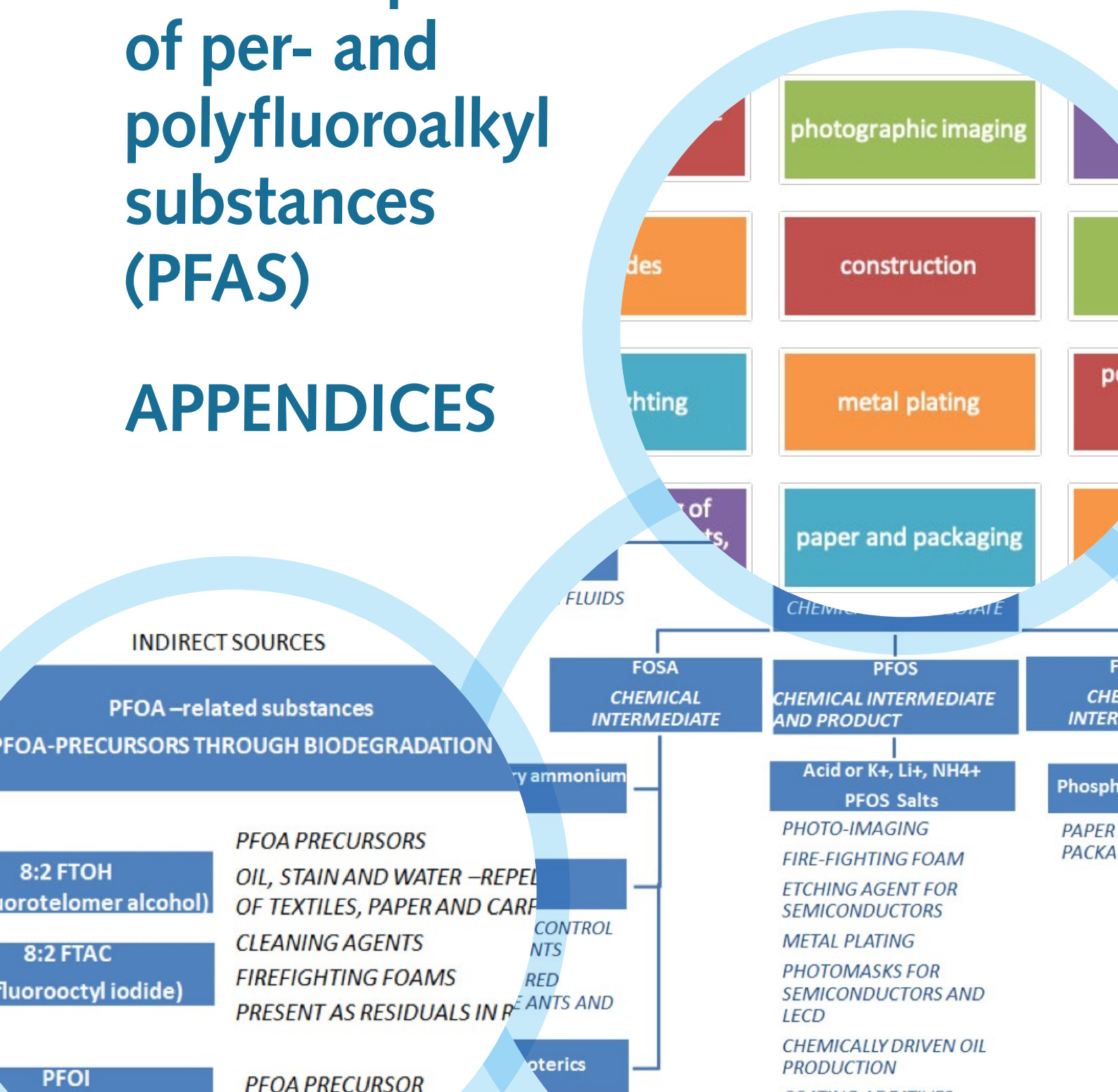


Scoping study for addressing risks to private water supplies from the presence of per- and polyfluoroalkyl substances (PFAS)

APPENDICES



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APPENDIX I

I.1 FAMILIES OF PFAS SUBSTANCES (NON-POLYMERS AND POLYMERS AND EXAMPLES OF INDIVIDUAL PFAS SUBSTANCES PER FAMILY SOURCE: BUCK ET AL 2011; OECD 2013.

Table AI1: Suite of PFASs discussed in this report.				
PFAS Classes	Chemical Name	Abbreviation	CAS RN	Molecular Formula
NON-POLYMERIC				
Perfluorocarboxylic Acids (PFCA)	Perfluorobutanoic acid	PFBA	375-22-4	C ₄ F ₇ COOH
	Perfluoropentanoic acid	PFPeA	2706-90-3	C ₅ F ₉ COOH
	Perfluorohexanoic acid	PFHxA	307-24-4	C ₆ F ₁₁ COOH
	Perfluoroheptanoic acid	PFHpA	375-85-9	C ₇ F ₁₃ COOH
	Perfluorooctanoic acid	PFOA	335-67-1	C ₈ F ₁₅ COOH
	Perfluorononanoic acid	PFNA	375-95-1	C ₉ F ₁₇ COOH
	Perfluorodecanoic acid	PFDA	335-76-2	C ₁₀ F ₁₉ COOH
	Perfluoroundecanoic acid	PFUnA	2058-94-8	C ₁₁ F ₂₁ COOH
	Perfluorododecanoic acid	PFDoA	307-55-1	C ₁₂ F ₂₃ COOH
	Perfluorotridecanoic acid	PFTTrDA	72629-94-8	C ₁₃ H ₂₅ O ₂
Perfluorosulfonic Acids (PFSA)	Perfluorobutane sulfonic acid	PFBS	375-73-5	C ₄ F ₉ SO ₃ H
	Perfluorohexane sulfonic acid	PFHxS	355-46-4	C ₆ F ₁₃ SO ₃ H
	Perfluorooctane sulfonic acid	PFOS	1763-23-1	C ₈ F ₁₇ SO ₃ H
	Perfluorodecane sulfonic acid	PFDS	335-77-3	C ₁₀ F ₂₁ SO ₃ H
	Perfluorooctane sulfonamide	FOSA	754-91-6	C ₈ F ₁₇ SO ₂ NH ₂
	N-methyl perfluorooctane sulfonamidoacetic acid	N-MeFOSAA	2355-31-9	C ₈ F ₁₇ SO ₂ N(CH ₃)CH ₂ CO ₂ H
Perfluoroalkane sulfonic acids (PFSIA)	N-ethyl perfluorooctane sulfonamidoacetic acid	N-EtFOSAA	2991-50-6	C ₁₂ H ₈ F ₁₇ NO ₄ S C ₈ F ₁₇ SO ₂ N(C ₂ H ₅)CH ₂ CO ₂ H
	Perfluorooctane sulfinic acid	PFOSI	647-29-0	C ₈ HF ₁₇ O ₂ S
Perfluoroalkyl phosphonic acids (PFPA)	Perfluorooctyl phosphonic acid	PFPA	252237-40-4	C ₈ H ₆ F ₁₃ O ₃ P
Perfluoroalkyl phosphinic acids (PFPIA)	Bis(perfluorooctyl)phosphinic acid	PFPIA	40143-79-1	C ₁₆ HF ₃₄ O ₂ P
Perfluoroalkane sulfonyl fluoride (PASF ¹)	Perfluorobutane sulfonyl fluoride	PBSF	375-72-4	C ₄ F ₉ SO ₂ F
	Perfluorooctane sulfonyl fluoride (POSF)	POSF	307-35-7	C ₈ F ₁₇ SO ₂ F
Perfluoroalkanesulfonamides(FAS)	Perfluorooctanesulphonamide	(FOSA) or (PFOSA)	754-91-6	C ₈ F ₁₇ SO ₂ NH ₂
Fluorotelomer (FT-) based substances	4:2-fluorotelomer unsaturated carboxylic acid	4:2 FTUCA	20825-07-4	C ₃ F ₁₅ CF=CHCOOH
	6:2-fluorotelomer unsaturated carboxylic acid	6:2 FTUCA	261503-40-6	C ₅ F ₁₅ CF=CHCOOH
	8:2-fluorotelomer unsaturated carboxylic acid	8:2 FTUCA	70887-84-2	C ₇ F ₁₅ CF=CHCOOH
	10:2-fluorotelomer unsaturated carboxylic acid	10:2 FTUCA	70887-94-4	C ₉ F ₁₅ CF=CHCOOH
	4:2-fluorotelomer sulfonic acid	4:2 FTSA	36839-98-2	C ₄ F ₉ CH ₂ CH ₂ SO ₃ H
	6:2-fluorotelomer sulfonic acid	6:2 FTSA	27619-97-2	C ₆ F ₁₃ CH ₂ CH ₂ SO ₃ H
	8:2-fluorotelomer sulfonic acid	8:2 FTSA	39108-34-4	C ₈ F ₁₇ CH ₂ CH ₂ SO ₃ H
	4:2 Fluorotelomer iodide	4:2 FTI	2043-55-2	C ₄ F ₉ CH ₂ CH ₂ I
	6:2 Fluorotelomer iodide	6:2 FTI	2043-57-4	C ₆ F ₁₃ CH ₂ CH ₂ I

¹ The Stockholm Convention reports them as PFOSF.

