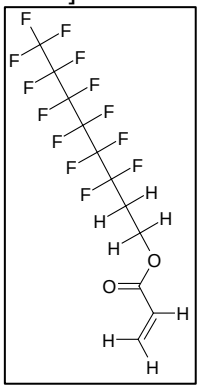
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
30381-98-7 -	Bis[2-[N-ethyl(perfluorooctane) sulfonamido]ethyl] phosphate, ammonium salt or ammonium bis(N-ethyl-2-perfluorooctane sulfonamideethyl) phosphate [SN-di-PAP/FT] 	Yes	No	No	3 (NL)	NL, IT, BE: containing no more than 15% ammonium mono(N-ethyl-2-perfluorooctane sulfonamidoethyl) phosphate as impurity. IT: maximum 0.5% w/w referred to finished and dried product. Not for foods for which the migration test with 20% ethanol is required according to 10/2011. The so treated paper and boards must comply with the requirements in the Title II, point I DM21/3/73. FR: 0.8 g/m² of material, do not put into contact with solid food containing alcohol. BE: Maximum amount on the surface of 8.3 mg/dm^2. Max. amount of fluor: 4.4 mg F/ dm^2 paper	NL: Evaluated in 1971&1973 (paper&board); Migration data: max: 0.24 mg/dm^2 max exposure: 0.14 mg/pers/day. Toxicity data: Available: oral LD_{50} (rat); oral 3 weeks rat oral 90-day study (rat & dog); Conclusion: (P)ADI: 3 mg/person/day.	NL II.1.2.2.r [paper/board] ESCO_IT_ D.M. 21/3/73 am DM 18/06/79 [paper/board] ESCO_FR_ Lettre circulaire du 3 septembre 1975. Publié dans la Circulaire du 29 mai 1978 relative aux MCDA (FR) [paper/board] BE_bijlage 4.III.3.1.7 [paper/board]

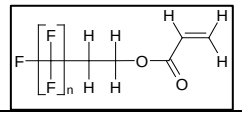
CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/ kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
NA	2-(Perfluorooctyl sulfonyl aminomethyl) ethyl methacrylate, **copolymer** [copolymer of fluorinated (meth)acrylate polymers]	No	No	Yes	1, as total fluor-ine	NL paper/cardboard: copolymers with 2,3-epoxypropyl methacrylate, ethoxyethyl acrylate and methacryloyl methyl-trimethyl ammonium chloride (forming 92265-81-1)	See monomers	NL II.1.2.2.r [paper/board]
NA	Esters of phosphoric acids, perfluoroalkysub-stitutes, ammonium salts, obtained from the reaction of 2,2'-bis[(alpha, omega-perfluoro C_4-C_{20} alkylthio)methyl]-1,3-propandiol, polyphosphoric acid and ammonium hydroxide [unknown]	Yes (polym eric addi-tive)	No	No	NA	IT: Maximum concentration in the finished dried product: 0.44%.	The substance has been evaluated by Italian authority and these evaluations are not publically available	ESCO_IT_ D.M. 21/3/73 am DM 24/09/96 [paper/board]
200013-65-6	Diphosphoric acid, **polymers** with ethoxylated, reduced methyl esters of reduced, polymerised and oxidated tetrafluoroethylene or Phosphoric acid ester of ethoxylated	Yes (polym eric addi-tive)	No	No	0.05 (NL)	DE: Max 1.5 %, based on the dry fibres weight.	Toxicity data (NL 2005): Not genotoxic DE: The substance has been evaluated by German authority and these evaluations are not publically available	NL II.1.2.2.n [paper/board] DE XXXVI.C.IV.2 2 and XXXVI/2.III.D .9 [paper/board]

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
	perfluoropolyetherdiol  monophosphate ester  diphosphate ester R= -O-$(CH_2CH_2O)_p$-CH_2CF_2O-$(CF_2CF_2O)_n$-$(CF_2O)_m$-CF_2-CH_2-$(OCH_2CH_2)_q$-O- [unclear which group]							
69991-62-4	Perfluoropolyether dicarboxylic acid [similar to PFECA, PFAE]	Yes	No	No	NA	DE paper/board: Max 0.5 %, based on the dry fibres weight. The correspondingly treated papers may not come into contact with aqueous and alcoholic foodstuff.	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.24 [paper/board]
355-43-1	1,1,1,2,2,3,3,4,4,5,5,6,6- Tridecafluoro-6-iodohexane [PFAI] 	No[14]	Yes[13]	No	NA	DE paper/board: 2-Propen-1-ol, reaction products with 1,1,1,2,2,3,3,4,4,5,5,6,6-tridecafluoro-6-iodohexane, de-hydroiodinated, reaction products with epichlorohydrin and triethylenetetramine with a fluorine content of 54 %, max. 0.5 %, based on the dry fibres weight.	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.26 [paper/board]

[13] Allowed as a monomer in a (co)polymer that can be used as starting substance or polymeric additive.

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
25038-02-2	Poly(hexafluoro-propylene oxide) [PFPE] 	No	No	Yes	NA	DE paper/board: polymer with 3-N-methylaminopropylamine, N,N-dimethyl dipropylene triamine and poly(hexamethylene diisocyanate), with a fluorine content of 59.1 %, max. 4 mg/dm^2	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.32 [paper/board]
647-42-7	3,3,4,4,5,5,6,6,7,7,8,8,8-Tridecafluoro-1-octanol [FT] 	No	Yes	No	NA	DE paper/board: Reaction product of hexamethylene-1,6-diisocyanate (homopolymer), converted with 3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluoro-1-octanol with a fluorine content of 48 %, max. 0.16 %, based on the dry fibres weight.	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.33 [paper/board]
e.g. 2144-53-8	3,3,4,4,5,5,6,6,7,7,8,8,8-Tridecafluorooctyl methacrylate, acetate and/or malate For single (methacrylate and acrylate) see CAS 17527-29-6) [monomer of (meth)acrylate polymers]	No	Yes	No	NA	DE paper/board: Copolymer with 2-diethylaminoethylmethacrylate, 2,2'-ethylendioxydiethyldimethacrylate, 2-hydroxyethylmethacrylate (CAS 863408-20-2) max. 1.2 %, based on the dry fibres weight.	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.25 and XXXVI/2.IIII.D.10 [paper/board]

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA[#$]	Monomer or other starting substance[$]	Poly-mer[$]	SML [mg/ kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
								
e.g. 17527-29-6	3,3,4,4,5,5,6,6,7,7,8,8,8-Tridecafluorooctyl acrylate (28, 29 and 39), or methacrylate (30, 31, 35 and 38), acetate or sodium salt [monomer of (meth)acrylate polymers] 	No	Yes	No	NA	DE paper/board: 28: Copolymer with 2-hydroxyethyl acrylate, polyethylene glycol monoacrylate and polyethylene glycol diacrylate with fluorine content of 35.4 %, max. 0.4 %, based on dry fibres weight. 29 and D.11: Copolymer with methacrylic acid, 2-hydroxyethylmethacrylate, and polyethylene glycol mono-acrylate, with a fluorine content of 45.1 %, max. 0.8 %, based on dry fibres weight 30: Copolymer with methacrylic acid, 2-diethylaminoethyl-methacrylate and acrylic acid, with a fluorine content of 45.1 %, max. 0.6 %, based on dry fibres weight.	The substance has been evaluated by German authority and these evaluations are not publically available	DE XXXVI.C.IV.28 and 29, 30, 31, 35, 38 and 39 and XXXVI/2.III. D.11 [paper/board]

CAS No. [FCM No.] (Ref No.)	Substance name and structure [subgroup, group]*	Additive or PPA#$	Monomer or other starting substance$	Poly-mer$	SML [mg/kg]	Restrictions and specifications	Migration and toxicity, as available in the indicated dossier	Source [type of FCM]
						31: Copolymer with methacrylic acid and 2-dimethylaminoethyl methacrylate, with a fluorine content of 44.8 %, max. 0.6 % based on dry fibres weight 35. Copolymer with 2-dimethyl-aminoethyl methacrylate, with a fluorine content of 45 %, max. 4 mg/dm^2 (CAS 479029-28-2) 38. Copolymer with 2-hydroxyethylmethacrylate, methacrylic acid, itaconic acid, max. 24 mg/dm^2. 39. Copolymer with 2-hydroxyethylmethacrylate, vinyl pyrrolidone, acrylic acid, with a fluorine content of 41.9 %, max. 1.0 %, based on dry fibres weight		
65605-70-1	Perfluoroalkyl acrylate [side-chain fluorinated acrylatepolymer] Example of monomer: 	No	No	Yes	NA	Note: this group also contains the PFAS in the row above.	The substance has been evaluated by Italian authority and these evaluations are not publically available	ESCO_IT_ D.M. 21/3/73 am D n.411, 1/12/2000 [paper/board]

* Subgroup (if available) and group according to the arrangement in Figure 1 and Figure 2. Groups are not indicated for monomers. Attribution to a group has been performed by expert judgement of the authors.
# PPA = Polymer Production Aid.
$ as indicated by EFSA 10/2011, or by expert judgement of the authors.
◇ Forbidden in food contact paper in USA since 2016: https://www.gpo.gov/fdsys/pkg/FR-2016-01-04/html/2015-33026.htmr.

6 Actual presence of PFASs in FCMs

Information on the actual presence of PFASs in FCMs was investigated by a search in the public literature on experimentally determined PFAS concentrations. The data were collated and analysed and graphically presented.

6.1 Literature search

Between 19 July and 13 August 2018, the public literature was consulted for the availability of concentration data of per- and polyfluorinated compounds in food contact materials. Literature was searched using Google Scholar. Various search terms were used, such as, but not limited to: ‘perfluoroalkyl and polyfluoroalkyl substances’, ‘food contact material’, ‘total organic fluorine’, ‘roasting bag’, ‘detection’, ‘PFAS, ‘non-stick product’, ‘PFOA’, ‘consumer product’, ‘perfluoroalkyl acids’, ‘cork’, and ‘tin-plate’. Articles illustrating concentration data of products dating from before 2009 were not included in the database, as regulation and use of PFASs has changed over the last decade, and these measurements were not considered in concordance with the current legislation on PFASs in food contact materials anymore.

The literature search resulted in 22 publications, among which publications from public health institutions, universities, and non-governmental organizations. The origin of the sampled products varied largely, but samples mainly came from the U.S, China, and countries in Europe, North Africa, and South-East Asia. Most publications focused on food contact paper, with the exception being non-stick silicone baking ware (Blom and Hanssen, 2015; Borg and Ivarsson, 2017), non-stick pans and irons (Herzke *et al.*, 2012), and roasting bags (Surma *et al.*, 2015). A considerable number of publications aimed (solely) to detect PFASs in popcorn bags (Blom and Hanssen, 2015; Borg and Ivarsson, 2017; Dolman and Pelzing, 2011; Liu *et al.*, 2015; Martinez-Moral and Tena, 2012; Moreta and Tena, 2013; Poothong *et al.*, 2012; Ritter *et al.*, 2017; Robel *et al.*, 2017; Shoeib *et al.*, 2016; Trier *et al.*, 2011; Yuan *et al.*, 2016; Zabaleta *et al.*, 2017; Zafeiraki *et al.*, 2014). Furthermore, wrappers for fast food and street food were often sampled (Dolman and Pelzing, 2011; Liu *et al.*, 2015; Poothong *et al.*, 2012; Ritter *et al.*, 2017; Shoeib *et al.*, 2016; Surma *et al.*, 2015; Trier *et al.*, 2011; Yuan *et al.*, 2016; Zabaleta *et al.*, 2016; Zafeiraki *et al.*, 2014).

Various classes of substances were included for detection in the analyses: perfluoroalkyl carboxylic acids, perfluoroalkane sulfonic acids, perfluoroalkyl phosphonic acids, perfluoroalkyl phosphinic acids, per- and polyfluoroether sulfonic acids, fluorotelomer alcohols, fluorotelomer acrylates, fluorotelomer sulfonates, , and polyfluoroalkyl phosphoric acid mono- and diesters (mono- and di-PAPs). Historically, the long-chain substances were of interest because of their hazard profile. However, the focus of the publications included in the database was also on the short-chain compounds, as it was suspected that the short-chain substances replaced the long-chain substances and hence the exposure to these substances could potentially be relatively high. By including

polyfluoroalkyl phosphoric acid mono- and diesters and fluorotelomer alcohols, also indirect exposure to perfluorocarboxylic acids was taken into account. It is known that these classes of substances are precursors of perfluorocarboxylic acids, and that degradation of PAP-containing materials contributes to the exposure of perfluorocarboxylic acids, as well as the migration of fluorotelomer substances (Trier *et al.*, 2017).

6.2 The publications' analytical methods and reporting of results

All publications aimed to detect per- and polyfluorinated compounds in food contact materials, but their methods and reporting of results varied considerably.
The most used detection method for individual PFASs was LC-MS/MS, followed by GC-MS (Blom and Hanssen, 2015; Dolman and Pelzing, 2011; Herzke *et al.*, 2012; Kotthoff *et al.*, 2015; Liu *et al.*, 2015; Moreta and Tena, 2013; Poothong *et al.*, 2012; Shoeib *et al.*, 2016; Surma *et al.*, 2015; Trier *et al.*, 2011; Vestergren *et al.*, 2015; Xu *et al.*, 2013; Yuan *et al.*, 2016; Zabaleta *et al.*, 2017; Zafeiraki *et al.*, 2014). The remaining publications measured total organic fluorine (TOF) (Borg and Ivarsson, 2017; Ritter *et al.*, 2017; Schaider *et al.*, 2017) or both (several) individual PFASs (by LC-MS/MS) and TOF (Borg and Ivarsson, 2017; Robel *et al.*, 2017). Most substances are best detected using LC-MS/MS. As the short-chain fluorotelomer alcohols and polyfluoroalkyl phosphoric acid esters are volatile compounds, they are usually detected using GC-MS.
The detection of PFASs in these publications was reported in a quantitative or qualitative manner. It is common to report the results of LC-MS/MS and GC-MS quantitatively. The FCM samples were either shredded or cut into pieces and the measured PFAS content expressed as µg/m^2 or µg/kg material. Furthermore, the raw data were not always presented, but rather summarized data given as a minimum, mean or median, and/or maximum value were found for the different sample classes. With regard to the TOF analysis, the results were reported in a qualitative manner, so either elemental fluor being present or absent, the number of samples containing fluor, or the percentage of samples containing fluor. Otherwise, the TOF results were reported as nmol fluor/cm^2.

6.3 Overview of PFASs experimentally determined in FCMs

An overview of the experimentally determined PFAS concentrations in FCMs is presented in Figure 3 to Figure 6.
It should be noted that data analysis is complicated because of the many different types of samples, using various detection methods, and reporting results in various units. Combining of results was performed, but resulted in some loss of information with regard to the origin of the samples (e.g. food contact material, food type packaged, processed or non-processed food, meant for hot or cold types of food).
In the figures all concentrations in µg/m^2 and µg/kg are included. No distinction is made between values of a single measurement and summary data, i.e. a point in the figure can represent one single sample or the minimum, mean, median or maximum of several samples.
PFASs were analysed in various products. These products were assigned to the following product categories:

- Baking aid (silicone); this group contains silicone baking moulds, ware and covers; and reusable baking liner.
- Baking paper or cup; this group contains cupcake, muffin and baking cups; and baking paper.
- Beverage cup or plate; this group comprises hot and cool beverage containers and cups; (cardboard) coffee cups; noodle cups; instant rice porridge cups; ice cream cups; paper cups, plates and tableware; and (cinema) popcorn boxes and buckets.
- Food container (cardboard); it was attempted to gather all products made of cardboard or thick paper into this category. (Thin) paper products are included in the category food wrapper or bag (paper). The food container (cardboard) category contains products described as fast food packaging, container and box; burger, lasagne and rice cardboard; cardboard fast food and pasta; dessert container; French fries box; fries holder; and pizza box.
- Food wrapper or bag (paper); it was attempted to gather all products made of (thin) paper into this category. Cardboard or thick paper food contact products are included in the category food container (cardboard). The food wrapper or bag (paper) category contains paper breakfast bag; donut bag and wrapper; (fast) food bag and wrapper; paper bag and wrapper, sandwich paper.
- Microwave bag; consisting mainly of microwave popcorn bags and popcorn bags without mentioning microwave, but also two samples of browning microwave bags.
- Miscellaneous food contact paper; products in this category were not described in sufficient detail to include them in the food container (cardboard) or food wrapper or bag (paper) categories. Products designated as food contact paper and paper box, without further specifications, were included in this category.
- Non-stick pans and pots; containing samples solely taken by Herzke at al. (Herzke *et al.*, 2012) and described as non-stick ware.
- Other; this group contains products which were not clearly described or did not fit one of the above categories, such as: foil paper wrapper; aluminium foil bag and wrapper; polystyrene coffee cup; polyethersulfone roasting bag; and milk bottle.