	8:2 Fluorotelomer iodide	8:2 FTI	2043-53-0	C ₈ F ₁₇ CH ₂ CH ₂ I
	4:2 Fluorotelomer olefin	4:2 FTO	19430-93-4	C ₄ F ₉ CH=CH ₂
	6:2 Fluorotelomer olefin	6:2 FTO	25291-17-2	C ₆ F ₁₃ CH=CH ₂
	8:2 Fluorotelomer olefin	8:2 FTO	21652-58-4	C8F17CH=CH2
	4:2-Fluorotelomer alcohol		2043-47-2	C ₄ F ₉ CH ₂ CH ₂ OH
	6:2-Fluorotelomer alcohol		647-42-7	C ₆ F ₁₃ CH ₂ CH ₂ OH
	8:2-Fluorotelomer alcohol		678-39-7	C ₈ F ₁₇ CH ₂ CH ₂ OH
Perfluoroalkyl iodides(PFAIs)	Perfluorohexyl iodide	PFHxI	355-43-1	C ₆ F ₁₃ I
Per- and polyfluoroalkyl ether (pfpe)-based compound				
Perfluoroalkyl aldehydes (pfals) and aldehyde hydrates(PFAL. H ₂ O _s)	Perfluorononanal	PFNAL	63967-40-8	C ₈ F ₁₇ CHO
POLYMERIC PFASs				
Fluoropolymers	Polytetrafluoroethylene (PTFE)			
	Polyvinylidene fluoride (PVDF)			
	Fluorinated ethylene propylene (FEP)			
	Perfluoroalkoxyl polymer (PFA)			
Side-chain fluorinated polymers				
Fluorinated (meth)acrylate polymers				
Fluorinated urethanepolymers				
Fluorinated oxetane polymers				
PERFLUOROPOLYETHERS				

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APPENDIX II MATERIAL AND METHODS IN DETAIL

II.1 LITERATURE REVIEW SEARCH TERMS AND OUTPUT

Table AII1. Words-phrases used as search terms or keywords in line with the objectives of the present scoping study.		
Key words	Search Engine	Output
Perfluorinated chemicals	WoS	394
+Scotland	WoS	0
+drinking water NOT blood NOT mouse NOT method	WoS	14
Perfluorinated +drinking water+threshold	WoS	10
"drinking water standards" monitoring PFC	WoS	0
PFC leachate OR foam OR contaminated land) (method or monitoring	WoS	12
Perfluorinated chemicals	Science Direct (title abstract or keywords)	482
+Scotland	Science Direct (title abstract or keywords)	0
+drinking water NOT blood Not mouse	Science Direct (title abstract or keywords)	5
Perfluorinated +drinking water+threshold	Science Direct (title abstract or keywords)	5
"drinking water standards" monitoring PFC	Science Direct (title abstract or keywords)	0
	Anywhere in article	10
Perfluorinated chemicals	Google Scholar (anywhere in the article)	4750
	In the title	221
+Scotland	Google Scholar (Anywhere in the article)	281
	(in the title)	0
+Scotland _drinking water NOT blood Not mouse OR rat Not method	Google Scholar (Anywhere in the article)	10
"drinking water standards" monitoring PFC	Google Scholar (Anywhere in the article) -2018	9
PFC and PFOA and PFOA leachate OR foam OR monitoring OR method "contaminated land" -remediation	Google Scholar (Anywhere in the article) -2018	9
PFC and PFOA and PFOA "leachate OR foam " "drinking water" -remediation -exposure -removal	Google Scholar (Anywhere in the article) -Any time	8
FAOLEX perfluor* "drinking water"		0

II.2 DATA SOURCES FOR MAPPING PFAS RISK TO PWS IN SCOTLAND

The data used for developing the GIS-based risk-mapping method refer to²:

1. A data set of PFAS concentrations in groundwater based on 40 borehole samples collected under SEPA's groundwater monitoring programme from 2013-2016 at 17 sites located in 10 different Local Authorities (LAs). Data source: SEPA. PFAS determination: LC-MS/MS using solid phase extraction (Isla Smail, SEPA, pers. com., June 2018).
2. Locations of Type A and Type B PWS as of 2015. Data source: DWQR.
3. Boundaries of Surface water bodies and groundwater bodies delineated to support the implementation of the WFD and Groundwater Daughter Directive. Data source: SEPA.
4. Boundaries of Local Authorities. Data source: Ordnance Survey (OS) UK.
5. Locations of airfields. Information on currently operating airports is available via the National Air Traffic Service (NATS) website (<http://www.nats-uk.ead-it.com/public/index.php.html>). This was supplemented with information from the UK Airfield Guide (<http://www.ukairfieldguide.net/>³). This website was downloaded using wget (<https://www.gnu.org/software/wget/>), then the pages with individual airfield information were parsed using Python to extract information on location, period of operation and any other notes of interest. Airfield location data were collated into a single file, loaded into ArcGIS (ver 10.5), plotted as a layer, and the sites in Scotland were selected for further analyses. The final dataset was saved as both a shapefile and geodatabase feature class. Number of sites: 396.
6. Locations of Fire Stations. A composite dataset was created based on:
 - a. The list of fire stations in each of Scottish Fire Service' divisions (East, West and North) (found in: <http://www.firescotland.gov.uk/>) was scraped from the website and parsed using Python to extract name and location information. The sites were assumed to be currently operational.
 - b. Data from a private website, UK Fire Stations (<https://www.ukfirestations.co.uk/>) was also used. This Information is presented by brigade (East, West and North) but includes sites which are not listed on the official Fire Service site. Location information was obtained in the same manner as above, but when plotted there are some discrepancies between locations for stations which appear on both lists. An 'eyeball' of a selection of sites suggests this is down to how the location information (postcode) of the station has been interpreted, and, interestingly, it seems that the unofficial site locations are more accurate, with the official site using the centroid of the postcode area, rather than the actual location of the station.
 - c. The Ordnance Survey Mastermap product (used under license, Crown Copyright 2018, All rights reserved. The James Hutton Institute, Ordnance Survey Licence Number 100019294) with feature description including the word 'fire' was also used. These were then reviewed to exclude records of no relevance (typically place names containing fire, fire lookout towers etc). Number of sites: 382.

Caveat: It was impossible to find out what type of materials these sites may store or use.

7. Fire and Rescue Services (FRS) data. A Freedom of Information request (FOI-007377-2018 CRM:0023091) was made to the Fire Service on 16/01/2018 asking for: location of fire-fighting training centres; location and any other information about use of foams at incidents (if this information is recorded in their system). The dataset was delivered in a EXCEL file which provided the following information:
 - Code and Date of incidence (2013-2017)
 - Incident category (Fire or Special Service, the latter not defined)
 - Incident type referring to: dwelling fire, vehicle fire, other building, other primary fire, refuse fire, outdoor fire, and Special Service (-RTC, -Hazardous Materials Incidents, -Spills and Leaks NOT RTC, Other transport incident).
 - Council (30 out of 32 Local Authorities) and Electoral Wards
 - Description, which potentially refers to type of foam used: High Expansion, Medium Expansion, Low Expansion, Other (CAFS).

² Where location information was given in WGS84 Web Mercator projection (for Google Maps), the dataset was re-projected to OSGB (British National Grid).

³ The site is maintained by Richard Flute who kindly gave permission to use the records.

- Number Used (25L Barrel), potentially referring to amount of foam used: 1-8.

Caveat: It is not known what type of air fighting foam in terms of PFAS composition was used in each incident or each incident type. No information on any environmental measures in the foam-impacted area were given.

8. Locations of high risk landfills. SEPA regulate these sites and make data on the location, size and waste types collected available for each site from 2010 - 2015 (<https://www.sepa.org.uk/environment/waste/waste-data/waste-data-reporting/waste-site-information/waste-sites-and-capacity-excel/>). The list was filtered to include only sites explicitly listed as 'landfill' and locations (as of 2015) with a Pollution Prevention Control permit receiving hazardous (special waste) and non-hazardous (household, industrial, commercial) waste and closed landfill locations. Number of sites: 439.

Caveat: As shown in Appendix III.1.8.2 there is no consistent pattern in the PFAA concentrations and composition in landfill leachate, therefore location of landfill sites is not sufficient information to understand which landfill sites function as direct PFAS sources to groundwater and PWS. For example, among EU Member States PFAA concentrations in landfill leachate range from <1 to 1800 ng/L.

9. Locations of Waste Water Treatment Plants. The OS Mastermap was used to identify locations by searching for 'water' and 'treatment'; having filtered for Scottish locations only. Sites referring to drinking water treatment were filtered out. Number of sites: 1075.

Caveat: The source of data has not been verified by Scottish Water and SEPA. Further, type of wastewater treatment and volume of effluent and sludge produced are unknown. Finally, fate of sludge is also unknown.

10. Locations of monitoring sites of the effluent discharged by WwTW, Combined Sewage Outflows and Leachate collection Works, all of which are regulated by SEPA. Source: SEPA.

11. Locations of businesses/industries potentially using PFAS. The only readily available dataset was found on the Companies House website (http://download.companieshouse.gov.uk/en_output.html). This dataset refers to on-going commercial/manufacturing activity and not to historic activity and lists businesses by Standard Industrial Classification (SIC) code (<http://resources.companieshouse.gov.uk/sic/>). Over 700 individual SIC codes with postcodes were included in the downloaded record, split into a number of broad categories which include agriculture/forestry, manufacturing, construction, wholesale and retail, transportation and storage, accommodation, financial services, and communications. Postcodes were georeferenced and plotted in ArcGIS to allow selection of only Scottish businesses. The list was then filtered to include only business types to address the findings of the literature review in Section 3 and Appendix III. . The types of businesses included in the study are presented in Table AII2. Number of sites: 628.

Caveat: Only the postcode of the listed address of the head office of the company is given, therefore it remains unknown where the environmental impacts of the activity and disposal of PFAS-contaminated waste apply. Further, it remains unknown whether and what types of PFAS substances are indeed used.