PFASs were sampled in various countries, which were included in the overview. In the study of Herzke *et al.* (Herzke *et al.*, 2012) and the Nordic Council of Ministers (Trier *et al.*, 2017) the FCMs were obtained in Norway/Sweden and Scandinavia, respectively. Other studies reported that samples were taken from one specific Scandinavian country. To avoid multiple combinations of Scandinavian countries in the resulting plots all data from Denmark, Norway and Sweden were gathered under Scandinavia.

Table 3: Abbreviations used in Figure 3 to Figure 6. General descriptions of the (sub)groups can be found in Section 2.2 and Figure 1.

Abbreviation	Substance	Subgroup	Group
FTA 6:2	6:2 fluorotelomer acrylate	Fluorotelomer acrylates (FTAs)	Fluorotelo-mer substances
FTA 8:2	8:2 fluorotelomer acrylate		
FTA 10:2	10:2 fluorotelomer acrylate		
FTOH 4:2	1H,1H,2H,2H,-perfluoro-1-hexanol	Fluorotelomer alcohols (FTOHs)	
FTOH 6:2	1H,1H,2H,2H,-perfluoro-1-octanol		
FTOH 8:2	1H,1H,2H,2H,-perfluoro-1-decanol		
FTOH 10:2	1H,1H,2H,2H,-perfluoro-1-dodecanol		
FTOH 12:2	1H,1H,2H,2H,-perfluoro-1-tetradecanol		
FTOH 14:2	1H,1H,2H,2H,-perfluoro-1-hexadecanol		
FTOH 16:2	1H,1H,2H,2H,-perfluoro-1-octadecanol		
FTOH 18:2	1H,1H,2H,2H,-perfluoro-1-eicosanol		
FTS 4:2	4:2 fluorotelomer sulfonate	Fluorotelomer sulfonate (anions) (FTSs)	
FTS 6:2	6:2 fluorotelomer sulfonate		
FTS 8:2	8:2 fluorotelomer sulfonate		
FTCA 5:3	5:3 fluorotelomer saturated acid	Fluorotelomer (saturated) carboxylic acids (FTCAs)	
FTCA 6:2	6:2 fluorotelomer saturated acid		
FTCA 7:3	7:3 fluorotelomer saturated acid		
FTCA 8:2	8:2 fluorotelomer saturated acid		
FTUCA 6:2	6:2 fluorotelomer unsaturated acid	Fluorotelomer unsaturated carboxylic acids (FTUCAs)	
FTUCA 8:2	8:2 fluorotelomer unsaturated acid		
PFOSA	perfluorooctane sulfonamide	Perfluoroalkane sulfonamides (FASAs)	PASF and PASF-based derivatives
PFBS	perfluorobutane sulfonic acid	Perfluorosulfinic acids (PFSAs)	Perfluoro-alkyl acids (PFAAs)
PFPeS	perfluoropentane sulfonic acid		
PFHxS	perfluorohexane sulfonic acid		
PFHpS	perfluoroheptane sulfonic acid		
PFOS	perfluorooctane sulfonic acid		
PFDS	perfluorodecane sulfonic acid		
PFBA	perfluorobutanoic acid	Perfluorocar-boxylic acid (PFCAs)	
PFPeA	perfluoropentanoic acid		
PFHxA	perfluorohexanoic acid		
PFHpA	perfluoroheptanoic acid		
PFOA	perfluorooctanoic acid		
PFNA	perfluorononanoic acid		
PFDA	perfluorodecanoic acid		
PFUnDA	perfluoroundecanoic acid		
PFDoDA	perfluorododecanoic acid		
PFTriDA	perfluorotridecanoic acid		
PFTeDA	perfluorotetradecanoic acid		
PFPeDA	perfluoropentadecanoic acid		

Abbreviation	Substance	Subgroup	Group
PFHxDA	perfluorohexadecanoic acid		
PFHpDA	perfluoroheptadecanoic acid		
PFODA	perfluorooctadecanoic acid		
PFHxPA	perfluorohexane phosphonic acid	Perfluorophos-phonic acids (PFPA)	
PFOPA	perfluorooctane phosphonic acid		
PFDPA	perfluorodecane phosphonic acid		
PFPiA 6:6	6:6 perfluorophosphinate	Perfluoroalkyl-phosphinic acids (PFPIAs)	
PFPiA 6:8	6:8 perfluorophosphinate		
PFPiA 8:6	8:6 perfluorophosphinate		
PFPiA 8:8	8:8 perfluorophosphinate		
PAP 6:2	6:2 perfluoroalkyl phosphate	Polyfluoroalkyl-phosphoric acid esters / Polyfluoroalkyl-phosphates / fluorotelomer phosphates (PAPs)	Fluorotelomer (FT) substances
PAP 8:2	8:2 perfluoroalkyl phosphate		
monoPAP 6:2	6:2 monosubstituted polyfluoroalkyl phosphate		
monoPAP 8:2	8:2 monosubstituted polyfluoroalkyl phosphate		
diPAP 6:2	6:2 disubstituted polyfluoroalkyl phosphate		
diPAP 8:2	8:2 disubstituted polyfluoroalkyl phosphate		

Figure 3 to Figure 6 show the concentration (both as µg/m^2 and µg/kg) of PFASs in different types of FCMs. In Figure 6 a subselection of PFASs was reported. These PFASs (PFHpS, PFOS, PFNS, PFDS, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA) are relatively potent, i.e. they have a Relative Potency Factor (RPF) larger than “one” compared to PFOA (Zeilmaker *et al.*, 2018). Note that the concentrations refer to concentrations *in* the FCM; rather than in foods that have been in contact with these FCMs.

The PFASs determined in FCMs can be the added functional substance. It is also possible that impurities or degradation products are found, which are referred to as Non-Intentionally Added Substance (NIAS).

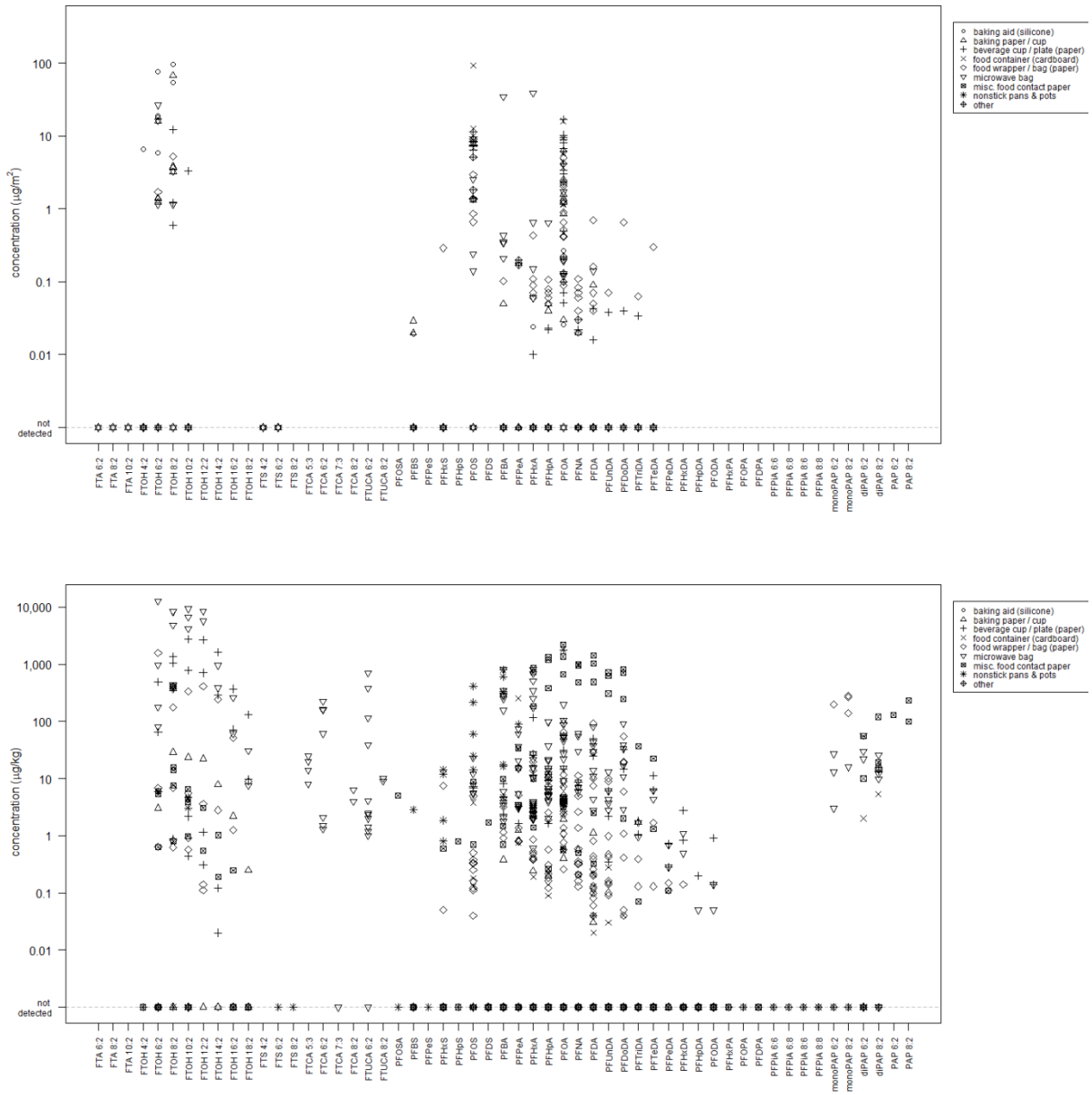


Figure 3: Concentrations in µg/m² (top) and µg/kg (bottom) on logarithmic scale for each individual PFAS. Measurements below the limit of detection or quantification are reported as "not detected". Symbols indicate the various product types as described in the legend. Note that the vertical axis are logarithmic. Concentrations (when above LOD) may vary with up to 6 orders of magnitude.

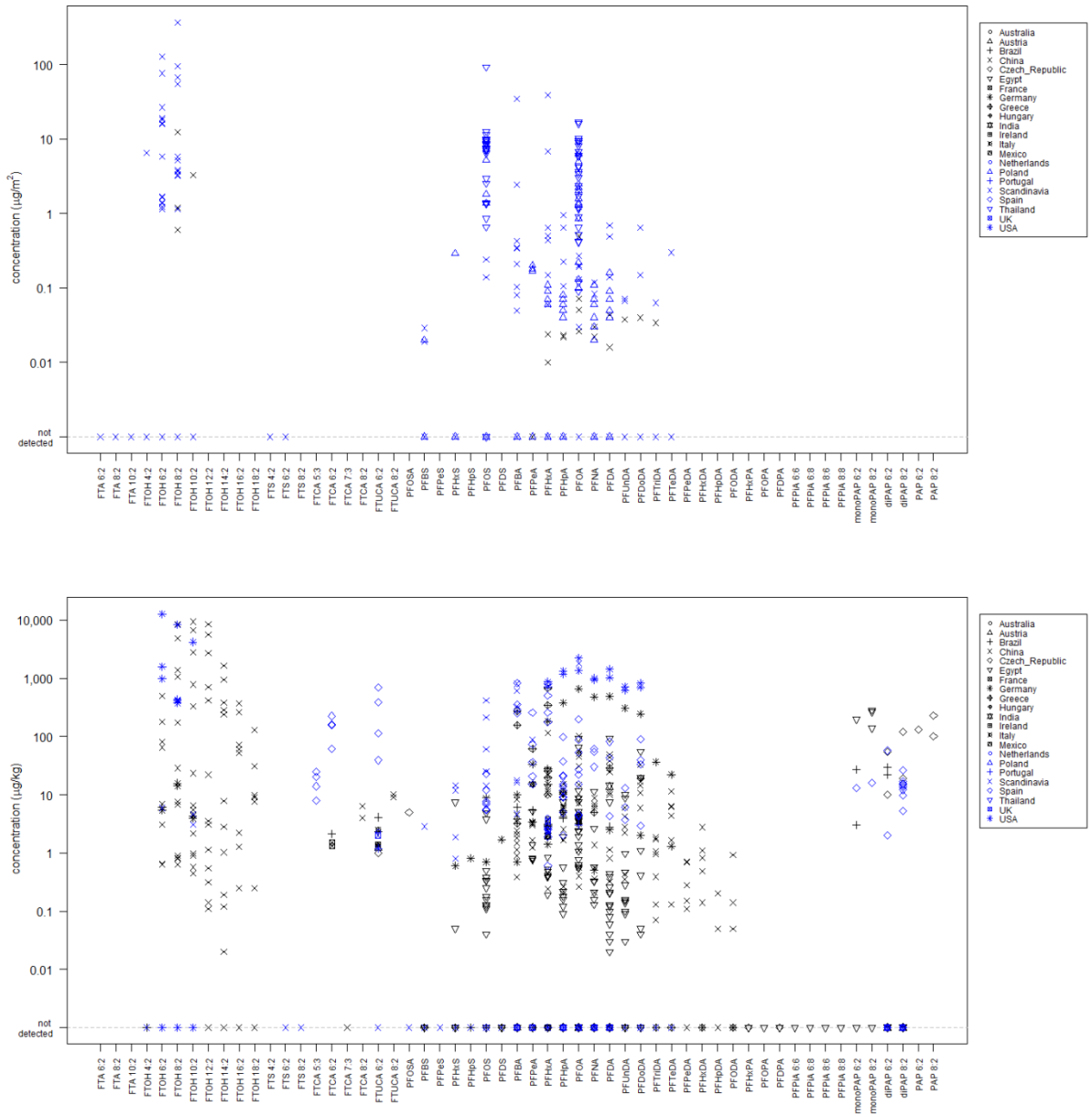


Figure 4: Concentrations in µg/m^2 (top) and µg/kg (bottom) on logarithmic scale for each individual PFAS. Measurements below the limit of detection or quantification are reported as "not detected". Symbols indicate the various countries in which samples were taken.

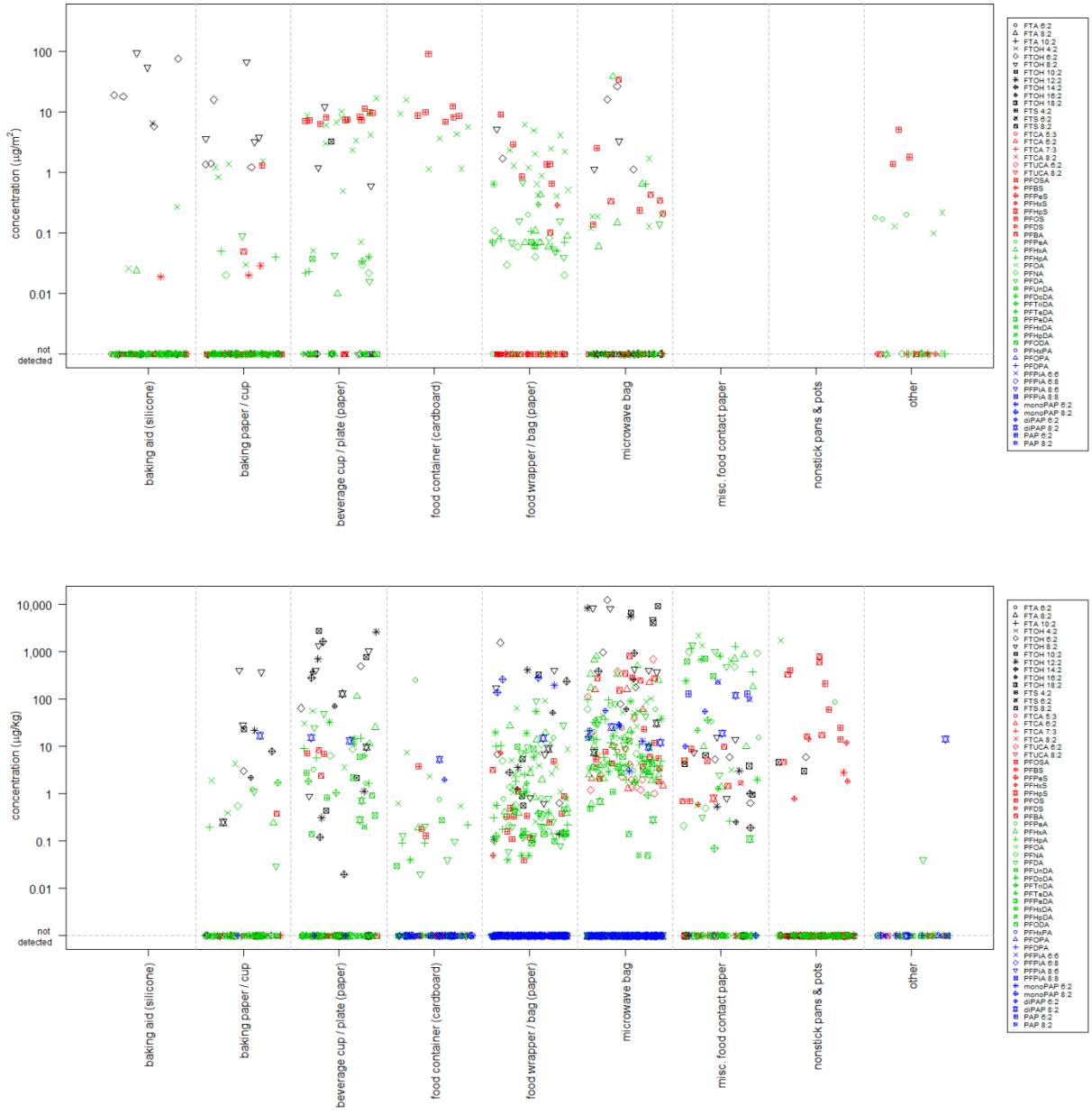


Figure 5: Concentrations in µg/m^2 (top) and µg/kg (bottom) on logarithmic scale measured in various product types. Measurements below the limit of detection or quantification are reported as "not detected". Symbols indicate the various PFAS as described in the legend.

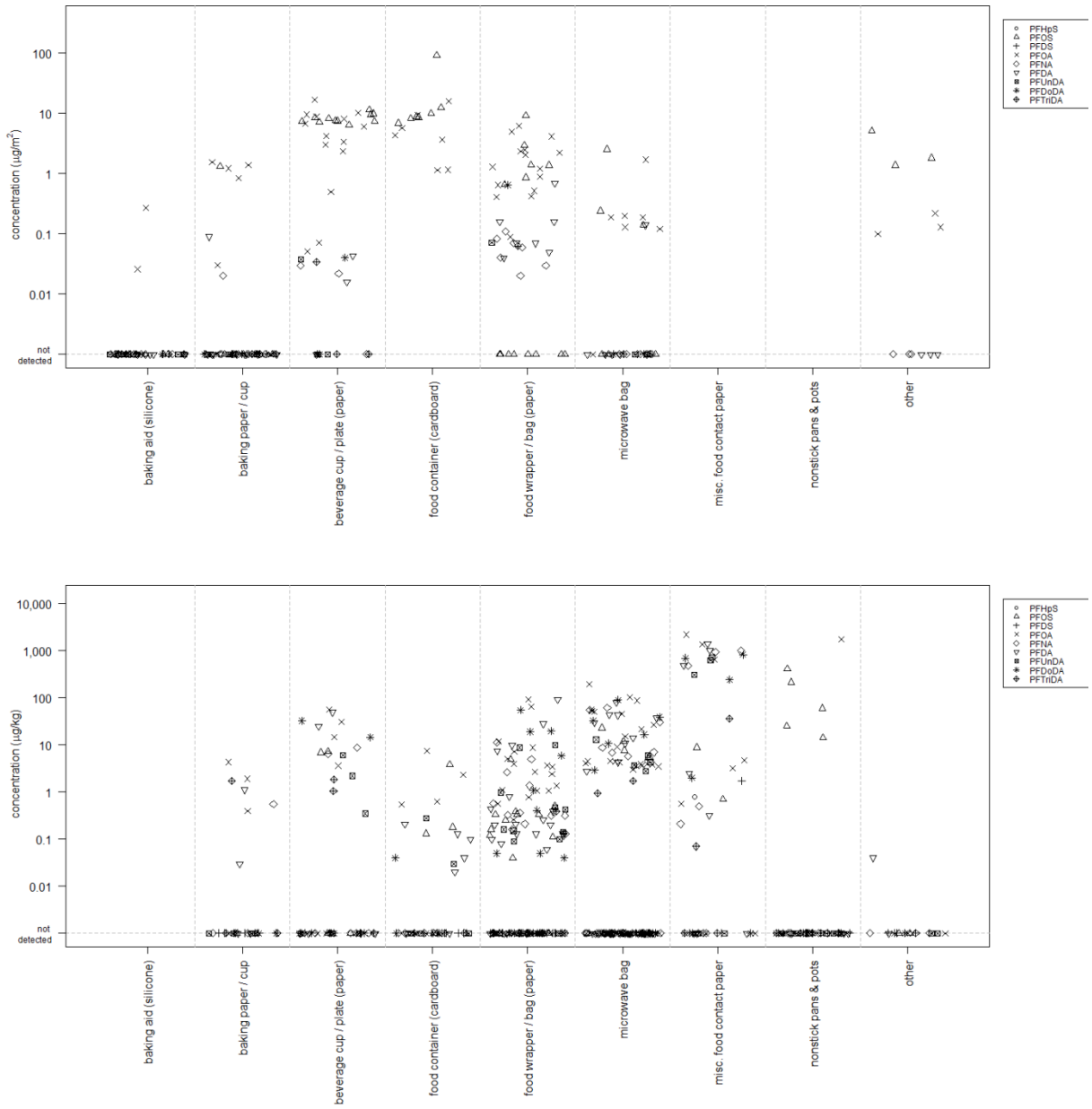


Figure 6: Concentrations in µg/m^2 (top) and µg/kg (bottom) on logarithmic scale measured in various product types. Measurements below the limit of detection or quantification are reported as "not detected". Symbols indicate a sub-selection of PFAS as described in the legend.

7 Experimentally determined migration of PFASs

Migration experiments described in EFSA opinions on plastic and coating generally indicated limited migration. In submitted dossiers for national authorization, migration of PFASs from paper and board was measured. In addition, a limited number of experiments on the migration of PFASs from paper and non-stick coatings in pans and other kitchen appliances, have been published in scientific literature. These papers are described below.

Xu *et al.* (2013) determined the concentrations of seven PFCAs and three PFSAs impurities in two commercially available, technical-grade food contact substances (FCSs): a di-perfluoro-alkyloxy-amino-acid (PAA) and a perfluoroalkyl phosphate surfactant (PAP). FSC is the starting material for application in or on products and can be applied in food contact paper and board. In addition, two paper food packaging samples, one produced with PAA and the other with PAP, were analysed on PFCAs and PFSAs, and the migration of the PFCAs into five food simulants from the two commercial packagings (obtained in the USA) was evaluated.
The FCS PAA and PAP contained concentrations of the seven measured PFCAs (PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA and PFDoDA) ranging between 0.1 and 0.3% in PAA and between 0.01 and 0.05% in PAP. The PAA food packaging sample contained 2.3 g/kg (0.2%) PAA and 0.0001 to 0.0002% of each PFCA. The PAP food packaging sample contained 5.5 g/kg (0.6%) PAP and 0.0001% of each PFCA. In none of the samples PFSAs (PFBS, PFHxS, PFOS) were detected. According to the authors, this is consistent with the synthesis of PAA and PAP.
Migration tests were performed to assess the migration of PFCAs from the two food contact papers into five food simulants (Miglyol oil (MO), MO with Tween 60 [0.5 w%] (TW), MO with soy lecithin [0.5 w%] (SL), ethanol [10%] in water [v%](EtOH) and acetic acid [3%](AA)) at 40°C from 2 to 240 h. In EtOH and AA migration reached its maximum within 24 hours, in MO after approx. 96 h, but in TW and SL the migration does seem to reach equilibrium within 240 h. For each PFCA, the highest migration (after 240 h) was reached in EtOH, followed by AA, and finally the three MO simulants. Migration depends also on the type of paper (PAA vs. PAP paper) and length of the alkyl chain. Highest migration of 100% was measured for PFHxA and PFHpA, from PAA paper using simulant AA. Migration of PFCAs decreased with longer alkyl chains (in PAA paper using AA) from 100% for PFHxA and PFHpA, to ~50% for PFOA and <5% for PFDoDA. In PAP paper, these percentages were approximately a factor 2 lower. In EtOH from PAA paper, migration percentages were between 65 and 100% for PFBA to PFDA, and decreasing to ~5% for PFDoDA. Again, in PAP paper, these percentages were approximately a factor 2 lower. After 240 h, migration to oily (MO, TW, SL) simulants varied between ~5 and ~50%.

Poothong *et al.* (2012) obtained 34 paper food packaging paper products from Thailand. The authors aimed to determine the PFOS and PFOA concentration in these products and their migration from the products. A very rigorous extraction method was applied, also for

assessing the migration. Therefore, the results should not be interpreted as migration information but rather as concentrations present in food packaging paper.

Yuan *et al.* (2016) argue that paper FCMs are often used for fast food in snack bars and take-away food in restaurants. Therefore, the contact with food is generally for short durations, and migration tests for a short time were conducted. The food simulants, water, ethanol–water solutions (v/v, 10:90, 30:70, 50:50) and oil, were preheated to 100 °C. A total of 10 mL of food simulants was transferred to paper bowls and kept for 15 min at room temperature. Only the bottom of each paper bowl had contact with the food simulants (contact area was 36 cm^2) The sum of 15 PFCAs was 0.006 µg/cm^2 in the paper, and the sum of 7 FTOHs was 0.3 µg/cm^2.
Of the 15 PFCAs (C_4-C_{18}), 9 PFCAs (C_4-C_{12}) were detected in water and 10% ethanol, 12 PFCAs (C_4-C_{14}) were detected in 30% ethanol, and all PFCAs were detected in 50% ethanol. None of PFCAs were detected in oil. Migration efficiencies of PFCAs from paper bowls into water (as percentage of the content in the paper packaging) depend on carbon-chain lengths and ranged from 0.005% for PFDoDA to 20% for PFBA. Compared with the results from water, migration efficiencies of PFCAs into ethanol solutions significantly increased with increasing ethanol level: 0.03–20% for 10% ethanol, 0.01–20% for 30% ethanol, and 0.2–30% for 50% ethanol. Again, the ranges reported for the three ethanol concentrations correspond to the highest to lowest carbon chain lengths.
The 6:2, 8:2, 10:2, 12:2, 14:2 and 16:2 FTOHs were detected in food simulants. Although measured, 18:2 FTOH was not detected. Similar to the results for PFCAs, migration efficiencies of FTOHs into water and 10% ethanol simulant were dependent on their carbon-chain lengths, ranging from 0.004% for 16:2 FTOH to 0.2% for 6:2 FTOH. The component ratios of ethanol in food simulants were also an important factor for the migration efficiencies of FTOHs from FCMs, and migration efficiencies increased to 0.009–3% for 30% ethanol and 0.06–10% for 50% ethanol. The migration efficiencies of FTOHs into oil ranged from 0.04% (16:2 FTOH) to 2% (8:2 FTOH), similar to those into 30% ethanol.

Fengler *et al.* (2011) tested the migration of fluorinated telomere alcohols (6:2-, 8:2-, and 10:2-FTOH) from muffin baking paper to butter, dough and Tenax (stimulant for dry and fatty foods) at various temperatures and durations. In dough, high migration of FTOHs up to 2-6 µg/dm^2 is measured. Migration patterns differ between the two tested doughs of which dough 1 is a dry and low-fat version of dough 2. The authors suggest that due to its higher humidity, dough 2 has lower concentrations (at most temperatures and durations, except one) compared to dough 1, because FTOH evaporates. Migration to butter varies between approximately 2-15 µg/dm^2 for 6:2 FTOH, 2-4 µg/dm^2 for 8:2 FTOH and 1-2 µg/dm^2 for 10:2 FTOH. Migration to Tenax ranges from ~25 to ~90 µg/dm^2. In all simulants migration decreases with increasing FTOH chain length. Furthermore, the dough and butter experiments indicate production of FTOH from precursor compounds.
Choi *et al.* (2018) purchased 312 products (139 frying pans, 132 baking utensils, 10 grill pans, 10 pots, 10 electric rice cooker pots and 11

non-stick baking papers) across Korea in 2014. Four different simulants (n-heptane, 50% ethanol, water and 4% acetic acid) were used for migration tests to detect four PFSAs (PFBS, PFHxS, PFOS, PFDS) and 12 PFCAs (PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTriDA, PFTeDA, PFHxDA, PFODA). For samples from container-type cooking materials, simulant was fully added to the container and the top was closed. For water and 4% acetic acid, the temperature was maintained at 100°C for 30 min, while for 50% ethanol and n-heptane, it was maintained at 70°C for 30 min and 25°C for 1 h, respectively. Other products were contacted with the simulants in a closed system at the same durations and temperatures as described above. No PFASs were detected in the migrations solutions from all 10 grill pans, all 10 pots, all 10 electric rice cooker pots and all 11 non-stick baking papers. Seven PFASs were found in migration solutions from 10 frying pans, including PFOA, PFNA, PFDoDA, PFTrDA, PFTeDA, PFHxDA and PFODA. PFNA was found in the simulants of two baking utensils. Migration (in µg/L simulant) was provided for these products. However, since no initial product concentrations are available, the migration cannot be expressed in percentages. The 50% ethanol simulant resulted in the highest frequency of detection, followed by 4% acetic acid, water, and n-heptane, indicating that e.g. alcoholic beverages or fatty foods are most likely to result in the migration of PFASs.
Additional experiments, applying two migration rounds, indicated that PFASs tend to migrate only upon the first use of perfluorocarbon-coated utensils.

Schlummer *et al.* (2015) measured the gaseous emissions of three PTFE coated and one ceramic coated pans under overheating conditions. In addition the gaseous emissions of one non-stick sandwich toaster and three non-stick waffle irons, heated under normal conditions, were measured. In all cases products were heated during one hour, and 2 or 3 consecutive trails were performed.
PTFE coatings are known to degrade when overheated, and release (gaseous) fluorinated acids. This was confirmed by this study, by measuring PFBA, PFPeA, PFHxA, PFOA, PFNA, PFDA, PFUnDA and PFDoDA after overheating. Although overheating of pans is not unintended use, it can be seen as a reasonable foreseeable situation that occurs incidentally. Results of this part of the experiment are presently not discussed any further.
Only trace emissions of PFCAs were recorded at normal use temperatures with levels in the range from 0.1 to 3.4 ng per item and hour. Most frequent and highest emissions were recorded for PFBA and PFOA. The emission of low amounts of PFBA at normal use conditions may be explained by the presence of PFBA in PTFE coated pans (Herzke *et al.*, 2012). The source of PFBA in PTFE is unclear, since there is no information on the application of PFBA in PTFE production. According to the authors, minor PFBA amounts may be due to a contamination during the supply chain. Or, as the authors speculate, these PFBA levels are caused by a slight thermolysis of PTFE at temperatures from 180 to 230 °C.
Surface temperature of PTFE, intentionally addition of PFOA during production, age of PTFE products, and chain length of emitted PFCA congeners are expected to be main parameters that significantly

influence the emission of PFCAs from PTFE. Thermolytic formation at normal use conditions (below 230 °C) seems to be low.
Another release mechanisms of PFCAs may be the volatilization of intentionally added PFOA. Residual levels of PFOA may remain in PTFE and will decrease further during the use phase of heated and PTFE coated products since repeated heating will volatilize PFOA. Therefore, the authors expect decreasing levels of PFOA with increasing age (or frequency of use) of PTFE products. On the other hand, new PTFE coated products may be even free of PFOA, due to the replacement of PFOA as emulsifier by other (PFAS-)substances such as HFPO-DA.
It should be noted that the experiments (under normal heating conditions) were performed on rather old kitchen appliances. When it is assumed that PFOA (and PFBA or other PFCAs) emissions decrease over time or frequency of use, then higher emissions could be expected from new non-stick kitchen appliances.