Table AII2. Types of businesses whose locations were georeferenced to inform mapping of PFAS risk to PWS.

Finishing of textiles
Manufacture of soft furnishings
Manufacture of woven or tufted carpet and rugs
Manufacture of other technical and industrial textiles
Manufacture of outerwear
Manufacture of workwear
Manufacture of paper and paperboard
Manufacture of dyes and pigments
Manufacture of plastics in primary forms
Manufacture of pesticides and other agrochemical products
Manufacture of paints, varnishes and similar coatings, mastics and sealants
Manufacture of printing ink
Manufacture of soap and detergents
Manufacture of clearing and polishing preparations
Manufacture of glues
Manufacture of basic pharmaceutical products
Manufacture of plastic packing goods
Manufacture of other plastic products
Manufacture of cement
Manufacture of other articles of concrete and plaster
Aluminium production
Treatment and coating of metals
Manufacture of photographic and cinematographic equipment
Specialised cleaning services
Other building and industrial cleaning activities
Washing and dry-cleaning of textile and fur products

APPENDIX III

III.1 GENERAL INFORMATION ON PFAS

PFAS are a family of more than 3000 man-made chemicals that have been extensively used since the 1950s as ingredients or intermediates of surfactants and surface protectors for a wide range of industrial and technological applications and consumer products around the world, including the UK (OECD 2013). On a global level, extensive evidence is available on the type of industries manufacturing and using PFAS and on PFAS concentrations in commercial products and in the environment. The most commonly studied PFAS are perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA); the next most commonly studied are perfluorohexane sulfonic acid (PFHxS) and perfluorononanoic acid (PFNA) (Buck et al 2011). Readers can look into more details in a number of reviews (Buck et al 2011; Knepper and Lange 2012; OECD 2013; Kotthoff et al 2015; UNEP-OECD 2015; Xiao 2017, EFSA 2008) and in regulatory documents related to the implementation and consultations under the Stockholm Convention (see also Section III.2.2).

III.1.1 Synthesis-Production

Manufacturing history could be a key factor underlying the concentrations and occurrence of PFAS in environmental samples. Two major processes exist for the production of PFAS, viz. Simons Electro-Chemical Fluorination (ECF), and telomerisation (TM) (Hekster et al 2003). TM produces primarily or exclusively linear PFAS, whereas ECF produces a mixture of branched and linear isomers⁴. The presence of 6:2 FTSA and 8:2 FTSA in the products indicates use of the TM process (Prevedouros et al 2006). PFAS is still being produced in Germany, Italy and 15 locations in China (Emerging Contaminants 2016).

- PFOS manufacturing was based on the ECF process using 1-octanesulfonyl fluoride and per-fluorooctanesulfonyl fluoride (POSF) as initial products. Since 1949, the company 3M was the major global producer of PFOS, sulfonamide derivatives, N-MeFOSE, NEtFOSE, and other PFOS-related substances (e.g. adipates, phosphate-esters, fatty acid esters, urethanes, copolymers and acrylates) as commercialised products (3M 2000a). The company 3M reported a total POSF's historical production of about 37,809 metric tons from its main production facilities in Decatur, Alabama and Antwerp, Belgium, and Cottage Grove, Minnesota (3M 2006). 3M terminated PFOS manufacture in 2002 but PFOS production grew rapidly in China, which is since the major PFOS manufacturer (Xie et al 2013).
- PFOA was first manufactured in 1947 by the ECF process by the company 3M (Prevedouros et al 2006). The ammonium perfluorooctanoate salt (APFO) is the most widely produced form used as an essential surfactant for the manufacture of fluoropolymers such as polytetrafluoroethylene (PTFE) aka Teflon or C8. The largest historic production sites for APFO through ECF were in the U.S. and Belgium, the next largest in Italy and Japan (EFSA 2008). PFOA emissions from the ECF-based production were mainly to water (3M 2000b). A recent report by WHO refers to two case studies of groundwater contamination with PFOA caused by industrial emissions: one of PFOA in the Veneto Region (Italy); the other caused by a chemical plant of Dupont in the USA (WHO 2016). In 2002, 3M phased out the production of PFOA through the ECF process. Other manufacturers, such as the companies Dupont, Daikin, Clariant, Asahi Glass and Atofina used TM (Emerging contaminants 2016), which produces substances that are PFOA precursors through biotransformation (Prevedouros et al 2006; Buck et al 2011). As of 2018, manufacture and import of PFOA has been phased out in the US as part of the PFOA Stewardship program⁵ but existing stocks of PFOA and some imported articles might still be used (US EPA 2018). PFOA production and use are restricted in the EU under the REACH Regulation (see Section III.2.2).
- As C8 compounds are being phased out, the production of PFAS containing C4 and C6 perfluorinated moieties has increased, and PFASs containing C2 and C3 perfluorinated groups connected by ether linkages are also being manufactured (Wang et al 2015). There is a trend within the PFAS manufacturing industry to change from longer perfluorinated chains to shorter ones (Scheringer et al 2014), as the longer chain substances have attracted attention from both the authorities and the media. The health and environmental effects of using shorter perfluorinated chain substances are, however, still uncertain (Wang et al 2013). It is becoming increasingly clear that chain length does play a significant role in the behaviour of the compound once PFAS are released into the environment.

⁴ The mixture of linear and branched isomers presents challenges in providing an accurate quantification of many PFAS in environmental matrices (Riddell et al 2009). Nevertheless, the study of linear and branched isomers is useful for understanding sources of PFAS (e.g. Benskin et al 2010), because the production of isomers varies by manufacturing process.

⁵ The companies participating in the PFOA Stewardship Program were global companies with business operations in United States and other countries. Under the PFOA Stewardship Program, each of the companies committed to work toward a global phase out of PFOA and related chemicals, both for U.S. operations and for the company's global business (US EPA 2018).

III.1.2 Physiochemical properties.

PFAS are both hydrophobic and lipophobic, highly fire resistant, highly weather and temperature resistant, and resistant to chemical and biological breakdown and some of them such as fluoropolymers have excellent dielectric properties (Kissa 2001; Posner 2012). Their unique properties are attributed to the presence of the perfluoralkyl functional chain and the extreme strength of the carbon-fluoride bond (Kissa 2001; Eschauzier et al 2013). Therefore, PFAS properties render them valuable ingredients for products requiring high versatility, strength, resilience and durability but also persistent in the environment (OECD 2013).

III.1.3 Applications

Historical and ongoing⁶ applications of PFAS encompass uses as precursors or raw materials in firefighting foams, rain clothes, ski waxes, paints, medical implants, non-stick frying pans and food packaging (e.g. Buck et al 2011; OECD 2013). A wide range of industries are involved in the use of PFAS (Figure III.1).



Figure III.1. Industries using PFAS substances. Source: Buck et al 2012; OECD 2013.

III.1.4 Occurrence

Within the past decade, several long-chain perfluoroalkyl acids (PFAA) have been recognised or nominated as persistent, bioaccumulative and toxic (UNEP 2009; 2015; 2017). Many PFAS are also considered as ubiquitous (e.g. Prevedouros et al 2006) and have been detected in:

- Remote regions and the ocean (Dreyer et al 2009).
- Wildlife (e.g. Dietz et al 2008; Reiner et al. 2011).
- Air, rain, snow, soil, sediment, surface water and groundwater (e.g. Kim and Kannan 2007; Ahrens et al 2014).
- Landfill leachate (Hamid et al 2018 and literature cited therein).
- Wastewater (Loos et al 2013 and literature cited therein).
- Food (EFSA 2008).
- Fire-fighting foam (OECD 2013).
- Drinking water (Kabore et al 2018 and literature cited therein; Post et al 2012).
- Human body, with PFAS accumulating in protein-rich blood and related organs and not in fatty tissue (e.g. Post et al 2013; Rapazzo et al 2017 and literature cited therein); PFAS with long carbon chains have been estimated to have half-lives in the body ranging from 2-4 years for PFOA, 5 to 6 years PFOS and 8 to 9 years for PFHxS (ATSDR 2017).

⁶ See Section III.2 for accepted uses and exemptions from restrictions.

It must be noted that sites of manufacture, use and disposal of consumer products containing PFAS are determinants of PFAS occurrence (Prevedouros et al 2006). However, the relative importance of direct (Section III.1.5) and indirect sources (Section III.1.6) vary for the different PFAS and may be different on a local, regional or global scale (Armitage et al 2009; Prevedouros et al 2006).

III.1.5 Direct sources of PFAS to the water environment

PFAS may enter the water environment through the following point and diffuse sources:

- Fugitive emissions⁷ or through waste streams (exhaust gases, trade effluent, sludge, solid wastes) from PFAS manufacturing or industrial user sites (e.g. Prevedouros et al 2006; Dauchy et al 2012a,b;), such as sites with industries mentioned in Figure III.1.
- Volatilization, wash-off or direct use in the environment from products containing PFAS at sites such as fire training sites and military or civil airports where fire-fighting foam and fluorosurfactants are used (UNEP 2009; Clara et al 2008; Gobelius et al 2018).
- Wastes containing PFAS, including use of insufficiently treated sewage sludge used as fertiliser (aka biosolid) (D'eon et al 2009; Wilhelm et al 2008; Lindstrom et al 2011; Gottschall et al 2010; Clarke and Smith 2011 and literature cited therein); leachate from landfills⁸ (Busch et al 2010; Hamid et al 2018); insufficiently treated wastewater effluent (Guo et al 2010; Schultz et al 2006; Chen et al 2018); and urban runoff (Murakami et al 2009; Hamid and Li 2016).