In conclusion, based on experimental studies described in literature, migration of a number of PFAS from especially paper and board FCMs occurs. Experimental findings on paper and board show that considerable amounts of PFCAs and FTOHs can migrate into food simulants, particularly when containing alcohol.

8 Overview toxicity and health based guidance values

8.1 Information on the toxicity and health based guidance values

Of all the PFASs, PFOA and PFOS have by far the largest toxicity database. Both substances have been studied extensively in experimental animals and in human epidemiology studies. Below the available evidence is briefly summarized. A further PFASs with a relatively large toxicological database is HFPO-DA (also referred to as GenX). Toxicological data are available on the ammonium salt of HFPO-DA, whereas read-across to these data has been applied for the acid form of HFPO-DA. HFPO-DA is currently used as a replacement of PFOA in the production of Teflon. The HFPO-DA toxicity data are also summarized below. Finally, an overview is provided on the toxicity of ADONA, which is another alternative for PFOA, based on the scientific opinion of EFSA (2011a) on this substance.

8.1.1 *PFOA*

Comprehensive reviews of the toxicological data base are provided by US-EPA (2016a), DWQI (2017b), EFSA (EFSA, 2018; EFSA *et al.*, 2018) and ATSDR (2018). RIVM (Zeilmaker *et al.*, 2016) has evaluated the toxicity of PFOA as part of its risk assessment of the emission of PFOA by the DuPont/Chemours chemical plant in Dordrecht, The Netherlands. As a supplement to this assessment, RIVM (2017) provided a review specifically of the available epidemiological studies for PFOA.

Both PFOA and its ammonium salt APFO have been used in toxicity experiments. Since in aqueous environments both PFOA and APFO lead to the presence of perfluorooctanoate as the dominant chemical species the read across from the salt to the acid is considered valid. Oral animal toxicity studies of short-term, subchronic and chronic duration are available in several species including monkeys, rats, and mice. In addition, developmental toxicity and reproductive toxicity and carcinogenicity were studied in mice and rats. These studies report developmental effects, liver and kidney toxicity, immune effects, and cancer (liver, testicular, and pancreatic). Developmental effects observed in animals include decreased survival, delayed eye opening and reduced ossification, skeletal defects, altered puberty (delayed vaginal opening in females and accelerated puberty in males) and altered mammary gland development.

Human epidemiology studies report associations between PFOA exposure and a number of disorders and diseases. The examined populations were workers at PFOA production plants, a high-exposure community population near a production plant in the United States (the C8 cohort) and members of the general population in the United States, Europe, and Asia. In its review of the epidemiological evidence for PFOA, RIVM (Rijs and Bogers, 2017) selected the following effects as reported in the available epidemiology studies for further evaluation: increased liver enzymes and liver disease, testicular and kidney cancer, pregnancy-induced hypertension and pre-eclampsia, decreased birth weight, increased serum uric acid concentration, ulcerative colitis, changes in blood lipid concentrations, decreased vaccination response and thyroid disorders. The weight of evidence for an association

between PFOA and these effects was concluded to be variable. For some effects inconsistencies were noted, for others the influence of possible confounders could not be ruled and for yet others the biological significance was doubtful. The associations of PFOA exposure and decreased birth weight and increased cholesterol were strongest. It is not certain whether PFOA is the true cause or whether there are other explanations for the observed associations.
(Rijs and Bogers, 2017). Because of the limitations in the available epidemiological evidence, most international organizations have chosen not to use these data for quantitative dose response analysis and risk assessment (for further discussion see below).
In the animal studies with PFOA, changes in liver weight and hepatocellular hypertrophy were the most common effects observed with or without other hepatic indicators of adversity. The liver contains the highest levels of PFOA when analysed after test animal sacrifice. The increase in liver weight and hypertrophy as seen in rodent studies may be associated with activation of the cellular peroxisome proliferator-activated receptor alpha (PPARα)[14]. The PPARα response in the liver is known to be more pronounced in rodents than in humans. But increased liver weight and hypertrophy were also observed in monkeys, which have similar PPARα to humans (Butenhoff *et al.*, 2002). In its 2016 risk assessment for PFOA, RIVM concluded liver effects to represent the most sensitive endpoint for PFOA-toxicity (Zeilmaker *et al.*, 2016). In the derivation of a health-based guidance value (HBGV) for PFOA, RIVM (Zeilmaker *et al.*, 2016) ,rejected a subchronic study in monkeys in which increased liver weight and hypertrophy were seen due to inconsistencies in the serum PFOA-levels that were reported for the different dose groups. However, the study did provide supportive evidence. For the derivation of the HBGV a subchronic study in rats (Perkins *et al.*, 2004) was used, which also included measurement of the serum PFOA levels. In deriving the HBGV for PFOA, the NOAEL from this rat study was divided by a reduced interspecies factor because of the known higher susceptibility to liver effects by PFOA in rodents compared to humans (for the complete derivation see below).
In reproduction toxicity studies in mice reduced fertility and reduced sperm counts were observed (NOAEL 2.5 mg/kg bw/day).
Developmental toxicity in mice and rats showed decreases in pup weight as the most sensitive effect (NOAEL 1 mg/kg bw/day). In addition, several developmental toxicity studies in mice showed delayed mammary gland development in female offspring at very low maternal dose levels. RIVM noted that other hormone-related parameters in these studies showed no effect and concluded that further research on this possible effect is needed. The biological significance of this effect is unknown and was not used for deriving a point of departure.

[14] PPARα is a transcription factor, i.e. a protein that binds to specific DNA-sequences, thereby regulating the production of mRNA. PPAR-alpha is a major regulator of lipid metabolism in the liver. Activation of PPAR-alpha promotes uptake, utilization, and catabolism of fatty acids by upregulation of genes involved in fatty acid transport, fatty acid binding and activation, and peroxisomal and mitochondrial fatty acid β-oxidation. PPAR-alpha is primarily activated through ligand binding. Known synthetic ligands include the fibrate drugs (used in humans to treat hyperlipidemia) and a number of insecticides, herbicides, plasticizers, and organic solvents collectively referred to as peroxisome proliferators. Endogenous ligands include fatty acids such as arachidonic acid as well as other polyunsaturated fatty acids and various fatty acid-derived compounds (Klaunig *et al.*, 2003).

Rat bioassays showed increased incidences of tumours in liver, testes and pancreas. Epidemiological studies in a population living in the vicinity of a PFOA production plant in the USA and in workers of this plant showed an association between PFOA exposure and testicular cancer and kidney cancer. As stated above, IARC concluded the rat bioassay results to represent limited evidence in experimental animals and the positive associations as seen in the epidemiological studies to represent limited evidence for a carcinogenic effect by PFOA in humans. Available information on PFOA genotoxicity and mechanistic information for the induction of the observed tumours indicates a non-genotoxic mode of action (DWQI, 2017b; US-EPA, 2016a; Zeilmaker *et al.*, 2016). For the derivation of a HBGV for PFOA this means that a threshold in its toxic action is assumed and that a HBGV can be derived via application of the appropriate assessment factors to a selected point of departure in the form of an adequate NOAEL or BMDL.

An important property of PFOA for HBGV derivation is its marked potential for bioaccumulation in humans, with an estimated half-life for clearance from human serum as long as 3-4 years. This contrasts with the half-life in experimental animals (monkeys, rats, mice), which is only several weeks at most (11-21 days).

For PFOA, HBGVs have been derived by a number of public organizations dealing with the risk assessment of substances. Table 4 provides an overview of these derivations.

EFSA (2008b) derived an HBGV as an external dose only, i.e. in ng/kg bw/day, by dividing an NOAEL for liver effects derived from rodent studies by several assessment factors. For the difference in toxicokinetics between rodents and humans an extra assessment factor of 2 was applied. ECHA/RAC (2015) used a mouse NOAEL of 1 mg/kg bw/day from a reproduction toxicity study with decreased pup weight as the critical effect. Based on measurement of the serum PFOA levels in this study the NOAEL was concluded to be equivalent with 20 µg/ml in blood serum. By applying a total assessment factor of 25 to this serum concentration an HBGV as internal dose of 0.8 µg/mL was derived. In these two HBGV-derivations the potential of PFOA for bioaccumulation in humans is only taken into account in qualitative way. Following the approach previously developed for polychlorinated dioxins, which are a group of substances with high potential for bioaccumulation in humans, RIVM, ATSDR, US-EPA and DWQI used a quantitative approach for calculation of the long-term human serum concentration equivalent with the mean animal serum concentration measured at the point of departure selected for HBGV-derivation.

Table 4. Overview of Health Based Guidance Values (HBGVs) for PFOA.

Organisation (reference)	**Year**	**Duration**	**HBGV**		**Critical effect**	**Species**
			In serum (ng/mL)	**External dose (ng/kg bw/day)**		
EFSA (2008b)	2008	Chronic	-	1500	Liver	Rat, mouse
ECHA/RAC (2015)	2015	-	800	-	Repro-duction	Mouse
ATSDR (2015)	2015	Subchronic (provisional)	173	20	Liver	Monkey

US-EPA (2016a)	2016	Chronic	142	20	Develop-mental[15]	Mouse
RIVM (Zeilmaker *et al.*, 2016)	2016	Subchronic	710	100	Liver	Rat
		Chronic	89	12.5		
DWQI (2017b)	2017	Chronic	14.5	2	Liver	Mouse
EFSA (EFSA, 2018; EFSA *et al.*, 2018)	2018	Chronic (provisional)	9.4[a]	6 [a] ng/kg bw/wk	Cholesterol	Human
ATSDR (2018)	2018	Subchronic (provisional)	22	3	Develop-mental[16]	Mouse

[a] These values are not supported by RIVM, see text for further details.

Recently, EFSA published an update of its previous evaluation for PFOA and PFOS. In this new opinion EFSA proposes a PFOA HBGV based on benchmark dose modelling of selected health effects as found in epidemiological studies. However, due to the nature of the scientific uncertainties described in this opinion and in the minutes of the expert meeting (EFSA *et al.*, 2018), and the possible application of the forthcoming Scientific Committee guidance on combined exposure to multiple chemicals, the conclusions of this assessment and the derived HBGVs are considered provisional. In finalizing its opinion for PFOA and PFOS, EFSA noted a substantive divergence of opinion with RIVM (Zeilmaker *et al.*, 2016) evaluation for PFOA. According the Article 30 of the EU Food Law (Regulation (EC) No 178/2002) such a divergence is to be formally addressed by EFSA in cooperation with the member state body in question. As a result, a joint document was presented clarifying the contentious scientific issues and identifying the relevant uncertainties in the data. As prescribed by Regulation 178/2002, this document was made public by EFSA (EFSA *et al.*, 2018). The questions that RIVM raised regarding the HBGV primarily pertain to the interpretation and analysis of data from epidemiological studies. RIVM holds the opinion that the scientific articles on which EFSA bases its recommendation do not contain enough data in order to derive a guidance value. Moreover, it is unclear whether, and to what extent, exposure to PFOA (or other perfluorinated compounds) causes the changes identified in the epidemiological studies. Lastly, RIVM raises questions about the data analysis method used to derive the guidance value. Therefore, RIVM remains of the opinion that the value of 12.5 ng/kg bw/day as derived by RIVM (Zeilmaker *et al.*, 2016) based on

[15] US-EPA (2016a) developed a number of candidate RfDs (=HGBVs) based on the following effects: increased liver weight in combination with liver cell necrosis in a subchronic rat study, decreases in pup weight in 2 developmental toxicity studies in mice, reduced pup ossification and accelerated male puberty in a further developmental toxicity study in mice, reduced IgM-response in a subacute study in mice, decreased growth and increased liver and kidney weight in a 2-generation study in rats. The RfD based on reduced pup ossification and accelerated male puberty in the developmental toxicity study by Lau *et al.* (2006) was chosen as the final value.

[16] ATSDR (2018) developed a number of candidate intermediate MRLs (=HBGVs for subchronic exposure) based on the following effects: increased locomotor activity in mice after in utero exposure, bone effects (altered long bone morphology + decreased bone mineral density) and increased locomotor activity in mice after in utero exposure, decreased no. of births in mice after in utero exposure, reduced ossification of proximal phalanges and advanced preputial separation in mice after in utero exposure, reduced weight gain and delayed eye opening in mice after in utero exposure, reduced antibody response and sRBC reponse in mice after subacute exposure, increased severity of chronic inflammation in liver of offspring in mice after in utero exposure, increased liver weight, hepatocellular hypertrophy and cell necrosis in rats after subchronic exposure. The provisional MRL based on bone effects and decreased locomotor activity in mice after in utero exposure as found at the only dose level in the developmental toxicity studies by Koskela *et al.* (2016) and Onishchenko *et al.* (2011) was chosen as the final value.

liver effects in animal toxicity studies, currently remains the preferred HBGV for chronic exposure to PFOA.

Further information on the molecular structure, restrictions and specifications for use in FCMs and information on the migration of PFOA can be found in Table 2.

8.1.2 *PFOS*

Comprehensive reviews of the toxicological data base of PFOS are provided by US-EPA (2016b), DWQI (2017a), EFSA (EFSA, 2018; EFSA *et al.*, 2018) and ATSDR (2018). The toxicity of PFOS was studied in a number of oral studies, including subacute studies (rat, mice), subchronic studies (rat, monkeys), neurotoxicity studies (rat, mouse), a 2-generation reproduction study (rats), developmental studies (rat, mouse) and a single chronic toxicity study in rats. In addition, hormonal disruption and developmental neurotoxicity were examined in several studies in rats and mice. Genotoxicity testing was done *in vitro* and, in a single study, *in vivo* (US-EPA, 2016b).
A large number epidemiology studies have studied the relationship between serum PFOS concentration and various health outcomes. These studies were evaluated by US-EPA (2016b), DWQI (2017a) and ATSDR (2018). The study populations consisted of workers at large-scale PFOS production plants in the USA and a residential population living near a PFOA production facility also in the USA (the C8 study). In addition, general population studies were done in Europe (Scandinavia, UK), Canada and the USA. Associations were reported between PFOS exposure and increased serum cholesterol and reproductive and developmental parameters. The strongest associations were found for increased total cholesterol and high density lipoproteins (HDLs). Associations, though less strong, were also observed between higher PFOS levels and decreases in female fecundity and fertility, in addition to decreased body weights in offspring, and other measures of postnatal growth. Several human epidemiology studies evaluated the association between PFOS and cancers including bladder, colon, and prostate, but these data present a small number or cases and some are confounded by failure to adjust for smoking. US-EPA (2016b) concludes that the associations for most epidemiology endpoints are mixed. Overall, the studies were used qualitatively in the identification of hazard, but not for the deduction of a quantitative dose response relationship. In the derivation of a HBGV for PFOS US-EPA used animal data (US-EPA, 2016b). DWQI (2017a) also reviewed the epidemiological studies for PFOS. It noted that some of these studies have yielded inconsistent results, lacked proper controlling for confounding, or could only provide weak suggestions of causality. For some effects (decreased vaccine response, increased serum uric acid/hyperuricemia, increased total cholesterol) DWQI considered the findings to be consistent among studies. Nevertheless, DWQI (2017a) concluded, as did US-EPA (2016b), that overall the epidemiological studies have limitations and in its derivation of an HBGV preferred an animal based point of departure over a human one. ATSDR (2018) reviewed the epidemiology data for PFOS and concluded there are two major limitations to establishing dose-response relationships based on the available epidemiology studies for PFOS and other PFASs. The first limitation identified by ATSDR (2018) is that the single exposure measurements in serum (at one point

in time only) used in the epidemiological studies constitute an uncertain indicator of chronic exposure given the fact that it is known that exposure levels were considerably higher in the past. The second major point of uncertainty is the fact that all studied populations most likely were exposed to a mixture of PFASs and that in most studies, this co-exposure was not quantified. Because of these limitations in the epidemiology data ATSDR (2018) used animal data in its derivation of MRLs for PFOS and several other PFASs.

In animal studies in mice, monkeys, and rats, PFOS induced a number of effects, the most important of which were liver effects (enlargement, histological effects, increased serum enzymes), neurological effects, immune effects and thyroid hormone effects. The studies showed increases in liver weight consistently at doses generally ≥ 0.5 mg/kg bw/ day. Co-occurring effects in these studies include decreased cholesterol, hepatic steatosis, lower body weight, and liver histopathology. For the liver effects in rodents the possible influence of PPARα was evaluated, leading to the conclusion that for PFOS a non PPARα mode of action most likely contributes to the rodent liver effects. Reproduction toxicity studies showed decreased pup survival and decreased pup weights. Additionally, in developmental neurotoxicity studies increased motor activity and decreased habituation and increased escape latency in the water maze test following *in utero* and lactational exposure to PFOS were found. Gestational and lactational exposures were also associated with higher serum glucose levels and evidence of insulin resistance in adult offspring. Limited evidence suggests immunological effects in mice (ATSDR, 2018).

PFOS has a harmonized European classification (EC Regulation No. 1272/2008, CLP) as carcinogenic category 2 (suspected of causing cancer), toxic for reproduction category 1B, inducing specific target organ toxicity (repeated exposure) in the liver (STOT RE 1 liver) and may cause harm to the breast-fed child (Lact).

IARC has not evaluated the data on carcinogenicity for PFOS. US-EPA (2016b) and DWQI (2017a) conclude that the epidemiological evidence for carcinogenicity is limited and does not allow a clear-cut conclusion. In the only animal carcinogenicity bioassay available, carried out in rats, increased incidences were found for liver adenomas/carcinomas and for thyroid follicular tumours. The increases, however, did not show a clear dose response relation. Genotoxicity studies for PFOS included an Ames test, mammalian-microsome reverse mutation assay, and an assay for chromosomal aberrations, an unscheduled DNA synthesis assay, and mouse micronucleus assay. The results were uniformly negative (DWQI, 2017a; US-EPA, 2016b). For the derivation of a HBGV for PFOS this means that a threshold in its toxic action is assumed and that a HBGV can be derived via application of the appropriate assessment factors to a selected point of departure in the form of an adequate NOAEL or BMDL.

Like PFOA, PFOS is eliminated from humans only very slowly with estimated average half-life values of 4.1–8.67 years. In contrast, half-life values for the monkey, rat, and mouse are 121 days, 48 days, and 37 days, respectively (US-EPA, 2016b).

For PFOS, HBGVs have been derived by a number of public organizations dealing with the risk assessment of substances. The table below provides an overview of these derivations.

EFSA (2008b) concluded that the liver was the major target for PFOS toxicity and used an NOAEL of 0.03 mg/kg bw/day from a subchronic

study in monkeys as the basis for a HBGV. In the derivation an extra factor of 2 was applied to compensate for uncertainties in connection to the relatively short duration of the key study and the internal dose kinetics (animal versus humans).
Similarly as for PFOA, later evaluations used the approach previously developed for polychlorinated dioxins, which quantitatively factors in the high potential for bioaccumulation in humans of PFOS. Thus, ATSDR, US-EPA and DWQI used a quantitative approach using a one-compartment model for calculation of the long-term human serum concentration equivalent with the mean animal serum concentration measured at the overall-NOAEL selected for HBGV-derivation. The RIVM has not derived an HBGV for PFOS.

Table 5. Overview of Health Based Guidance Values (HBGVs) for PFOS.

Organisation [reference]	Year	Duration	HBGV		Critical effect	Species
			In serum (ng/mL)	External dose (ng/kg bw/day)		
EFSA (2008b)	2008	Chronic	-	150	Liver, thyroid hormone, HDL	Monkey
ATSDR (2015)	2015	Sub-chronic (provi-sional)	404	30	Liver	Monkey
US-EPA (2016b)	2016	Chronic	199	20	Develop-mental[17]	Rat
DWQI (2017a)	2017	Chronic	23	1.8	Immune suppres-sion	Mouse
EFSA (EFSA, 2018; EFSA *et al.*, 2018)	2018	Chronic (provi-sional)	23[a]	13[a] ng/kg bw/wk	Choleste-rol	Human
ATSDR (2018)	2018	Sub-chronic (provi-sional)	22	1.7	Develop-mental[18]	Rat

[a] These values are not supported by RIVM, see text for further details.

[17] US-EPA (2016b) developed a number of candidate RfDs (=HGBVs) based on the following effects: increased serum ALT and serum BUN in a subchronic rat study, decreased pup survival in a developmental toxicity study in rats, increased motor activity and decreased habituation in a neurodevelopmental study in rats, decreased pup weight in a 2-generation reproduction study in rats, decreased pup weight in a 1-generation reproduction study in rats (dosing to dams only), decreased pup survival in a 1-generation reproduction study in rats (dosing to dams only). The RfD based on decreased pup weight at birth in a 1-generation reproduction study in rats (with treatment of dams only) by Luebker *et al.* (2005a) was chosen as the final value.
[18] ATSDR (2018) developed a number of candidate intermediate oral MRLs (=HBGVs for subchronic exposure) based on the following effects: increased rat pup mortality and lung histopathology in a developmental toxicity study in rats, decreased pup weight per litter in a 1-generation reproduction study in rats, delayed eye opening in F1 pups and transient decrease in F2 pup weights during lactation in a 2-generation study in rats, delayed eye opening in a developmental study in mice, Increased locomotor activity and concurrent failure to habituate to test environment in male pups in a developmental study in rats with dosing up to postnatal day 20, increased neonatal mortality in a developmental study in rats, liver effects in subchronic study in monkeys. The provisional MRL based on delayed eye opening in F1 pups and transient decrease in F2 pup weights during lactation in a 2-generation study in rats by Luebker *et al.* (2005b) was chosen as the final value.

Recently, EFSA published an update of its previous evaluation for PFOA and PFOS. In this new opinion EFSA proposes a PFOS HBGV based on benchmark dose modelling of selected health effects as found in epidemiological studies. However, due to the nature of the scientific uncertainties described in this opinion and in the minutes of the expert meeting (EFSA *et al.*, 2018), and the possible application of the forthcoming Scientific Committee guidance on combined exposure to multiple chemicals, the conclusions of this assessment and the derived HBGVs are considered provisional. In finalizing its opinion, EFSA noted a substantive divergence of opinion with the RIVM (Zeilmaker *et al.*, 2016) evaluation for PFOA. As explained above, this divergence was discussed by EFSA and RIVM (EFSA *et al.*, 2018). This discussion is focussed on PFOA but is also relevant for PFOS because the epidemiological data are very similar for the two substances. The questions that RIVM raised regarding the HBGV primarily pertain to the interpretation and analysis of data from epidemiological studies. RIVM holds the opinion that the scientific articles on which EFSA bases its recommendation do not contain enough data in order to derive a guidance value. Moreover, it is unclear whether, and to what extent, exposure to PFOS (or other perfluorinated compounds) causes the changes identified in the epidemiological studies. Lastly, RIVM raises questions about the data analysis method used to derive the guidance value.

8.1.3 *HFPO-DA (GenX)*

Hexafluoropropyleneoxide dimer acid (HFPO-DA), also referred to as GenX, is used to denote two substances with code names FRD-902 and FRD-903. FRD-902 is the dimer ammonium salt (ammonium-2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)-propanoate; CAS no. 62037-80-3) and FRD-903 is the dimer acid (2,3,3,3-tetrafluoro-2-(heptafluoropropoxy)propanoic acid; CAS no. 13252-13-6).

FRD-902 is used as production aid in the production of Teflon In this application FRD-902 replaces PFOA. During the production process FRD-903 is emitted to air and FRD-902 and FRD-903 are emitted to surface water.

In 2016 RIVM carried out a risk assessment for exposure to HFPO-DA via air of the population living in the vicinity of the DuPont/Chemours plant in Dordrecht. All available toxicological studies were performed with FRD-902. Read-across of the toxicological properties of FRD-902 to FRD-903 is considered justified (Beekman *et al.*, 2016).

The available toxicity data for the RIVM (Beekman *et al.*, 2016) evaluation were obtained from the ECHA registration dossier of that point in time. The registration dossier comprised animal data on toxicokinetics and a number of animal studies (carried out according to OECD guidelines), i.e. subacute and subchronic oral toxicity studies in rats and mice, an oral developmental toxicity study in rats, genotoxicity studies *in vitro* and *in vivo*, an oral reproduction toxicity study in mice and a chronic oral toxicity/carcinogenicity study in rats. No epidemiological studies are available for HFPO-DA.

The biokinetics of HFPO-DA were studied in rats, mice and monkeys (Gannon *et al.*, 2016). The results indicate that HFPO-DA in these

species has a lower potential for bioaccumulation compared to PFOA (half-lives between hours and days and between hours and weeks for HFPO-DA and PFOA respectively). Whether this lower potential for bioaccumulation by HFPO-DA also applies to humans is not known because data for HFPO-DA on bioaccumulation in humans are lacking.

In the animal toxicity studies in rats and mice, HFPO-DA caused increases in liver weight, induced changes in clinical chemistry parameters related to liver toxicity (AST, ALT, ALP), caused changes in red blood cell parameters (decreased serum cholesterol and increased albumin (A), decreased globulin (G), and increased A/G ratio), induced liver hypertrophy and liver microscopical changes, and induced tumours in testes, liver and pancreas. The mechanism for these effects is possibly by activation (directly or indirectly) of the peroxisome proliferator-activated receptor alpha (PPARα). This is, amongst others, indicated by a treatment-related increases in fatty acid beta oxidation in rodents upon exposure to HFPO-DA. Rodents are known to be more susceptible to PPARα mediated liver effects than are primates. For PFOA the interaction with PPARα in rodents has been studied extensively (especially many studies in mice) and the effects seen in rodents with HFPO-DA closely match those seen with PFOA. It should however be noted in this context that the liver effects for PFOA in rodents have been demonstrated to be in part non PPARα mediated, and whether this also is the case for HFPO-DA still needs further research. And, on the other hand, it should also be noted that although rodents are more susceptible than humans, PPARα mediated effects do occur in humans: this is clearly demonstrated by the fact that certain drugs (fibrates) that act via PPARα activation are applied in human medicine as a treatment for cholesterolemia. Based on the above, the hepatic effects as seen in rodents after treatment with HFPO-DA and PFOA are considered relevant for the risk assessment for humans. This is in agreement with RIVM (Beekman *et al.*, 2016; Zeilmaker *et al.*, 2016), US-EPA (2016a), DWQI (2017b) and ATSDR (2018).

All genotoxicity tests carried out with HFPO-DA (Ames-test, *in vitro* mammalian cell gene mutation assay, *in vitro* mammalian cell chromosome aberration test, *in vivo* micronucleus-test in mice) were negative (no effect). As already indicated above the only study on carcinogenicity (2-year study in rats) showed increased incidences of tumours in testes, liver and pancreas. RIVM (Beekman *et al.*, 2016) concluded that the available mutagenicity studies and mechanistic information indicate a non-genotoxic mode of action for the observed tumours in the 2-year study.

In the RIVM (Beekman *et al.*, 2016) evaluation, for HFPO-DA an overall NOAEL of 0.1 mg/kg bw/day was selected as the basis for deriving the general population limit values for oral and inhalation exposure. This oral NOAEL was derived from the chronic oral study in rats with increased albumin/globuline ratio in serum as the critical effect (LOAEL 1.0 mg/kg bw/day). Overt liver toxicity was only present at 50 mg/kg bw/day and above in this study. For the general population inhalation limit value the oral NOAEL of 0.1 mg/kg bw/day was converted to an inhalation NOAEC of 0.087 mg/m^3 (by dividing by the rat inhalation

volume of 1.14 m^3/kg bw/day). For the inhalation limit value the following assessment factors were applied to the oral NOAEL:

- interspecies (for remaining toxicodynamic differences, according to REACH guidelines): 1.8
- intraspecies (standard factor according to REACH guidelines): 10
- extra factor for possible bioaccumulation (based on PFOA-data): 66.

Thus, a tentative general population limit value of 73 ng/m^3 was derived. This value is tentative because the half-life for HFPO-DA in humans is unknown. To fill the data gap for HFPO-DA on bioaccumulation in humans an extra factor based on PFOA-data was applied.

Similarly for the oral tolerable daily intake (TDI), assessment factors were applied according to REACH guidelines:

- interspecies (standard interspecies for difference in kinetics): 4
- interspecies (interspecies for remaining toxicodynamic difference): 1.8
- intraspecies (standard factor): 10
- extra factor for possible bioaccumulation (based on PFOA-data): 66.

This resulted in a tentative oral TDI (Tolerable Daily Intake) of 21 ng/kg bw/day.

In the USA work is ongoing with the aim to derive a general population limit value for HFPO-DA. This is by the North Carolina departments of Environmental Quality (NC-DEQ) and Health and Human Services (NC-DHHS) and by the US-EPA. Information presented at the North Carolina state website 'https://deq.nc.gov/news/hot-topics/GenX-investigation' shows that the evaluation there is still ongoing. In 2017 NC-DEQ/DHHS used a subchronic NOAEL of 0.1 mg/kg bw/day from a 90-day mouse study as the basis for deriving a drinking water advisory but further evaluation of the toxicological dataset for HFPO-DA is in progress and different options for deriving the drinking-water advisory are still under discussion in the North Carolina Science Advisory Board (latest meeting on June 18, 2018). In the meeting of the North Carolina Science Advisory Board of June 2018 US-EPA gave an update about an ongoing evaluation for HFPO-DA by US-EPA aimed at developing toxicity values (RfDs for different durations) for the substances. Recently this has led to a public US-EPA draft (US-EPA, 2018) in which proposals for these are developed. In this draft, US-EPA (2018) derives a subchronic Reference Dose and a chronic Reference Dose, both based on single liver cell necrosis as seen in parental males in an oral reproductive/developmental toxicity study in mice with an NOAEL of 0.1 milligrams per kilogram per day (mg/kg/day). Using benchmark dose modelling, a $BMDL_{10}$ of 0.15 mg/kg bw/day was derived for the critical effect. This POD was converted to a human equivalent dose of 0.023 mg/kg bw/day by allometric scaling. The following uncertainty factors were applied to this POD: 10 for intraspecies variability, 3 for interspecies differences, and 3 for database deficiencies, including immune effects and additional developmental studies. This gave a subchronic RfD of 0.0002 mg/kg bw/day. For deriving a chronic RfD an

additional uncertainty factor of 3 was applied for extrapolation from subchronic to chronic duration (US-EPA, 2018).

For HFPO-DA no harmonized classification is available. In RIVM (Beekman *et al.*, 2016) the classification as proposed by the REACH registrant was reviewed. The conclusion was that the RIVM agrees with the classification as Acute Tox. 4; H302 and Eye Damage 1; H318 as proposed by the registrant. The proposal for STOT RE 2 based on liver and red blood cell effects was evaluated as inconclusive by RIVM. The registrant did not propose classification for carcinogenicity but RIVM considers Carc Cat 2 (suspected human carcinogen) justified based on the tumours seen in the chronic rat study. The registrant also proposed no classification for reproductive toxicity and RIVM agrees that the effects seen in de relevant studies would normally not lead to classification. However, one has to note that the results from the reproduction/developmental toxicity study in mice do not allow for final conclusions regarding the reproductive effects because the highest dose level tested only exerted minimal effects in the parental animals. RIVM also agrees that the available information does not warrant classification for mutagenicity. RIVM (Beekman *et al.*, 2016) also included a preliminary assessment by RIVM of the PBT-properties of HFPO-DA. The conclusions:

- being a perfluorinated compound, HFPO-DA is almost certainly persistent or very persistent;
- for possible bioaccumulation in humans no conclusion could be reached because of lack of data in this species;

HFPO-DA is less toxic compared to PFOA but no definitive conclusion on the T criteria can be reached since the substance is considered borderline T for STOT RE (Beekman *et al.*, 2016).