III.1.6 Indirect sources to the water environment

PFAS may enter the water environment and drinking water through the indirect sources, i.e. substances that may biotically or abiotically transform to PFAS and are known as PFAS precursors (Buck et al 2011). The significance of indirect sources can only be assessed after the identification of precursor substances. A way of doing this is through Total Oxidizable Precursor Assays aka TOPA (e.g. Houtz et al 2013; Houtz and Sedlak 2012; Weber et al 2017; Casson and Chiang 2018). TOPA is an "aggressive" chemical process which does not quantify nor identify the structures of individual PFAS precursors nor can it be relied upon to quantify PFAS in a sample or transformations under natural (field conditions). However, it represents a "worst-case" or potential scenario, which may be useful for predicting PFAS in drinking water or generally (Weber et al 2017; Casson and Chiang 2018). For example TOPA has shown that the 6:2 FTSA precursor is primarily transformed into PFBA, PFPeA, and PFHxA during oxidation, while 8:2 FTSA is primarily transformed into PFPeA, PFHxA, PFHpA, and PFOA (Houtz et al 2013; Houtz and Sedlak 2012; Weber et al 2017; Casson and Chiang 2018). FOSA is expected to oxidise into PFOA (Houtz et al 2013).

Aerobic and anaerobic biotransformation of precursor compounds. Biotransformation may explain increase of PFOS and PFOA from influent to effluent and sludge at WWTW as well as from raw to treated drinking water. Examples of biotransformation pathways are given below.

- o In general, perfluoroalkyl acids (PFAA)-precursors include low-molecular weight substances (e.g., fluorotelomer alcohols (FTOHs) or N-alkyl substituted perfluorooctane sulfonamides (FOSAMs), but potentially also high molecular weight commercial polymeric materials and phosphate-surfactants (Trier et al 2011). For example, fluorotelomers—when released into the environment—transform into shorter chain PFAAs (Kim et al 2014).
- o 8:2 fluorotelomer alcohol (FTOH) can transform aerobically (Wang et al 2005) and anaerobically (Li et al 2018) to PFOA and other perfluorocarboxylates (PFCA) containing less than eight carbons (see Appendix I).
- o Fluorotelomer olefins (FTO) degrade completely and rapidly, but are expected to form low yields of PFOA and PFNA and related substances once released to the environment (Young and Mabury 2010).
- o Perfluoroalkane sulfonamido-derivatives (e.g. N-ethyl perfluorooctane sulfonamidoethanol-EtFOSE) degrade to PFOS (Rhoads et al 2008).
- o The FOSEs and FOSA have been shown to transform abiotically to PFOS and PFOA (D'Eon and Mabury, 2007; Martin et al 2006) as well as via biotic transformation to PFOS (Tomy et al 2004)

⁷ Fugitive emissions of gases or vapors from pressurized equipment due to leaks and other unintended or irregular releases of gases, mostly from industrial activities.

⁸ Mass flows from the landfill into the aqueous environment can range between 0.08 and 956mg/day (Busch et al 2010).

- o diPAPS may biodegrade to perfluorooctanoic acid (PFOA) (D'eon and Mabury 2007).
- o PFOSA can transform to PFOS (Rhoads et al 2008).
- o The C4 compounds MeFBSA and MeFBSE could possibly degrade to the C4 homologues of PFCAs and PFSA's (D'eon et al 2006; Martin et al 2006).

Atmospheric degradation of precursor compounds is considered the major source of PFAS pollution in remote areas (Loewen et al 2008).

A recent study by Weber et al (2017) along a transect downgradient of a Aqueous Fire Fighting Foam (AFFF) source site and infiltration bed beneath a WWTW discharge point showed that some PFAS precursors (such as 6:2 and 8:2 FTSA and FOSA) are quite mobile, suggesting that the monitoring of these precursors downgradient of point sources is essential to accurately predict future PFAS concentrations.

III.1.7.1 PFAS transport pathways from direct sources to water resources

The following surface transport pathways and inventories can be differentiated:

- Pathways to surface waters. PFAS may enter rivers, streams, lakes and coastal waters in the form of WWTW effluent, atmospheric deposition, and land runoff from urban and, potentially, farmland⁹ areas, airports, military bases and fire-fighting training areas (Schultz et al 2006; Lindstrom et al 2011; Loos et al 2009).
- Pathways to groundwater. PFAS may contaminate groundwater through: leaching from farmland treated with wastewater and biosolid (Skutlarek et al 2006 ; Lindstrom et al 2011); infiltration of contaminated surface waters (e.g. Weber et al 2017); leaching from landfills (Hamid et al 2018); and leaching from sites where fire-fighting foams have been applied (e.g. Houtz et al 2016; Weber et al 2017).
- Pathways to remote environments. PFAS may reach areas away from human activity through long-range atmospheric and oceanic currents transport (Dreyer et al 2009).

III.1.7.2 PFAS sub-surface sources, transport pathways and sinks

The unsaturated zones beneath AFFF-impacted land and the infiltration beds are believed to be continuing sources of PFAS to the aquifer decades after source removal (Weber et al 2017). Groundwater is the main freshwater sink for PFOA and PFOS (Zareitalabad et al 2013) and potentially for other PFAS. A review of PFAS concentrations and composition in a wide range of environmental media showed that groundwater was the only aquatic matrix where no PFAS with a carbon chain longer than ten was reported (Rayne and Forest 2009). However, there is limited knowledge of the complex processes controlling PFAS sub-surface fate and transport. The evidence found refers to the following key factors influencing sub-surface pathways:

1. PFAS chain length: several studies show that leaching potential of PFAS decreases with increasing chain length. For example:
 - (i) Sepulvado et al (2011) detected short-chained PFAS in soil cores from biosolids-amended soils to depths of 120 cm, suggesting transport of these substances within the soil profile over time and a higher transport potential for short-chain PFCs in soils amended with municipal biosolids.
 - (ii) Higgins and Luthy (2006) estimated in laboratory experiments that PFAS distribution coefficients in organic sediments (K_{oc}^{10}) generally increase by between 0.3 and 0.6 log units with each additional CF_2 unit (moiety).
2. Sorption- and desorption potentials of the different PFAS: laboratory experiments and field studies show that short-chained PFAS are more likely to be found in aqueous matrices (e.g. soil leachate) whereas long chain and more lipophilic PFAS are predominantly found adsorbed on soil matrices. For example:
 - (i) Perfluorinated carboxylic acids are usually found as dissociated anions in environmental media and do not sorb substantially on sediment particles (e.g. Vierke et al 2013).

⁹ where PFAS contaminated biosolid has been applied.

¹⁰ The soil organic carbon-water partitioning coefficient is the ratio of the mass of a chemical adsorbed in the soil per unit mass of organic carbon in the soil per the equilibrium chemical concentration in solution. It is the "distribution coefficient" (K_d) normalized to total organic carbon content. KOC values are useful in predicting the mobility of organic soil contaminants; higher KOC values correlate to less mobile organic chemicals while lower KOC values correlate to more mobile organic chemicals. K_d (L/kg) is the ratio of a chemical's sorbed concentration (mg/kg) to the dissolved concentration (mg/L) at equilibrium, as illustrated below: $K_d(L/kg) = \text{Sorbed Concentration (mg/kg)} / \text{Dissolved Concentration (mg/L)}$. Source: State of Washington (n.d. and literature cited therein).

- (ii) Higgins and Luthy (2006) showed that sorption of perfluorinated carboxylic acids to organic particles increases with the length of the perfluoroalkyl chain but it is still orders of magnitude lower than usually observed for typical POPs, which may explain the long transport of PFAS in aquatic media particularly in the ocean.
 - (iii) At least two studies based on laboratory experiments showed that perfluorinated sulfonic acids adsorb more than perfluorinated carboxylic acids (Higgins and Luthy 2006; Enevoldsen et al 2010).
 - (iv) McGuire et al (2014) suggested that higher perfluorinated carboxylate log K_{oc} values may be due in part to carboxylate precursor transformation during the sediment extraction process. Other field studies, however, showed the opposite pattern (Kwadijk et al 2010).
 - (v) Laboratory experiments by Enevoldsen et al (2013) showed that in soils where desorption of PFAS substances is lower than adsorption, soil can act as a protective barrier towards groundwater.
 - (vi) Leaching experiments performed by Gellrich et al (2012a;b) showed that the short chain PFAS elute without adsorbing on solid but PFAS_{>C8} could not be detected in the soil leachate after 2 years. These experiments also demonstrated that larger and more lipophilic molecules (e.g. PFHxA and PFHxS) can displace shorter PFAS (e.g. PFBA) from their binding sites in the soil.
 - (vii) Laboratory experiments by Ahrens et al (2011) found that PFOA was present mainly in the dissolved phase at low suspended particulate matter concentrations in rivers.
3. Groundwater flow and velocity (hydraulics), which are key determinants of the time between introduction of PFAS to the ground and the distance travelled by these substances. The evidence base on the influence of groundwater flow and velocity on PFAS transport comes exclusively from a field study by Weber et al (2017) at Cape Cod Massachusetts. This study showed that:
- (i) With a groundwater velocity of 0.42m/day and a log K_{oc}=3.37, it could take PFOS 15 years to travel a distance of 780 m from the site of introduction into the groundwater beneath a AFFF source site.
 - (ii) Knowing the last year the PFAS was emitted from the source site, allows for estimating the time taken for PFAS to travel in the unsaturated soil zone. This time may be between 3 and 30 years for AFFF-impacted sites depending on K_{oc}, unless there occurs precursor transformation.
 - (iii) PFOA, PFNA, and PFAAs with C4–C7 chain lengths are predicted to be transported farther than PFOS based on lower in situ K_{oc} values. This is differential transport. For example, PFOS was detected in groundwater 4km downgradient of a AFFF source site; however, greatest PFOS concentrations were recorded beneath source sites (e.g. AFFF-impacted land and infiltration beds from a WwTW discharges). Highest PFOA was detected approximately 960 m downgradient a AFFF source site in response to geochemical conditions. PFHxS were greatest in the groundwater near a AFFF source site and shallow groundwater. PFHpA, PFHxA, and PFPeA were detected deeper in groundwater and farther downgradient, indicating that shorter chain length PFAS are more mobile than PFOS both vertically and horizontally.
4. Soil and groundwater geochemistry.
- (i) Hydrophobic interactions between the organic carbon content of the sediment and the PFAS chain and electrostatic interactions affected by the PFAS type determine the rate of PFAS sorption on soil particles, which is a slow process especially at higher soil pH and low soil calcium concentrations (Higgins and Luthy 2006; Johnson et al 2007; Tang et al 2007).
 - (ii) A study found that increased sediment organic carbon and ionic interactions resulting from wastewater loading in the groundwater beneath aWwTW discharge point may enhance PFAS sorption beneath the infiltration beds through both hydrophobic and electrostatic interactions (Weber et al 2017). However, PFAS remobilisation may be observed following cessation of disposal, owing to (1) reduction in dissolved ion concentrations; (2) desorption of sediment organic carbon; and (3) dissolution of positively charged iron and manganese oxide grain coatings following the onset of anoxic conditions beneath the infiltration beds. Weber et al (2017) also suggested that anoxic conditions caused by wastewater discharges may influence biotic precursor transformation.

III.1.8 PFAS Source Sites (also known as hotspots)

Elevated PFAS concentrations in groundwater (and surface water) have been observed near the so called “PFAS hotspots”, which refer to sites with detectable and elevated PFAS concentrations and associated with direct sources of PFAS to the water environment (see review by Cousins et al 2016; Houtz et al 2016; Hu et al 2016; WHO 2016; Gobelius 2018):

- Chemical plants
- Landfills
- Waste water treatment works (WWTW)
- Domestic and military airports
- Oil, gas and mining production sites
- Firefighter training areas

Point sources such as chemical plants, landfills, airports and oil, gas and mining production sites function as PFAS source sites due to the fugitive emissions or production of waste streams whereas firefighter training areas and, potentially landfills, function as source sites through land runoff and leaching. It must be noted that the literature searches delivered only two studies assessing the PFAS risk to drinking water supplies (Section III.1.8.1).

To better understand the risk to drinking water from these sources sites, the sections below review types of PFAS substances and concentrations in landfills (Section III.1.8.2), firefighting foams (Section III.1.8.3), and in WwTW effluent and sludge/biosolid (Section III.1.8.4).

III.1.8.1 Monitoring to assess PFAS to drinking water supplies

- Hu et al (2016) developed a statistical framework for investigating whether increased PFAS concentrations in drinking water are associated with the number of point sources within a catchment at the scale of a waterbody. They used a multivariate spatial regression model for waterbodies with detectable PFAS that adjusts for correlations and co-location among point sources; a natural log transformation was used to normalize the distribution of individual PFAS. The study used publicly available drinking water concentration data for six PFAS from the US EPA's third Unregulated Contaminant Monitoring Rule (UCMR3), including perfluorobutanesulfonic acid (PFBS), perfluorohexanesulfonic acid (PFHxS), perfluoroheptanoic acid (PFHpA), PFOA, perfluorooctanesulfonic acid (PFOS), and perfluorononanoic acid (PFNA). The use of the UCMR3 database allowed Hu et al (2016) to discuss its utility for identifying sources of PFAS to US drinking water supplies, locations of vulnerable populations, and priorities for future monitoring.
- WHO (2016) described the hydrogeological study that was carried out to investigate the distribution of groundwater and the extension and flow velocity of the PFAS contamination plume caused by a PFOA manufacturing plant in the Region of Veneto, Italy. These data allowed for estimations of the duration of contamination, which dated back more than 20 years. Soil monitoring showed a widespread presence of PFAS, not only in areas where human activity or erosion could have produced contamination, but also in those assumed to be free of it.

III.1.8.2 Landfills

Every-day consumer products and packaging containing PFAS are sent to landfills at the end of their life cycle. Recent studies on the PFAS content of every-day consumer products used in Germany, Norway, USA and China (hence potentially in other countries too, including the UK) showed the presence of individual PFAS in varying concentrations (Kotthoff et al 2015; Herzke et al 2012; Yuan et al 2016). Samples of carpets, outdoor clothing, leather and AFFF used in Germany as well as other products such as paint, non-stick ware, water proofing agents, ski waxes, leather care sprays and some cleaning agents used in Germany and Norway contained PFOA, FTOH, FTS, PFOSA, PFBS, PFHxS, PFBA, and PFPA at concentrations above limit of quantification (LOQ) (Kotthoff et al 2015; Herzke et al 2012). Another study found that eco-friendly paper tableware and popcorn bags used in USA and China contained very high concentrations of total FTOH (Yuan et al 2016).

Up to 70 different substances from all classes of PFAS have been detected in landfill leachate in North America, Europe, China and Australia, as shown in a recent comprehensive review of studies conducted until 2017 (Hamid et al 2018 and literature cited therein). Despite the great variation in individual PFAS concentrations between countries and types of landfills within the same country, some patterns in PFAS composition of leachate were observed in the available literature (Hamid et al 2018 and literature cited therein):

- Shorter-chain perfluoroalkyl acids (PFAA) ($C \leq 6$) are generally more abundant than longer-chain homologues, with perfluorobutanoic acid (PFBA), perfluorobutane sulfonate (PFBS) and perfluorohexane sulfonic acid (PFHxS) exhibiting the highest concentrations. This was explained as preferential release and leaching of short-chain PFAA from solid waste, consistent with their higher water solubilities and lower organic carbon-water partition coefficients relative to longer-chain homologues. Potentially, it also reflects the industrial and regulatory shift from PFOS use towards shorter-chain compounds since 2000¹¹.
- Total perfluoroalkyl carboxylic acids (PFCA) dominate the total PFAS concentration in the USA, Canada, Australia and may exceed 5000ng/L but there is great variation in concentrations. The difference in concentrations could result from variation in waste composition, climatic conditions, age of the landfill and/or leachate management system (i.e. leachate recirculation vs flow-through).
- Among European Union Member States¹², no consistent pattern in the PFAA concentrations in landfill leachate could be observed except that PFOS and PFOA have higher levels than short-chain PFAA. Concentrations reported were in the range of <1 -1800 ng/L. However, some landfills showed a predominance of short-chain PFAA in Germany and in Spain.
- Despite its restricted use, perfluorooctanoic acid (PFOA) remains one of the most abundant substances in landfill leachates presumably because of the high frequency of PFOA detection in consumer products and its historical use in leather, textiles, paper and electronics industries.
- PFAS-precursors (e.g. fluorotelomer-based compounds) were also abundant in landfill leachates at concentrations comparable to, or higher than, the most frequently detected PFAS.
- The total concentration of perfluoroalkyl acids (PFAA) in the leachate from closed landfills (20 to 40 years ago) ranges from hundreds to a few thousands of ng/L.
- Aerobic and anaerobic biotransformation processes control the release of PFAA from solid waste to leachate in landfills¹³.
- Landfills emit elevated concentrations of PFAS to the ambient air such as fluorotelomer alcohols (FTOH), suggesting that landfills are potential sources of atmospheric PFAS.

III.1.8.3 Fire-fighting foams

PFAS are used in small amounts to produce various fire-fighting foams such as aqueous film-forming foams (AFFFs), alcohol-resistant aqueous film-forming foams (AR-AFFF), fluoroprotein foam (FP) and film forming fluoroprotein (FFFP) (OECD 2013). According to the Fire Fighting Foam Coalition all modern AFFF agents contain fluorotelomer-based fluorosurfactants, which do not contain or break down into PFOS or PFHxS (FFFC 2017). The composition of AFFF-impacted groundwater and surface water depends on both historic and current uses of AFFFs. Historically, longer chain PFAS such as PFOS and PFOA and PFAS precursors were used as the main components in AFFF; today, C8 PFAS have been replaced by C6 or shorter PFAS although AFFFs may contain trace quantities of PFOA or PFOA-based substances as an unintended by-product of the surfactant manufacturing process (FFFC 2017). Change in geochemical conditions in the unsaturated zone of aquifers or in rivers may lead to release of old PFOS that was immobilised (adsorbed) to particles in soil and sediment around fire training sites, airports or military bases and generally AFFF-impacted areas (Hedlund 2016; Weber et al 2017; Dauchy et al 2017).

AFFF are released into the environment during fire-related emergencies or training: civilian airports, military fire training areas and firefighter training sites have been found to be PFAS source sites (Hu et al 2016; Houtz et al 2013; Ahrens et al 2015; Banzhaff et al 2017). Studies at sites where AFFF have been used, show that PFAS concentrations in the groundwater and surface waters surrounding these sites are significantly higher compared to non AFFF-impacted sites (Ahrens et al 2015; Houtz et al 2013; Filipovic et al 2015; Dauchy et al 2017), dropping with increasing distance from the source, probably due to dilution and partitioning in different media (Ahrens et al 2015).