Further information on the molecular structure, restrictions and specifications for use in FCMs and information on the migration of HFPO-DA can be found in Table 2.

8.1.4 *ADONA*

3H-perfluoro-3-[(3-methoxy-propoxy)propanoic acid], ammonium salt is also referred to as ADONA, and has been used as a replacement of PFOA. According to the scientific opinion of EFSA on the substance, ADONA is non-genotoxic (EFSA, 2011a). It was tested in three *in vitro* genotoxicity tests with and without S9 mix from phenobarbitone/β-naphthoflavone-induced rat livers. The substance was found neither mutagenic in bacteria (Salmonella typhimurium strains TA1537, TA1535, TA98, TA100 and in E. coli WP2 uvrA) nor in mammalian cells (V79/HPRT). In the chromosomal aberration study with human peripheral lymphocytes, the test substance induced statistically significant increases in the frequency of cells with aberrations. Two *in vivo* studies, a chromosome aberration test in rat and a mouse micronucleus test, are covering the same endpoint and showed no indication of clastogenic potential *in vivo*.
Furthermore, a repeated-dose toxicity study was performed. In a subchronic oral rat study, haematotoxicity and liver toxicity were observed in male rats at 10 mg/kg bw/day. According to the EFSA scientific opinion, the NOAEL in this study was 3 mg/kg bw/day.

In an oral gavage developmental toxicity study in rats, treatment-related effects were reported on the survival of dams and pups (at 270 mg/kg bw/day and higher), on food consumption, dam body weight gain and on pup weight per litter (at 90 mg/kg bw/day and higher). According to the EFSA scientific opinion, the maternal and developmental NOAELs in this study were 30 mg/kg bw/day.
Finally it was mentioned that kinetic data indicated a serum half-life in male rats of 44 hours. The dose was faster eliminated in female than in male rats (EFSA, 2011a).
Further information on the molecular structure, restrictions and specifications for use in FCMs and information on the migration of ADONA can be found in Table 2.

8.2 Mixture toxicity

The wide range of industrial and commercial applications of PFASs results in exposure to a mixture of individual PFASs. As with other substances, the risk assessment of PFASs is hitherto carried out at the level of individual mixture components. This approach starts with the derivation of the highest chronic human daily intake which is without adverse effects, i.e. the Health Based Guidance Value (HBGV). However, as only HBGVs for PFOA and PFOS are available this approach cannot be applied to other PFASs.
Alternatively, the PFASs risk assessment may be performed by assuming the combined toxicity of PFAS mixture components to be based on the concept of dose-addition (EFSA, 2008a; 2013). In Zeilmaker *et al.* (2018) this concept is developed for several PFCAs, PFSAs and HFPO-DA (also referred to as GenX), assuming that they act in *a similar manner, with the same mechanism/mode of action*, resulting in dose-responses with the same shape but with different potencies for each of the individual substances (see Figure 7). Liver toxicity was considered to be the most sensitive toxic endpoint. Based on available subacute and subchronic oral toxicity studies in rodents, relative potency factors (RPFs) were derived for 20 individual PFASs, including GenX. Basically, the RPF method scales the dose of each substance, according to its potency, to a dose of the Index (reference) Compound (IC), with the IC having a RPF equal to 1. Combining the occurrence of each mixture component with its specific RPF value then expresses each of the mixture components in terms of IC equivalents. Summing over all mixture components then leads to mixture exposure expressed in term of IC equivalents. The latter then can be compared with IC HBGV, i.e. the HBGV of PFOA or PFOS.
Note that the presented approach can be applied to any PFAS containing matrix, i.e. food, soil, or PFASs migrated from consumer products or food contact materials.

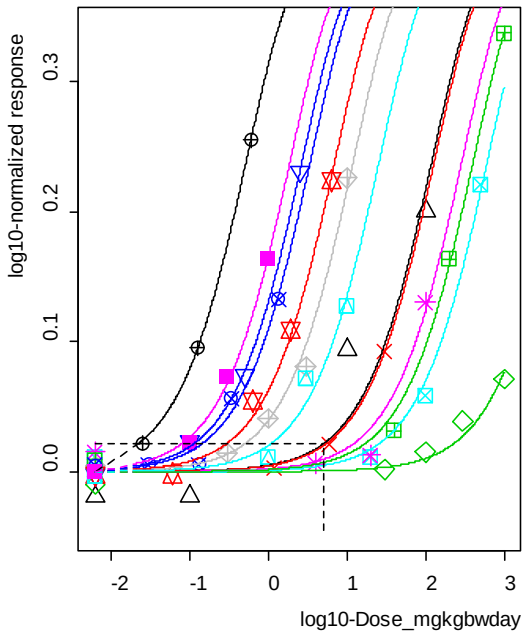


Figure 7: Dose responses of relative liver weight (in male rat) of a series of individual PFASs after normalizing to background. The dose responses have the same shape, i.e. the curves are parallel on the log-dose scale, but each PFAS has a different potency: different doses are required to result in the same increase in response (Zeilmaker et al., 2018).

Hence, with the proposed RPFs the combined effect on the liver for 20 PFASs can be estimated. However, more PFASs exist that might contribute as well, but for which insufficient toxicity data are available to derive RPF-values.

Table 6: Relative Potency Factors (RPFs) derived for several PFCAs, PFSAs and HFPO-DA (also referred to as GenX) from relative liver weight data. RPFs values using PFOA as the Index Compound, obtained from Zeilmaker et al. (2018), see also Figure 7.

PFAS group	PFAS	RPF
PFSA	Perfluorobutanesulfonate (PFBS)	0.001
	Perfluoropentane sulfonic acid (PFPeS)	0.001 ≤ RPF ≤ 0.6
	Perfluorohexanesulfonate (PFHxS)	0.6
	Perfluoroheptane sulfonic acid (PFHpS)	0.6 ≤ RPF ≤ 2
	Perfluorooctanesulfonate (PFOS)	2
	Perfluorodecane sulfonic acid (PFDS)	2
PFCA	Perfluorobutyrate (PFBA)	0.05
	Perfluoropentanoic acid (PFPeA)	0.01 ≤ RPF ≤ 0.05
	Perfluorohexanoate (PFHxA)	0.01
	Perfluoroheptanoic acid (PFHpA)	0.01 ≤ RPF ≤ 1
	Perfluorooctanoic acid (PFOA)	1
	Perfluorononaoic acid (PFNA)	10
	Perfluorodecanoic acid (PFDA)	4 ≤ RPF ≤ 10
	Perfluoroundecanoic acid (PFUnDA)	4
	Perfluorododecanoic acid (PFDoDA)	3
	Perfluorotridecanoid acid (PFTrDA)	0.3 ≤ RPF ≤ 3
	Perfluorotertadecanoic acid (PFTeDA)	0.3
	Perfluorohexadecanoic acid (PFHxDA)	0.02
	Perfluorooctadecanoic acid (PFODA)	0.02
PFECA	Hexafluoropropylene oxide-dimer acid (HFPO-DA) (FRD-902/FRD-903; also referred to as GenX)	0.06

Note that the presented dose addition method (Zeilmaker *et al.*, 2018) is based on effects on the liver as observed in animals studies, and is thereby in concordance with the risk assessment performed by several international scientific institutions (e.g. ATSDR and US-EPA). However, recently EFSA's CONTAM Panel reviewed PFOA and PFOS toxicity and has advocated the use of human epidemiological data over animal toxicity data as the starting point for the derivation of a HBGV. To which extent the RPFs are applicable to the epidemiological endpoints is unknown. For application in humans it is assumed that the potencies between substances within rat are equal to those within humans.

In 2019, EFSA aims to finalise an opinion on the human health risks related to the presence of perfluoroalkylated substances in food other than PFOS and PFOA (EFSA-Q-2017-00549). For this opinion, the possible application of a mixture approach will be considered. It is currently not known which method will be applied to assess a PFAS mixture. According to the disclaimer in EFSA's PFOA and PFOS opinion, at least a forthcoming EFSA guidance on harmonised methodologies for risk assessment of combined exposure to multiple chemicals (EFSA-Q-2015–00007) will be considered (EFSA, 2018).

9 Incineration and persistency of PFASs

Municipal waste, including FCMs, is often incinerated. Municipal waste incineration is carried out between 850 and 1100 °C (Huber *et al.*, 2009; Royal Haskoning, 2010). Some information is available on the destruction of PFASs at thermal heating, mostly on PFAS polymers. Between 400 and 600 °C, PTFE and most other fluoropolymers (FPs, see Figure 2) start to degrade (Huber *et al.*, 2009). At temperatures between 400 and 600 °C, the main degradation products of polytetrafluoroethylene (PTFE) polymers and most other fluoropolymers appear to be fluoroalkanes and alkenes, hydrogen fluoride, oxidation products (epoxides, aldehydes and acids), and fluoropolymer particulates (Huber *et al.* 2009). According to Watanabe *et al.* (2018), PFOA is destructed for 90% at 700 °C (referring to Takemine *et al.* 2013, article in Japanese), whereas a full-scale waste incinerator destructed a polymer chlorofluoro carbon with an efficiency of >99%. In laboratory studies PFOS degraded to more than 99% at 600 °C (Kucharzyk *et al.*, 2017). In the temperature range 700-1050 °C, CF4, CHF_3, C_2F_6, tetrafluoroethene (TFE) and hexafluoropropene (HFP) are major products, depending on incineration conditions like temperature, moisture, oxygen content, use of catalysts etc. (Huber *et al.*, 2009). Huber *et al.* did not assess whether all fluoropolymers were degraded. Based on the data available, they concluded that incineration of fluoropolymer containing products can contribute considerably to greenhouse gas emissions (Huber *et al.*, 2009).

Some studies investigated whether PFOA could be formed as degradation product after incineration of fluoropolymers. Taylor *et al.* (2014) did not find the formation of PFOA during incineration at 1000 °C of commercial fluorotelomer-based polymers (FTBPs). In this case, mainly hydrogen fluoride (HF) was formed during combustion (Taylor *et al.*, 2014). Also Yamada *et al.* (2005) did not find formation of PFOA under typical municipal incineration conditions of polyester/cellulose fabric treated with a fluorotelomer-based acrylic polymer.

Although the information on degradation of PFASs at incineration in general is limited, it seems possible that gaseous substances like CF_4, CHF_3, C_2F_6, trifluoroacetic acid (TFA), tetrafluoroethene (TFE) and hexafluoropropene (HFP) are formed that are ozone-depleting agents and/or potent greenhouse gases. Also, the formation of short-chain fluoro degradation products is possible, especially in case of lower temperature or incomplete incineration. The presence and persistence of these short-chain fluoro degradation products should be considered in view of their environmental effects. Via the environment or at incineration, human may be exposed as well. For example, gases TFE and HFP may cause cancer and may cause damage to organs (ECHA, 2018a; b). Some of the degradation products can react further, depending on the conditions. Because the conditions in the laboratory studies described in literature often differ from those in actual waste incinerators, it remains unknown to which extent these gaseous and short-chain fluoro substances are formed and emitted at incineration. For example, as opposed to the situation in waste incinerators, destruction of PFAS in laboratory studies is typically investigated by heating without flames.

As developed by a group of experts, the UN Environment (2017) recommends incineration at high enough temperatures as best environmental practices for PFOS and related substances, in order to thermally mineralise the fluorinates substances.

Since no information was obtained on the actual emissions of degradation products of PFASs from waste incineration plants, no conclusions can be drawn on the contribution of PFASs in FCMs to the greenhouse effect and environmental risks. Further research is therefore recommended.

10 Discussion and conclusion

10.1 PFAS definition and relevance of PFAS groups for toxicity

At present, there is no internationally agreement on the definition of PFASs. The most recent OECD report on PFASs (OECD, 2018) describes PFASs as substances containing a $–C_nF_{2n}–$ moiety (with n≥3) or a $-C_nF_{2n}OC_mF_{2m}–$ moiety (with n≥1 and m≥1). The advantage is that, in contrast to the previous definition (of $–C_nF_{2n+1}$, with n≥1), this definition excludes some small (volatile) fluorinated substances (with $–C_nF_{2n}–$, n<3), while in the meantime it covers possible PFOA and PFOS alternatives, which do not contain a $–C_nF_{2n+1}$ moiety. More specifically, some PFASs may be replaced by substances in which one fluor atom of the CF_3 group normally seen at the end of a chain, is changed into e.g. chloride. If PFASs are defined by containing a $–C_nF_{2n}–$ moiety rather than C_nF_{2n+1}, these substances are – under the most recent OECD definition – still considered PFASs. This increases the attention paid to such substances when they are proposed as candidates to the positive lists of various regulations on FCMs.

In concordance with Kemi (2015) and Trier *et al.* (2017), the authors of the present report hypothesise that the physical properties and toxicity of PFASs may be related to a general (fluoro)surfactant structure, i.e. a hydrophobic (water repellent) perfluorinated alkyl chain in combination with a hydrophilic (water soluble) group. Some substances may contain an additional “spacer” group, which links these two together (Figure 8). The water-soluble component can be made up of a wide range of different groups: a) anionic, such as carboxylates, sulfonates and phosphates, b) cationic, such as quaternary ammonium, c) non-ionic, such as acrylamide oligomers and polyethylene glycols, and d) amphoteric, such as betaines, sulfobetaines and amine oxides (KemI, 2015; Trier *et al.*, 2017).

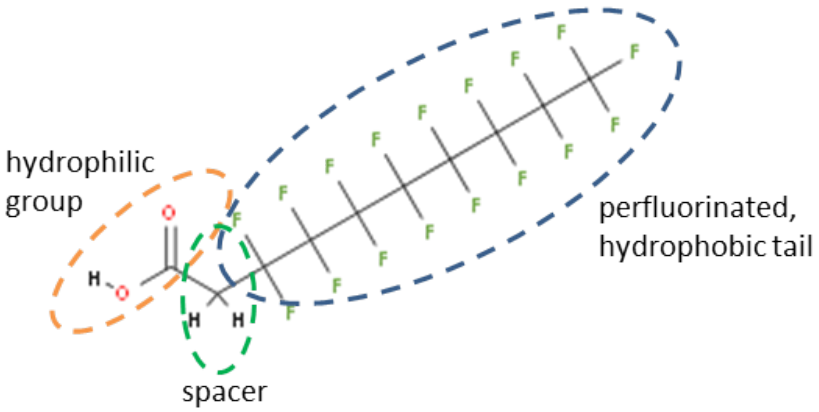


Figure 8: Illustration of a general structure of a fluorosurfactant.

If this is true, for risk assessment, emphasis should be on (linear) PFAAs, including PFCAs, PFSAs, per- and polyfluoroether acids and their

possible precursors (such as PAPs), as well as fluorotelomers. Other groups may be of lesser importance.

Conclusion: Based on the general structure of fluorosurfactants and current scientific insights on potential health and environmental risks, the focus for further research and monitoring should be on the (linear) PFAAs (including per- and polyfluoroether acids), their precursors, as well as fluorotelomers.

10.2 Legislation

For several PFAS, regulatory initiatives are being developed in the context of the REACH, the CLP and the POP regulation. As the concerns for PFAS relate to both human health and the environment, it can be expected that their use in FCM materials in time will be impacted by these activities (see Section 3.2 for further details).