Baduel et al (2015) and Weber et al (2017) showed that AFFFs usage at fire training sites can be a long term source even after the sites have been closed for fire training. Likewise, Moody et al (2003) demonstrated that groundwater from wells near a

¹¹ For example, 3M has commercialized surface treatment products containing C4-based side-chain fluorinated polymers since 2003 (Wang et al 2013).

¹² Leachate in European landfills has been studied in: Spain (Fuertes et al 2017), Finland (Perkola and Sainio 2013), Norway (Kallenborn et al 2004), Sweden (Woldegiorgis et al 2006), Denmark (Bossi et al 2008), and Germany (Busch et al 2010).

¹³ Several studies have found that the presence of precursor substances for PFAA and changes in leachate pH, EC, and 24-h precipitation are important factors controlling PFAS occurrence in leachate (e.g. Liu and Avendano 2013; Benskin et al 2012).

decommissioned airforce base in Michigan contained PFOS, PFHxS, PFOA, and PFHxA ranging in concentration from 3 to 120 µg /l five or more years after the last known use of AFFF onsite. Several studies report the presence of 6:2 and 8:2 FTSA in the groundwater and surface water close to AFFF-impacted sites (Houtz et al 2016; Dauchy et al 2017; Hedlund 2016; Banzhaff et al 2017; Weber et al 2017). Hedlund 2016 analysed both surface -and groundwater samples close to fire training sites across Sweden and found relatively high fractions of 6:2 FTSA, which is contained in fluorotelomer-based AFFF.

It must be noted that a recent study by Barzen–Hanson et al (2017) identified 40 new PFAS classes and 17 already reported PFAS classes in AFFF and AFFF-impacted groundwater in the vicinity of 15 US military bases. Of the newly discovered PFAS substances found only in AFFF-impacted groundwater, 11 are ECF-derived and two are FT-derived, which suggests that both ECF- and FT-based PFAS in fire-fighting foams are persistent in the environment (Barzen-Hanson et al 2017).

III.1.8.4 Waste Water Treatment Works (WwTW): effluent, sludge and biosolid

The discharge of municipal (treated) wastewaters is one of the principal routes of entry of PFAS into the aquatic environment. The number of WwTW in a catchment is positively associated with the presence of PFAS and particularly PFOS and PFOA in drinking water, although less important than industrial sites and military fire training sites (Hu et al 2016; Sinclair and Kannan 2006; Valsecchi et al 2015). Several studies have shown that the PFAS are non-reactive to typical wastewater (and drinking water) treatment processes, and only long-chain PFAS are effectively removed by sorption (Schultz et al 2006; Appleman et al 2014; Guo et al 2010). Generally, PFOA is fully discharged as effluent into receiving rivers, while about half of PFOS is retained in the sewage sludge (Becker et al 2008, 2010; Guo et al 2010; Huset et al 2008).

Generally, PFOS and PFOA remain the main forms of PFAS in municipal WwTW effluent five to ten years after the major phase-outs of C8-based PFAS commenced in the USA (Schultz et al 2006; Ahrens et al 2009; Guo et al 2010; Klosterhaus et al 2013). C8- based PFAS can undergo transformation during treatment processes , but their products are often other measureable PFAS that contain a similarly sized or shorter carbon chain PFAS substances, which may be PFOS or PFOA precursors (Rhoads et al 2008; Schultz et al 2006; Guo et al 2010). Thus, some PFAS substances measured in effluent may have lower concentrations in influent than in the effluent as a result of biotransformation of precursors during the activated sludge process (Guo et al 2010; Houtz et al 2016).

Effluent. PFAS in WwTW effluent may reflect current use products, along with PFAS in historical products still in use or slowly released into wastewater and different sources.

- Houtz et al 2016 showed that AFFF may be a primary source of PFAS contamination to effluent receiving influent from airports, and that PFAS in AFFF-impacted wastewater may occur at much higher levels than in typical municipal wastewater. Airports use AFFF in emergencies and for periodic proficiency testing. As Houtz et al 2016 noted, in the USA when AFFF is employed for emergencies in airports or military bases, runoff from the area of usage is typically diverted and collected for off-site treatment and disposal or diverted to contribute to industrial trade effluent.
- Guo et al (2010) showed that in South Korea domestic and industrial WwTW discharged treated effluent with more PFAS from plants located in big cities than those located in rural areas, suggesting household sewage is one of the significant sources of PFAS contamination in the environment. No PFAS were detected in livestock wastewater, indicating that livestock WwTW were not the source of PFAS to natural/remote water bodies. The highest concentrations of PFOA (615 ng/L) were detected in the influent from paper and battery manufacturers and relatively high concentrations (i.e. between 10 and 100ng/L) for PFOS, PFHpA and PFNA were measured in the effluent from paper, textile and leather industries. These findings were considered to be consistent with the historic and ongoing use of these PFAS as a surface coating for paper, carpets and furniture products
- Boulanger et al (2005) studied the composition, biotransformation and origin of eight PFAS surfactants in WwTW effluent from both domestic and industrial sources in Iowa, USA. PFOS and PFOA were above LOD in effluent and river water near discharge points; N-EtFOSA was detected in influent, effluent and river water. The authors found that transformation of precursors within wastewater treatment is not an important source of PFOS and PFOA compared to direct use and disposal of products containing the end products as residual amounts.
- A study of several small rivers in Germany concluded that the majority of PFAS entered the rivers via municipal WwTW (Becker et al 2008).
- Schultz et al (2006) determined the mass flows of selected PFAS in the effluent from a municipal wastewater treatment plant serving a population of approximately 50,000 people in the Pacific Northwest, USA and found that mass flows of the 6:2 fluorotelomer sulfonic acid (6:2 FTSA) and PFOA were unchanged as a result of wastewater treatment, which indicates that conventional wastewater treatment is not effective for removal of these compounds. They also found that PFHxS, and

perfluorodecanoate (PFDA) decreased during trickling filtration and primary clarification, while activated sludge treatment decreased the mass flow of PFHxA. On the other hand, perfluorodecane sulfonate (PFDS), PFOS and PFOA increased during activated sludge treatment, potentially as a result of transformation of precursor molecules.

- Ahrens et al 2009 studied the effluent of 10 WwTW in the River Elbe to the German Bight, six of which received only domestic influent (population served=14,000 -150,000 people) and four received mixed domestic and industrial/commercial influent (population served=25,000 to 2.2 million people. Total PFAS concentration in the effluent from WWTP receiving domestic and industrial influent was more than 4 times higher (171ng/L) than the effluents of WwTW without industrial waste water parts (36 ng/L). PFBS and PFHxS were detected in all WwTW effluents. PFOS was not detected in the effluent of domestic WWTP serving <20,000 people but reached 82.2ng/L in the effluent from mixed WwTW serving 650,000 people. Likewise 6:2 FTS had high concentration (32.8 ng/L) in the effluent from a mixed WwTW serving 2.2M people. C5–C10 PFCAs were found in all WwTW effluents, with PFOA being the predominant substance (concentration range: 12.3-77.6 ng/L). Potential precursors, PFOSA, MeFBSA and MeFBSE, were found in all WwTW. total PFAS concentrations in the WwTW effluents (mean=99ng/L) was approximately 5–10 times higher than the total PFAS concentrations of the river water (mean=19 ng/L), indicating that WwTW are potential sources of PFAS in the freshwater and the marine environment.
- Appleman et al 2014 evaluated the PFAS removal efficiency of water treatment techniques for secondary and a tertiary wastewater treatment systems producing effluent reused for other purposes including potable uses. Each wastewater treatment plant was sampled twice in 2011/12. The study demonstrated variable but limited removal efficiency for the following substances after secondary and tertiary treatment (in parenthesis, range of concentrations after treatment in ng/L): PFBA (< 5.0 – 29); PFHxA (10.0 – 90); PFPeA (9.4 – 370); PFHpA (2.6 – 18); PFOA (9.3 – 220); PFNA (3.6 – 11); PFDA (1.5 – 82); PFUnA (< 0.50 - 1.4); PFDaA (< 0.25 - 2.7); PFBS (1.7 - 8.6); PFHxS (2.7 – 10); PFOS (4.3 – 17). However, reverse osmosis, one of the costliest treatment approaches, reduced all PFAS in the treated effluent below detection level.
- Chen et al (2017) studied 17 PFASs in influents, effluents, and activated sludge solutions from municipal waste water treatment plants along the Yanghe River, China, a country where PFOS and PFOA are still (as of 2018) manufactured. The substances PFBA, PFOA, PFHxA, PFPeA, PFBS and PFOS) were detected in influents and effluents in comparable concentrations indicating that WwTW do not efficiently remove PFASs from wastewater. PFAS removal efficiencies differed among WwTW and were influenced primarily by the treatment process used at each, and followed the order: Cyclic Activated Sludge System (CASS, 32.2%) > Orbal Oxidation Ditch (OD, 17.5%) > Anaerobic–Anoxic–Oxic (A2/O, –1.49%). CASS removed more efficiently short-chain PFAS compared to the other systems.

Biosolids. Several studies have shown that sludge (biosolids) from WwTW may contain significant amounts of PFOS and PFOA and their precursors and significantly contaminate water resources. Sewage sludge concentrations have mainly been reported in the US, largely a consequence of the analytical difficulties associated with quantification of PFAS in sewage sludge matrices (Clarke and Smith 2011 and literature cited therein). Based on US studies, PFOS and PFOA are the predominant PFAS in sludge: PFOS concentration ranges between 0.008 and 3.12 mg/kg/dry weight(dw); and PFOA concentration ranges between 0.002 and 0.244 mg/kg/dw (3M Environmental Laboratory 2001 cited in Clarke and Smith 2011; Sinclair and Kannan 2006; Loganathan et al 2007). In the USA, no differences in PFAS levels were apparent in sludges from urban and rural WwTW, however, major seasonal variations in concentrations were observed (Loganathan et al 2007). A study from Denmark showed that the range for PFOS in sludge (0.005 to 3.12 mg/kg/dw) was comparable with that in the US but the range for PFOA (0.001 to 0.02 mg/kg/dw) was much lower (Bossi et al 2008).