The legislation of FCMs that applies to PFASs in the Netherlands and within Europe is inconsistent and complex. For plastics (and regenerated cellulose, which is not relevant for PFAS), the European legislation contains positive lists of substances. There are no European harmonised positive lists for other FCM matrices. The EU legislation on plastics (EU 10/2011) also includes a few substances that can be used in coatings. National regulations apply for the non-plastic materials, such as paper and board, rubber, silicone, and coatings. Due to mutual recognition of legislation between European countries, insight into the national legislations of other member states is required. This also means that it is not always clear what is allowed in the FCMs that are imported into the Netherlands. During the last decade very few new substances have been registered for rubber and paper and board under the Dutch “Warenwetregeling verpakkingen en gebruiksartikelen”, suggesting that substances might be used in FCMs that have not been added to the positive list, by lack of submission of an application. This indicates that substances might be used in FCMs that have not been added to the Dutch positive list (see also Section 10.3).

Furthermore, some substances are mentioned on positive lists since mid-1960. This especially holds for paper and board and rubber because almost all substances on these lists originate from the 20th century. For these substances, the underlying information is hard to trace, and if it is found, studies are only briefly summarized. For example, SN-di-PAP and mono- and di-PAP (see Table 2) were added to the positive list of paper and board in 1971, and the evaluations of the toxicity studies were found albeit very limited information was presented.

In the meantime, scientific insights in the behaviour and toxicity of these substances have evolved. The bioaccumulating potency of PFASs was for instance unknown at the time that these substances were put on the positive list; toxicity requirements are more extensive for substances that might be able to accumulate. Furthermore, no restrictions have generally been set for impurities in the authorised substances. Given the very low ADI’s for PFCA’s and their relative high migration potential from paper and paperboard (see Section 10.3 and 10.4), a specific restriction on the level of these impurities in the

starting substance might be needed. So, for substances that have been on one of these positive lists since decades, it is therefore recommended to gather the information available and re-assess the safety of the substance, including that of its impurities. Or, if insufficient information is available, generating additional data should be considered.

The inconsistency and complexity in legislation is also due to the fact that the various regulations mainly contain starting substances (including monomers), but also list polymers. Furthermore, groups of substances that contain multiple substances, e.g. C_6-C_{16} with a similar structure, are mentioned in these lists instead of individual substances. Finally, it should be noted that fluorotelomer alcohols have been found in silicone baking aids (Blom and Hanssen, 2015). There is no regulation for silicone baking aids in the Netherlands apart from the general requirement for food contact materials that “it should be safe” (EC, 2004, article 3). Therefore, substances used in silicone baking aids in the Netherlands are not restricted to substances included in the positive lists.

Comparing the PFAS entries between legislations appears to be difficult. This is due to 1) the high number of synonyms that exist for some of the PFASs, 2) the use of monomers (as regulated in EU legislation) that can form different oligomers or polymers that are not exactly described, 3) a series of different substances that may fall under the same entry (for example substances of different chain length), and 4) many PFASs do not have a CAS number and CAS numbers are often not mentioned on the positive lists of legislation, which means that a comparison based on CAS numbers cannot be made either.
Hence, further harmonisation of legislation within Europe for FCMs such as paper and board, rubber and coatings, may increase the transparency of the legislation applicable to PFASs. Alternatively, a substance specific European regulation on “PFASs in FCMs” could be developed, similar to the regulations considering e.g. bisphenol-A, and epoxy derivatives.

Other issues in the regulation that should be considered are:

- Migration limits usually refer only to the parent compounds, i.e. Non-Intentionally Added Substances (NIAS) are not considered. This includes impurities and reaction or degradation products that may also migrate.
- Migration limits do not consider wear of the products. However, wear may influence the migration properties and may be relevant for re-usable articles such as silicone baking aids, coatings on kitchen utensils and rubber conveyer belts.

Conclusion: The present legislation related to FCMs is complex and inconsistent across different national and EU regulations. As a result, the legislation related to PFASs in FCM is not fully clear. In addition, little attention is paid to NIAS, like impurities, and their potential health risks. Some substances are on a positive list (of allowed substances) in paper and board since decades, but the toxicity dossier is limited, difficult to retrieve, and may need re-assessment in view of current scientific insights.

European harmonisation of legislation, comprising all types of FCMs, would allow better and more effective regulation of PFASs used in FCMs and result in a better understanding of the PFASs allowed in FCMs. This would simplify adherence to the regulation and facilitate efficient enforcement.

10.3 Actual presence versus legally allowed PFASs in FCMs

Comparing the data on PFASs that were described in experimental studies to be present in FCM to the legally allowed PFASs, i.e. on positive lists in applicable regulations (Table 2), appeared to be difficult. This is due to the same four issues (see Section 10.2) hampering the comparison of PFAS entries between the various legislations. In addition, some of the substances may degrade to other PFASs. For example, fluorotelomers and PAPs may degrade to perfluorinated carboxylic acids (PFCAs) such as PFOA or perfluorinated sulfonic acids (PFSAs) (Fengler *et al.*, 2011; Wang *et al.*, 2014a; b).

Considering PFAAs (including PFEC/SA):
PFSAs and PFESAs are not mentioned in any FCM legislation. Of the PFCAs, only PFOA is allowed as a polymer production aid in the production of polymer coatings processed at high temperatures, for example in non-stick pans (see Table 2). Six PFECAs are allowed in FCMs. Five of them are allowed to produce coatings, requiring high processing temperatures, and one is mentioned in legislation on paper and board. Two PFP(I)As are registered in paper and board legislation. PFP(I)As have not been found in the few paper and board samples investigated (Figure 3 to Figure 6). As indicated, PFCAs are not mentioned on the positive list for paper and board. Nevertheless, a series of PFCAs are found in paper and board, as well as in a few cases in silicon baking aids.
PFCAs, including the substances PFOA and the toxicologically even more potent PFNA, has also been determined in the starting material for treatment of paper and board FCMs (Xu *et al.*, 2013). This suggests that the presence of these PFCAs in paper and board may be due to impurities or degradation products in the starting material (i.e. the technical-grade food contact substance used in production) or (un)intentional misuse of PFASs in FCM. Furthermore, Trier *et al.* (2017) indicates that non-intentional contamination of PFASs in paper and board may occur, e.g. if processing water is used in the paper manufacturing or due to recycling of paper.

Considering fluorotelomers:
A number of fluorotelomers or precursors thereof (e.g. fluorinated (meth)acrylate polymers) are allowed in paper and board. Such substances have also actually been determined in several paper/board matrices (see Figure 3 to Figure 6). The fluorinated side-chain of these substances may be released, and may further degrade to PFAAs.

Conclusion: Whether PFASs that are experimentally determined in FCMs are legally allowed, is not easily assessed. Although PFCAs are not allowed in paper and board, several PFCAs have been experimentally determined in these FCMs. This can be due to the presence of impurities in the technical-grade food contact substances (FCSs, used to treat the

paper and board), the presence of degradation products, contamination via e.g. recycled material or non-compliance with the legislation.

Furthermore, several FTs are allowed for use in paper and board. Given the toxicological relevance of PFCAs and FTs (see Section 10.1), their potential for migration (see Section 10.4) and the limited information available, further research to enable assessment of the potential health risk of PFCAs and FTs in paper and board is recommended.

No information is available on the presence of PFAS and PFAS impurities in plastic (including coatings) and rubber used in products on the market and very limited information on silicon baking aids.

10.4 Migration of PFASs from FCMs versus PFASs on positive list

The publicly available information on migration is rather limited. However, some noteworthy observations can be made based on the available studies. Overall, within the same type of FCMs, the migration of PFASs out of FCMs seems to increase with decreasing molecular size of the substance, and increasing contact time and temperature. In addition, migration depends on the type of food or simulant. Highest migration of PFAS from paper and board was observed into ethanol. These general observations were also mentioned by others like Trier *et al.* (2017) and Begley *et al.* (2005; 2008).

Under normal use conditions, low concentrations of PFCAs are released from non-stick coatings from kitchenware into food simulants and air. It is assumed that this is because PFAS are embedded in a polymer matrix, and are less subject to migration. It was also speculated (Schlummer *et al.*, 2015) that the low concentrations of PFCAs that are released are caused by slight thermolysis of fluoropolymers at high, but normal cooking temperatures.

It is observed that, although not allowed in food contact paper, PFCAs may be present in paper and board up to high concentrations as impurities or degradation products (see Section 6.3). Also, migration of PFCAs and FTOH from paper and board to food (simulants) is observed and can be substantial (up to 50% migration has been observed after a few hours for several PFASs, especially into ethanol containing matrices).
A very rough estimate on the exposure of several PFCAs from paper and board can be made. High concentrations of PFOA in paper and board FCMs are about 10 µg/m^2 (see Figure 5 and 6). Assuming a worst case estimate that is used as a default in the legislative evaluation for admittance of substances in FCMs of 1 kg food per day that has been in contact with 6 dm^2 of FCM, and a (probably worst-case) migration of 50% (see Chapter 7), the intake would be 0.3 µg PFOA per person. For a person of 60 kg, this would result in 5 ng/kg body weight/day. Although this is below the limit for external intake of PFOA according to RIVM of 12.5 ng/kg body weight/day, the exposure of PFOA via FCMs may be considerable. When considering the mixture of PFASs for which a relative potency factor has been proposed (see Chapter 8.2) and applying the same assumptions as for PFOA, an indication of the

cumulative exposure would be about 25 ng PFOA equivalents/kg body weight/day, with PFOS and PFOA contributing most.
As indicated, this exposure estimation is based on very limited data and can only be used to stress the importance of further information on the migration of these substances from paper and board FCMs.

Conclusion: Limited information is available on the migration of PFASs from FCMs. PFASs are usually only placed on positive lists with restrictions to assure that their migration into food will be limited. If such a requirement cannot be met, further (extensive) toxicity data are needed to enable a more detailed risk assessment. However, experimental findings on paper and board show that considerable amounts of PFCA and FTOH can migrate into food simulants, especially when containing alcohol.

Based on the limited data available (Chapter 6 and 7), a very rough and probably conservative estimation of the cumulative exposure from paper and board FCM can be made for PFOA, PFOS and other PFCAs for which a Relative Potency Factor has been proposed (Chapter 8.2). The exposure to PFOA equivalents can exceed the health based guidance value for PFOA. This estimate is highly uncertain and merely indicates the need for further data.

Information on migration of other PFAS from paper and board (e.g. PAPs) and for other FCMs (e.g. silicon baking aids) used in products on the market is lacking and further research is recommended.

Considering their embedding in FCMs and the available information on migration and content of PFASs, highest priority for human health risk assessment of PFASs in FCMs should be on its use in paper and board. Coatings are considered to be of lesser concern, because of the negligible release of PFASs from polymers. The priority for rubber and silicones is unclear because of lacking information and unknown strength of the incorporation of the PFASs into the polymeric matrix.

10.5 Difference TOF and sum of individual PFASs

The Nordic Council (Borg and Ivarsson, 2017) showed a comparison between summed levels of a number of individual PFASs and Total Organic Fluoride (TOF) in FCMs and other consumer products. For the four microwave popcorn bags investigated, the sum of measured PFASs represented between 0.001 and 0.05% of the TOF concentration. For two baking papers this sum of measured PFASs[19] represented up to 11% of the TOF concentration and for two popcorn papers 0.005 or 16%. This difference can be explained in several ways. First, the individual PFASs that were measured do not cover the entire spectrum of (allowed) non-polymer PFASs. Of the non-polymer PFASs, PFCAs with long chain length (e.g. C_{13}, C_{16}, C_{18}), PFSIAs, PFPAs and PFPIAs, and fluorotelomer carboxylic acids (FTCA and FTUCA) are not determined.

[19] The following individual PFASs were measured in several products: perfluorinated carboxylic acids (PFCAs): PFBA, PFHxA, PFOA, PFNA, PFDA; perfluorinated sulfonic acids (PFSAs): PFBS, PFHxS, PFOS; fluorotelomer alcohols (FTOHs): 4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH; fluorotelomer acrylates (FTAs): 6:2 FTA, 8:2 FTA, 10:2 FTA; fluorotelomer sulfonates (FTSs): 4:2 FTS, 6:2 FTS, 8:2 FTS. perfluorooctane sulfonamides (FOSAs): MeFOSA, EtFOSA; perfluorooctane sulfonamidoethanols (FOSEs): MeFOSE, EtFOSE (Borg and Ivarsson, 2017).

Alternatively or in addition, oligomers or polymers that can be applied to paper are not investigated, which can consist of fluorinated polymers which have perfluorinated sidechains attached to a carbon backbone. Hence, the small fraction of TOF that can be explained by the measured sum of several individual PFASs, can be due to the presence of non-polymer PFASs (or their degradation products) that are not measured or PFAS polymers.
The TOF concentrations in some silicon baking aids and baking paper are similar or in some cases even higher than for popcorn bags (for the latter up to 90 000 µg fluor/m^2 has been found). It is therefore likely that these high concentrations are due to the use of polymer PFASs, as polymers can have a high fluoride content.
Considering the high concentrations of TOF found in FCMs, a better understanding of the constitution of TOF would be relevant for health risk assessment.

In addition, at incineration, PFASs may be converted into fluorinated gases and short-chain fluoroalkanes. Considering the high TOF concentration that can occur in FCMs (up to 90 000 µg fluor/m^2) and the persistency and potential environmental effects of potential degradation products at incineration, further research on the emission by incineration plants and potential environmental risk is recommended. Also, the formation of greenhouse and ozone reducing gases should be included.

Conclusion: Total Organic Fluoride (TOF) concentrations in FCMs can be many fold higher than the sum of individual PFASs measured. This can either be explained by non-polymer PFASs that are not individually measured, or the presence of polymers.
Considering the high TOF concentration that can occur in FCMs and the persistency and possible effects of potential degradation products at incineration, more information on the actual emission of degradation products of PFASs from waste incineration plants is considered relevant.

10.6 Toxicity

Toxicological studies have focussed mainly on a few PFASs, such as PFOA, PFOS and HFPO-DA (also referred to as GenX) (see Chapter 8), of which risk assessments are generally performed per substance. However, exposure is likely to occur to multiple PFASs. Given that PFASs can have the same mode-of-action, with varying potency, the risk of the mixture should preferably be considered. A start for risk assessment of the mixture is being made with the relative potency factors developed for about 20 PFASs by Zeilmaker *et al.* (2018). In 2019, EFSA aims to write an opinion on the possible application of a mixture approach for PFASs.

Conclusion: Risk assessment of PFASs should preferably be performed for mixtures. Further development of relative potency factors for other PFASs like fluorotelomers is therefore relevant. Also extension of the development of relative potency factors for alternatives of PFOA and HFPO-DA, like ADONA, is recommended.

10.7 Recommendations on risk assessment of PFASs in FCMs

In order to do a risk assessment of PFASs in FCMs, information on exposure and hazard should be combined. At present, too limited information is available to make a reliable estimation of the exposure to PFASs from FCMs. A very rough estimation suggests that the exposure to PFOA equivalents can exceed the health based guidance value for PFOA (Section 10.4). This merely indicates the need for further data. Especially information on the migration of impurities and contaminants – also PFASs – from different types of FCMs is missing. The present limited information suggests that PFCAs and FTs may migrate (up to 50% migration after a few hours, especially into ethanol containing food simulants) from paper and board. Information on migration of other PFAS from paper and board (e.g. PAPs) and for other FCMs (e.g. silicon baking aids) is lacking and further research is recommended. Performing a risk assessment is considered opportune especially for PFAAs, FTs and PAPs (that may degrade to PFAAs), as in addition to their presence in and migration out of FCMs, their general structure indicates their relevance for risk.
Depending on the available information (see Section 10.8), in addition to information on exposure also more toxicity data may be required to perform a risk assessment.

10.8 Not investigated in the present study

Several aspects related to PFASs in FCMs were not investigated in detail.

- No toxicity information of PFAS other than PFOA, PFOS, HFPO-DA (GenX) and ADONA (see Chapter 8) has been gathered. Some further information may be available in the public literature, and it is recommended to get an overview.
- Information on the degradation pathways and rate of PFASs has not been investigated, but is relevant to assess which degradation products can be formed and under what conditions (in FCMs, food or the environment).
- Information on the presence and concentration of PFASs in food has not been investigated. These PFASs may have migrated from FCMs, but can also originate from environmental contamination.
- Information on migration of PFASs from plastic FCMs was obtained from EFSA opinions. Migration from other FCMs could be present in underlying national registration dossiers; no attempt has been made to retrieve dossiers from other countries than the Netherlands.
- Information on the mechanical wear of repeatedly used FCMs such as coatings on pans has not investigated.