Field investigations demonstrated that PFOA in biosolids applied to agricultural land increase PFOA concentrations in tile drainage above detection limits (Gottschal et al 2010). Lindstrom et al 2011 estimated that land application of approximately half of the biosolids generated by WwTW may contribute to human exposure through subsequent contamination of water, food, livestock, and wildlife.

Biosolids application resulted in one of the largest PFAS drinking water contamination events in Europe (Wilhelm et al 2008). In this particular case, industrial waste with high concentrations of PFAS was manufactured into a soil improver by a recycling company and spread by farmers on agricultural land of the rural area Sauerland, Germany and led to substantial environmental pollution. In parts of the affected area, perfluorooctanoic acid (PFOA) concentrations in drinking water were >500ng/L (Wilhelm et al 2008).

Sepulveda et al (2011) reported levels of PFAs in agricultural soils amended with typical municipal biosolids: PFOS was the

dominant PFA in both biosolids (80-219 ng/g) and biosolids-amended soil (2-483 ng/g). Concentrations of all PFAS in soil increased linearly with increasing biosolids loading rate. More recently, Rainey (2017) reported average concentrations of individual PFAS in biosolids based on NEBRA's¹⁴ monitoring of PFAS risk from biosolids in 27 sampling points in the NE USA. The highest average concentrations were recorded for PFBA (34µg/kg), PFOS (34µg/kg), PFPeA (22.5µg/kg), PFHxS (13.3µg/kg) and for PFHeA (11µg/kg) (Rainey 2017).

III.1.9 Exposure to PFAS-contaminated drinking water - Health effects of PFAS exposure.

Studies on the adverse effects PFAS exposure to humans refer mainly to the relationship of PFAS serum levels and a disease or a biochemical indicator of a disease (e.g. cholesterol). A review of studies on the effects of PFAS on human health by Lei et al (2015) showed that a clear cause-effect relationship between exposure to PFAS and cancer and human metabolic diseases has not been established yet. However, PFAS in serum have been found to be related to reproductive dysfunction and thyroid disease in the general population (Lei et al 2015 and literature cited therein).

With regards to exposure to PFAS drinking water, it is not well understood what level of PFAS concentration in drinking water can be considered as the effect dose. Nevertheless, positive correlations have been found between PFAS levels in human serum and the consumption of PFAS contaminated drinking water (Emmett et al 2006; Hölzer et al 2008; Steenland et al 2009). For PFOA in particular, a positive linear relationship has been reported between drinking water levels and human serum concentrations in populations served by public water supplies (Post et al 2009) and in communities served by private wells (Hoffman et al 2010). Post et al (2012) reviewed available information on PFOA, its sources and occurrence in drinking water, toxicokinetics, and health effects. Information covered in their review "suggests that the continued human exposure to even low concentrations of PFOA in drinking water results in elevated body burdens that may increase the risk of health effects."

Individual PFAS health effects. The US Agency for Toxic Substances and Disease Registry¹⁵ (ATSDR 2018a), reports adverse effects of PFOS, PFOA, PFHxS, and PFNA on: growth, learning, and behaviour of infants and older children; a woman's chance of getting pregnant; body's natural hormones; cholesterol levels (increase); the immune system; and on the risk of cancer (increase). The 'long-chain' PFASs and PFCAs and their corresponding anions have been shown to be more bioaccumulative than their short-chain analogues (Martin et al. 2003a, 2003b; Butt et al 2007; Conder et al. 2008). Adverse human health effects of PFOS exposure may be also related to PFOS precursors together with demonstrated or potential environmental concentrations that may exceed the effect dose for certain higher trophic level biota such as fish (UNEP/POPS/POPRC.2/17/Add.5 2006; EFSA 2008). PFOA exposure has been associated with numerous health endpoints in the general population, communities with contaminated drinking water, and workers, such as bladder and kidney cancers, cardio-and cerebro-vascular disease, diabetes, endocrine disruption, thyroid disease, and decreased fetal growth (see reviews by Post et al 2012 and UNEP/POPS/POPRC.11/5 2015 and literature cited therein). Lastly, a wide range of studies demonstrated adverse health effects of PFHxS exposure on the nervous system and brain development, the endocrine system, including in particular the thyroid hormone system; cholesterol levels, causing increase; metabolism; and on children exposed to the substance prenatally (UNEP/POPS/POPRC.13/4 2017 and literature cited therein). However, little is known about the combined toxicity of PFAS exposure.

III.2 POLICY FRAMEWORK

Developing and implementing evidence-based approaches to PFAS risk reduction is one of the greatest challenges faced by governments, the industry, scientists and advocacy groups alike. A recent survey by UNEP-OECD (2015) in a number of countries around the world including the EU, showed that the major challenges as regards PFAS reduction emerge from lack of (i) robust scientific evidence on the PFAS content of industrial and commercial products and of PFAS levels in the environment and humans in relation to PFAS emissions; (ii) cost-effective and technically suitable alternative materials; and (iii) cost-effective and available technologies to dispose and/or destroy PFAS-contaminated materials. For PFAS substances such as PFOS and PFOA, robust evidence is available and this helped to lay down restrictions of use, production and emissions on international and national level. Here, the key policy frameworks underpinning PWS regulation in Scotland and PFAS control outwith the DWD (See Section 1: Introduction) are reviewed to substantiate the assessment of risk from current (ongoing) and historic direct

¹⁴ NEBRA (Northeast Biosolids and Residuals Association) is a non-profit professional association advancing the environmentally sound and publically supported recycling of biosolids in New England, New York and eastern Canada.

¹⁵ ATSDR is involved in epidemiological studies at several PFAS-contaminated sites across the USA, either directly or through assisting state and federal partners. As of now, most sites are related to drinking water contamination connected with PFAS production facilities or fire training areas where aqueous film-forming firefighting foam (AFFF) was regularly used (ATSDR 2018b).

and indirect sources of PFAS in Scotland. It must be borne in mind that an extensive review of the governance and regulation of PFAS is beyond the scope of this report.

III.2.1 Background on the regulation of water quality in PWS in Scotland

Approximately 3.5 % of the population in Scotland is served by PWS; however, much larger numbers of the public may use premises with a PWS in the course of leisure activities (DWQR 2015). The Private Water Supply (Scotland) Regulations 2006 as amended in 2017 (hereafter reported as The Regulations) transpose the DWD requirements in the PWS context. The Regulations identify two types of supplies. Type A-PWS are defined as those serving more than 50 people or a commercial or public use. Type B-PWS are those serving fewer than 50 people. The Regulations place a duty on local authorities to monitor each Type A-PWS at specified minimum frequencies in line with Annex II of the DWD, and carry out risk assessments. Local authorities must also test water quality in Type B-PWS upon request by their owners or users.

Monitoring. This is crucial in ensuring provision of safe and wholesome water to PWS users in accordance with the standards set for each parameter in Annex I of the DWD. The Regulations require that the Drinking Water Quality Regulator (DWQR) reports the results of PWS monitoring, checks that local authorities are applying monitoring and risk assessments in their area, and provides guidance and clarification to local authorities. Currently, Part C of Schedule 3 of The Regulations mentions that the minimum sampling frequency for a parameter may be reduced under paragraph 3(b)(ii) only if the risk assessment confirms that no factor (that can be reasonably anticipated) is likely to cause deterioration of the quality of the water. Further, a local authority can remove a parameter from the list of parameters to be monitored (under paragraph 3(b)(iii)) only if the risk assessment confirms that no factor that can be reasonably anticipated is likely to cause deterioration of the quality of the water.

Risk assessment. Part 3 of The Regulations requires that the risk assessment must seek to establish whether (i) the measures in place to control risks to human health from the catchment area through abstraction, treatment and storage to distribution are working effectively, and (ii) water at tap meets the water quality standards specified in Article 14 of Part 4¹⁶. In addition, the risk assessment must assess available information to establish whether water quality standards are met and the parametric values for indicator parameters are not being exceeded.

III.2.2 The Stockholm Convention

The Stockholm Convention on Persistent Organic Pollutants (POPS) regulates use of pollutants that meet the following screening criteria: persistence, bioaccumulation, long-range transport and adverse effects/toxicity. As of 2018, the Convention prescribes restrictions for the use of PFOS its salts and related substances, which are in place since 2009 (UNEP/POPS/COP.4/38 2008, Decision SC-4/17). Figure III.2a shows the PFOS related substances and their applications; Figure III.2b shows the accepted PFOS purposes and exemptions from restrictions under the Convention. It must be noted that proposals are in place for adding in the list of POPs regulated by the Convention two more groups of PFAS substances: PFOA, its salts and related substances (UNEP/POPS/POPRC.11/5 2015) which are already regulated under the EU REACH Regulation (see below); and perfluorohexane sulfonic acid (PFHxS), its salts and related substances (UNEP/POPS/POPRC.13/4 2017). Bitesize information for the range of PFHxS substances and their applications nominated under the Convention is given in Box III.1.

¹⁶ These standards refer to the following. Firstly, tap water must not contain: (a) a microorganism, parasite or substance which (in number or concentration) poses a potential danger to human health; or (b) a microbiological or chemical parameter which exceeds its parametric value. Secondly, at the point of compliance, the sum of the following must not exceed 1 mg/l: (a) nitrate (in mg/l) in the water, divided by 50; plus (b) nitrite (in mg/l) in the water, divided by 3.