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Appendix A: Regulations searched for the presence of PFASs

The following regulations were searched for PFASs and monomers for fluorinated polymers to get an overview of their use in FCM.

The regulations in which PFASs and monomers were encountered are indicated in bold.

EU Regulations
General EU legislation on FCM[20] contains of the framework Regulation No 1935/2004 (EC, 2004), which provides a harmonised legal EU framework setting out the general principles of safety and inertness for all FCMs. A second regulation, Regulation No 2023/2006 (EC, 2006), ensures that the manufacturing process is well controlled so that the specifications for FCMs remain in conformity with the legislation. In addition to the general legislation, certain FCMs are covered by specific EU measures:

- **Commission Regulation No 10/2011 on plastic materials (EU, 2011)**
- Commission Regulation No 450/2009 on active and intelligent materials (EC, 2009)
- Commission Regulation No 282/2008 on recycled plastic materials (EC, 2008)
- Council Directive 84/500/EEC on ceramic articles (EC, 1984)
- Commission Directive 2007/42/EC on materials and articles made of regenerated cellulose film (EC, 2007)

Other additional legislation is in place to regulate the use of specific substances and products originating or consigned from China or Hong Kong. None of this additional legislation addresses occurrence of PFASs in FCM.

National Regulations: the Netherlands
In the Netherlands, Part A of the Dutch Commodities Act Regulation on Packaging and Consumer Articles Coming into Contact with Foodstuffs (WVG, 2014) (latest amendment in 2016) has set a list of authorized substances, restriction limits and other requirements for 12 types of food contact material and articles, listed here:

I. plastics;
II. paper and cardboard;
III. rubber products;
IV. metals;
V. glass and glass ceramics;
VI. ceramic materials and enamels;
VII. textile products;
VIII. foil made of regenerated cellulose;
IX. wood and cork;
X. coatings;
XI. colourants and pigments;

[20] https://ec.europa.eu/food/safety/chemical_safety/food_contact_materials/legislation_en

XII. epoxy polymers.

PFASs and monomers are included in the regulations (in bold) on plastics, paper and cardboard, rubber products and coating. In the regulations on metals and textile products no specific PFASs or monomers are mentioned, but the specific migration limit for fluorinated substances is set to 1 mg total fluorine/kg. In paper and cardboard and rubber products the specific migration limit for fluorinated substances is also set to 1 mg total fluorine/kg.

National Regulations: Germany
German BfR has issued recommendations on the health assessment of food contact materials. The BfR Recommendations are not legally binding (i.e. they are not equivalent to e.g. SMLs). However, they do represent the current state of the scientific and technical knowledge for the conditions under which food contact materials made from silicones, paper, rubber etc., meet the requirements of § 31, para 1, German Food and Feed Code (Lebensmittel-, Bedarfsgegenstände- und Futtermittelgesetzbuch, LFGB) as well as those of Article 3, para 1 a of the Regulation (EC) No 1935/2004 in respect to their health safety.

The list of authorized food contact substances are included in various chapters of the BfR recommendations[21]. The various types of FCMs assessed by BfR are listed below. PFASs were identified in the chapters (in bold) on paper and board for food contact (Chapter XXXVI), paper and paperboard for baking purposes (Chapter XXXVI/2) and temperature resistant polymer coating systems for frying, cooking and baking utensils (Chapter LI).

I. High Polymers Containing Plasticizers.
II. Plasticizer-free polyvinyl chloride, plasticizer-free copolymers of vinyl chloride and mixtures of these polymers with other copolymers and chlorinated polyolefins containing mainly vinyl chloride in the total mixture.
III. Polyethylene.
V. Polystyrene Produced exclusively from the Polymerisation of Styrene.
VI. Styrene Copolymers and Graft Polymers, and Mixtures of Polystyrene with other Polymers.
VII. Polypropylene.
IX. Colorants for Plastics and other Polymers Used in Commodities.
X. Polyamides.
XI. Polycarbonates and Mixtures of Polycarbonates with other Polymers or Copolymers.
XII. Unsaturated Polyester Resins.
XIV. Polymer Dispersions.
XV. Silicones.
XVI. Polyvinyl Ethers.
XVII. Poly(terephthalic acid diol esters).

[21] https://BfR.ble.de/kse/faces/DBEmpfehlung_en.jsp

XX. Polyisobutylene, Isobutylene Copolymers and Mixtures of Polyisobutylene with other Polymers.
XXI. Commodities based on Natural and Synthetic Rubber.
XXII. Polymers Based on Esters of Acrylic and Methacrylic Acids, their Copolymers, and Mixtures of these with other Polymers.
XXV. Hard Paraffins, Microcrystalline Waxes and Mixtures of these with Waxes, Resins and Plastics.
XXVIII. Cross-Linked Polyurethanes as Adhesive Layers for Food Packaging Materials.
XXX. Conveyor Belts Made from Gutta-Percha and Balata.
XXXIII. Acetal resins.
XXXIV. Vinylidene Chloride Copolymers with a Predominant Content of Polyvinylidene Chloride.
XXXV. Copolymers of Ethylene, Propylene, Butylene, Vinyl Esters and Unsaturated Aliphatic Acids, and their Salts and Esters.
XXXVI. Paper and Board for Food Contact.
XXXVI/1. Cooking Papers, Hot Filter Papers and Filter Layers
XXXVI/2. Paper and Paperboard for Baking Purposes.
XXXVI/3. Absorber pads based on cellulosic fibres for food packaging.
XXXVII. Polybutene-(1).
XXXIX. Commodities Based on Polyurethanes.
XLI. Linear Polyurethanes for Paper Coatings.
XLII. Plasticizer-Free Chlorinated Polyvinyl Chloride, Plasticizer-Free Chlorinated Copolymers of Vinyl Chloride and Mixtures of these Polymers with other Copolymers.
XLIII. Poly(4-methylpentene-1).
XLIV. Artificial Sausage Casings.
XLVI. Cross-linked Polyethylene.
XLVII. Toys Made from Plastics and other Polymers, and from Paper and Paperboard.
XLVIII. Materials for Coating the Outside of Hollow Glassware.
XLIX. Soft Polyurethane Foams as Cushion Packaging for Fruit.
L. Copolymers and Graft Polymers of Acrylonitrile.
LI. Temperature Resistant Polymer Coating Systems for Frying, Cooking and Baking Utensils.
LII. Fillers.
LIII: Absorber pads and packagings with absorbing function, in which absorbent materials based on cross-linked polyacrylates are used, for foodstuffs

National Regulations: Spain
In Spain, the national regulation Real Decreto 847/2011 (Ministerio de Sanidad Politica Social e Igualdad, 2011) has to be met for the following food contact materials:

a) Adhesives
b) Elastomers and natural and synthetic rubbers
c) Ion exchange resins
d) Silicones
e) Varnishes and coatings
f) Plastic materials acting as support for polymerization production

PFASs and monomers were encountered in the regulation (part B) on other components for the manufacture of food contact materials within the scope of the Royal Decree.

National Regulations: Belgium
In Belgium, the main legislation regulating food-contact materials and articles the Royal Decree of 11 May 1992[22] and its amendment[23]. This law also sets specific requirements for certain food-contact materials such as glass**,** metal and alloys**,** paper and board**,** varnishes and coatings. PFASs were only encountered in annex 4 (on paper and board) of the Royal Decree.

ESCO WG
In 2012 the EFSA-ESCO working group on non-plastic food contact materials published a compilation of lists of substances for non-plastic food contact materials, evaluated in Member States, Switzerland and Norway (EFSA-ESCO WG, 2012). This compilation of lists has no legal status, but is rather an inventory of the substances present in national legislations or recommendations in member states, on coatings, colorants, cork & wood, paper & board, printing inks, rubber and silicones.
Several PFASs were encountered in this compilation. The entries by the Netherlands, Germany and Spain were considered to be covered by the individual national regulations mentioned above. Entries by the Czech Republic did not mention PFASs. In the entries of France and Italy PFASs were encountered in the lists of paper & board, rubber and silicones.

Sources of information that were not used
The following other sources of information were identified, but not used to fill out Table 2 for specific reasons mentioned for each source.

Council of Europe Resolutions
The Council of Europe (CoE) has established general recommendations for various types of food contact materials. Specific requirements such as positive lists and restriction conditions are set in the Appendixes of the CoE resolutions. The resolutions are not legally binding before they are transferred into national law. Therefore, this source of information is not used to fill out Table 2. PFASs and monomers mentioned in the recommendations of the CoE are listed in Appendix B of this report.

Nordic council of Ministers
The Nordic council of Ministers (of Denmark, Norway, Sweden, Finland, Iceland, Faroe Islands, Greenland and Åland) has published a series of guidelines with requirements on in-house control and the documentation for the assurance of compliance with Nordic food contact legislation on printing inks, paper & board and metals & alloys (Alsing Pedersen *et al.*, 2012; Licht Cederberg *et al.*, 2015; Nordic Council of Ministers, 2008). No PFASs are mentioned in these guidelines. However, in the metals & alloys document future work on PFASs is recommended: “Many metal

[22] https://www.health.belgium.be/fr/node/25016
[23] https://www.health.belgium.be/sites/default/files/uploads/fields/fpshealth_theme_file/rd_25_09_2016_varnish.pdf

food contact materials are coated – like aluminium frying pans coated with the fluoropolymer Teflon® (PTFE). Knowledge on impurities in the metals used and release when the coating is no longer intact is limited."

National Regulations: France
The French national regulation concerns the following food contact materials:

- Rubber [Order of 9 November 1994]
- Silicone elastomer [Order of 25 November 1992] ;
- Aluminum [Order of 27 August 1987];
- Stainless steel [Order of 13 January 1976].

Other specific measures concerning food contact materials are in force:

- Decree of 28 June 1912 [packaging of foodstuffs] and decree of 19 November 1945 [measuring instruments and receptacles];
- Order of 8 September 1999 [cleaning products for materials and articles intended to come into contact with foodstuffs, products and beverages for human and animal consumption] and Decree No. 73-138 of 12 February 1973 .
- Order of 12 August 1986 [ionizing radiation treatment of materials and articles placed or intended to be brought into contact with foodstuffs, products and beverages intended for food];

More info can be found here. It is assumed that the ESCO WG list already covers the PFASs mentioned in the French legislation. Therefore, the French national regulations were not used to fill out Table 2.

National Regulations: Italy
In Italy, the national regulation Decreto Ministeriale of 21 March 1973 (latest amendment in 2015) has set detailed requirements on various food contact materials

- Decreto Ministeriale of 21 March 1973 in Italian
- 2009 Amendment
- 2013 Amendment
- 2015 Amendment (only related to stainless steel)

It is assumed that the ESCO WG list already covers the PFASs mentioned in the French legislation. Therefore, the French national regulations were not used to fill out Table 2.

WIV-ISP list
Database of Substances known by Member States of Council of Europe and used in Food Contact Materials[24]. This database is an initiative of Belgian authorities and is supported by contributions of the Council of Europe (CoE): the Belgian Scientific Institute for Public Health is managing the database of substances known by the member states of the Council of Europe (CoE) and used in Food Contact Materials.
It is assumed that the regulations in Section 5.1 already cover the PFASs allowed in the EU. Therefore, the WIV-ISP list was not used to fill out Table 2.

[24] https://fcm.wiv-isp.be/Default.aspx

JRC baseline study
In this JRC study (Simoneau *et al.*, 2016) national regulations on food contact materials were compared, but no exhaustive positive lists were presented. Therefore, the JRC study was not used to fill out Table 2.

Appendix B

Table B-1. PFASs mentioned in ResAP by the Council of Europe (https://www.edqm.eu/en/resolutions-policy-statements). These substances are mentioned in List B or C, indicating that the risk assessment of substances in List B and C is unclear according to current standards or is lacking.

- Resolution ResAP on PAPER AND BOARD MATERIALS AND ARTICLES INTENDED TO COME INTO CONTACT WITH FOODSTUFFS Version 4 – 12.02.2009. https://rm.coe.int/CoERMPublicCommonSearchServices/DisplayDCTMContent?documentId=09000016804e4794
- Resolution ResAP on COATINGS INTENDED TO COME INTO CONTACT WITH FOODSTUFFS
- Version 3 - 12.02.2009 https://rm.coe.int/CoERMPublicCommonSearchServices/DisplayDCTMContent?documentId=0900001680515 6e5
- Resolution ResAP on SILICONES USED FOR FOOD CONTACT APPLICATIONS Version 1 – 10.06.2004 https://rm.coe.int/CoERMPublicCommonSearchServices/DisplayDCTMContent?documentId=09000016804e206f
- Resolution ResAP on RUBBER PRODUCTS INTENDED TO COME INTO CONTACT WITH FOODSTUFFS. Version 1 – 10.06.2004 https://rm.coe.int/09000016804e9fce
- Resolution ResAP on CORK STOPPERS AND OTHER CORK MATERIALS AND ARTICLES INTENDED TO COME INTO CONTACT WITH FOODSTUFFS. Version 2 – 05.09.2007 https://rm.coe.int/CoERMPublicCommonSearchServices/DisplayDCTMContent?documentId=09000016804e47f6
- Resolution ResAP on ION EXCHANGE AND ADSORBENT RESINS USED IN THE PROCESSING OF FOODSTUFFS. Version 3 – 28.01.2009 https://rm.coe.int/CoERMPublicCommonSearchServices/DisplayDCTMContent?documentId=09000016804daaf3

CAS No	Substance name [group]*	Remarks	Source
67969-69-1	N-Ethyl-N-(2-hydroxyethyl)perfluorooctanesulfonamide phosphate, diammonium salt [SN-mono-PAP/PFPA]		ResAP paper/board
68310-75-8	(Perfluorooctylsulfonylaminopropyl) trimethylammonium iodide		ResAP paper/board
25268-77-3	Acrylic acid, N-methylperfluorooctanesulfonamido-ethyl ester		ResAP paper/board

CAS No	Substance name [group]*	Remarks	Source
	[PASF-based derivative]		
NA	2-(Diethylamino)ethyl methacrylate – 2,3-epoxypropyl methacrylate – perfluoroalkyl(C_4-C_{18})ethyl acryl		ResAP paper/board
NA	2-(Dimethylamino)ethyl methacrylate – perfluoroalkylethyl acrylate – vinyl acetate, **copolymer**		ResAP paper/board
479029-28-2	Methacrylic acid, 2-(dimethylamino)ethyl ester, **polymers** with gamma-omega-per- fluoro-C_{8-14}-alkyl acrylate, acetates, N-oxides [fluorotelomer]	Voluntary withdrawn (Clariant)	ResAP paper/board list B
783306-31-0	Methacrylic acid, 2-(dimethylamino)ethyl ester, **polymers** with gamma-omega-per- fluoro-C_{8-14}-alkyl acrylate, N-oxides		ResAP paper/board
e.g. 92332-25-7	Phosphoric acid, mono- and bis(gamma, omega-perfluoroalkyl) esters, compounds with diethanolamine		ResAP paper/board
92265-81-1	2,3-Epoxypropyl methacrylate - 2-ethoxyethyl acrylate - N-methylperfluorooctane-sulfonamidoethyl acrylate - trimethylethanolammonium chloride methacrylate, **copolymer**		ResAP paper/board
9011-17-0	Vinylidene fluoride hexafluoropropylene **copolymers**	Toxicity is determined by monomers, see CAS 116-15-4 and 75-38-7	ResAP rubber
25190-89-0	Vinylidene fluoride, hexafluoropropylene, tetrafluoroethylene **copolymers**	Toxicity is determined by monomers, see CAS 116-14-3, 116-15-4 and 75-38-7	ResAP rubber
64706-30-65	Rubber, fluorinated		ResAP silicones

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