Box III.1. Bitesize information on the types of PFHxS substances proposed to be restricted under the Stockholm Convention. Source: UNEP/POPS/POPRC.13/4 2017 and literature cited therein.

- PFHxS is a member of the same chemical category as perfluorooctane sulfonic acid (PFOS).
- 48 PFHxS-related/precursor/polymer substances which all contain the fluorinated alkyl moiety C₆F₁₃SO₂ have been identified.
- Used as: surfactants; to make fluoropolymers; water- and stain protective coatings for carpets, paper and textiles; and as raw materials to produce PFAS based products such as surfactant and surface protection products. but are also unintentionally produced during industry processes
- Present in: some fire-fighting foams (as building blocks), paper, water proofing agents; and textiles treatments and other products.
- PFHxS, its salts and related substances are also unintentionally produced during industry processes.
- Examples of PFHxS, its salts and related substances are:

Perfluorohexane sulfonic acid (PFHxS)

Perfluorohexane sulfphonamide

PFHxS Salts: Perfluorohexanesulfonate ammonium, Perfluorohexanesulfonate potassium,

Potassium N-ethyl-N[(tridecafluorohexyl)sulfonyl]

Perfluorohexane sulfonyl fluoride(PFHxSF) used as precursor/start material of PFHxS.

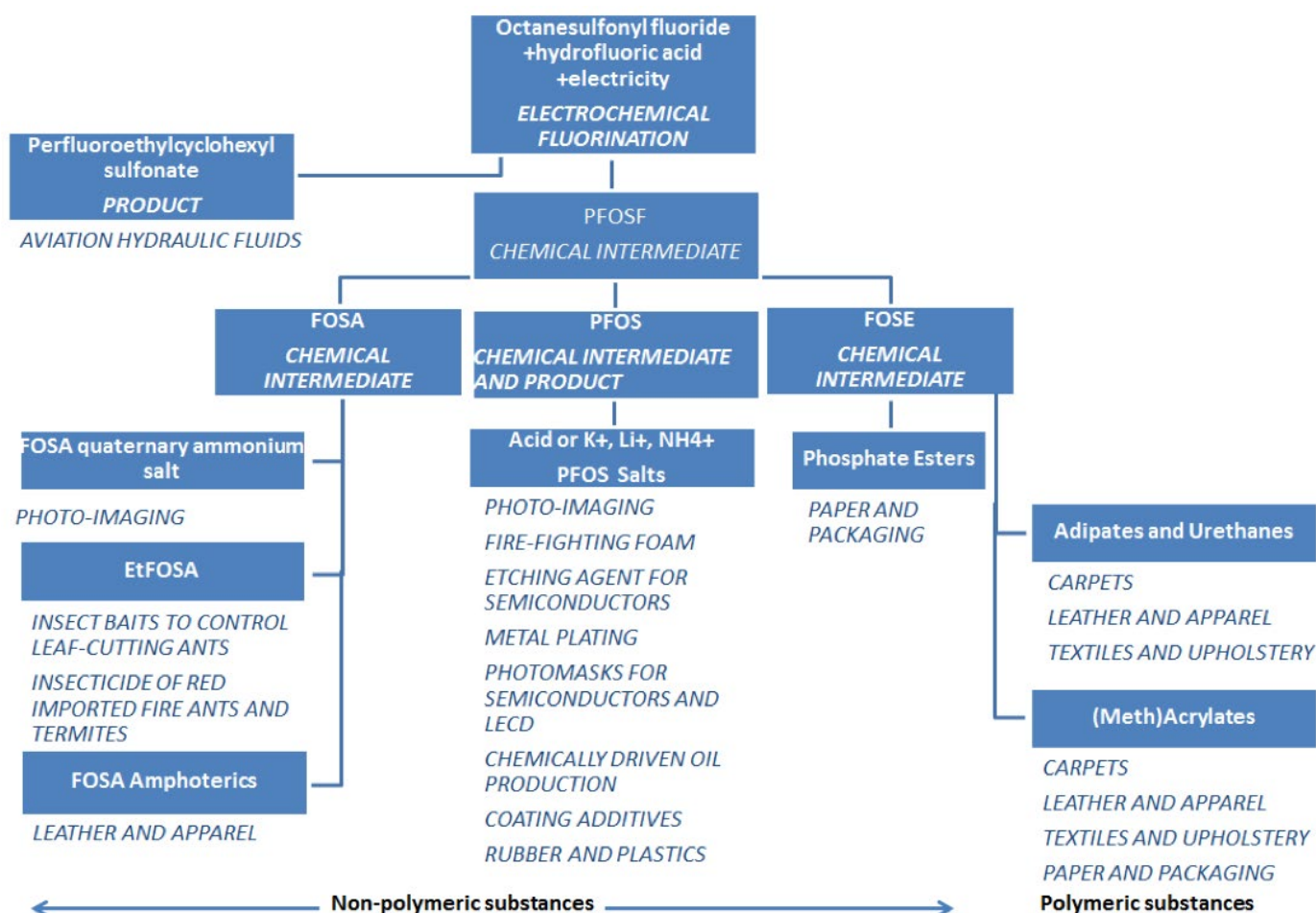


Figure III.2a. Process scheme and major product categories and applications of PFOSF, PFOS and PFOS-related substances, such as FOSA and FOSE and the non-polymeric and polymeric derivatives. Source: UNEP/POPS/COP.4/38 2008.

Acceptable purposes under the Stockholm Convention	Exempt uses under the Stockholm Convention	Exempt uses under Regulation (EC) 850/2004
<ul style="list-style-type: none"> •Photo-imaging •Photo-resist and anti-reflective coatings for semi-conductors •Etching agent for compound semi-conductors and ceramic filters •Aviation hydraulic fluids •Metal plating (hard metal plating) only in closed-loop systems •Certain medical devices (such as ethylene tetrafluoroethylene copolymer (ETFE) layers and radio-opaque ETFE production, in-vitro diagnostic medical devices, and CCD colour filters) • Insect baits for control of leaf-cutting Brazilian ants from <i>Atta</i> spp. and <i>Acromyrmex</i> spp. <p>•Fire-fighting foam (until 2011).</p>	<ul style="list-style-type: none"> •Photo masks in the semiconductor and liquid crystal display (LCD) industries •Metal plating (hard metal plating) •Metal plating (decorative plating) •Electric and electronic parts for some colour printers and colour copy machines • Insecticides for control of red imported fire ants and termites •Chemically driven oil production <p>•Expired exemptions*</p> <ul style="list-style-type: none"> •Carpets •Leather and apparel •Textiles and upholstery •Paper and packaging •Coatings and coating additives •Rubber and plastics 	<ul style="list-style-type: none"> •until 26 August 2015, wetting agents for use in controlled electroplating systems; •photoresists or anti -reflective coatings for photolithography processes; •photographic coatings applied to films, papers, or printing plates; •mist suppressants for non-decorative hard chromium (VI) plating in closed loop systems; •hydraulic fluids for aviation

Figure III.2b. List of acceptable purposes and time-limited exemptions for the use of PFOS, its salts and PFOSF decided by the COP to the Stockholm Convention on POPs (UNEP/POPS/COP.4/38 2008; BAT/BEP Group of Experts 2017) and list of uses exempted from restrictions under Regulation (EC) 850/2004.

European Union implementation of the Stockholm Convention

The European Community (EC) ratified the Stockholm Convention on 16 November 2004. Individual states as well the European Community as a regional economic integration organisation has to establish an Implementation Plan to show the concrete action that will be taken against the POPs listed in the Convention. Regulation (EC) No 850/2004 of 29 April 2004 (as amended) complements earlier EC legislation on POPs and aligns it with the provisions of the Stockholm Convention. The Regulation 850/2004 contains provisions regarding production, placing on the market and use of chemicals, management of stockpiles and wastes, and measures to reduce unintentional releases of POPs, including PFOS. Furthermore, Member States must set up emission inventories for unintentionally produced POPs, national implementation plans (NIPs) and monitoring and information exchange mechanisms. The EC Implementation Plan, which complements the national plans of the EU Member States, was adopted on 9 March 2007 (EC 2007). The uses of PFOS and related substances exempted from restrictions under Regulation (EC) 850/2005 are presented in Figure III.2b.

Further information on the UK implementation of this legislative framework is discussed in Section 3.3.

III.2.3. REACH Regulation

The Regulation EC 1907/2006 aka REACH (Registration, Evaluation, Authorization, and Restriction of Chemicals) restricts the use of substances where risks to human health and the environment cannot be managed and encourages the substitution of the most hazardous substances with less dangerous alternatives. In 2017, the EU published measures to regulate PFOA, its salts and related substances under Annex XVII of Regulation EU 2017/1000 amending REACH). REACH refers to PFOA salts and polymers having a linear or branched perfluoroheptyl group with the formula C₇F₁₅ or perfluorooctyl group with the formula C₈F₁₇ directly attached to another carbon atom, as one of the structural elements. The restriction puts a limit on PFOA in products and mixtures at 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.

The PFOA amendment will be implemented in phases, starting 4 July 2020 and Figure III.3a shows examples of PFOA, its salts and related substances; Figure III.3b shows the time-limited exemptions from banning PFOA as a constituent, a mixture or an article of industrial and commercial products. It must be noted that in 2017, PFHxS and its salts were added to the REACH candidate list of substances of very high concern (Article 57(e) of the REACH Regulation); see also Box 1.

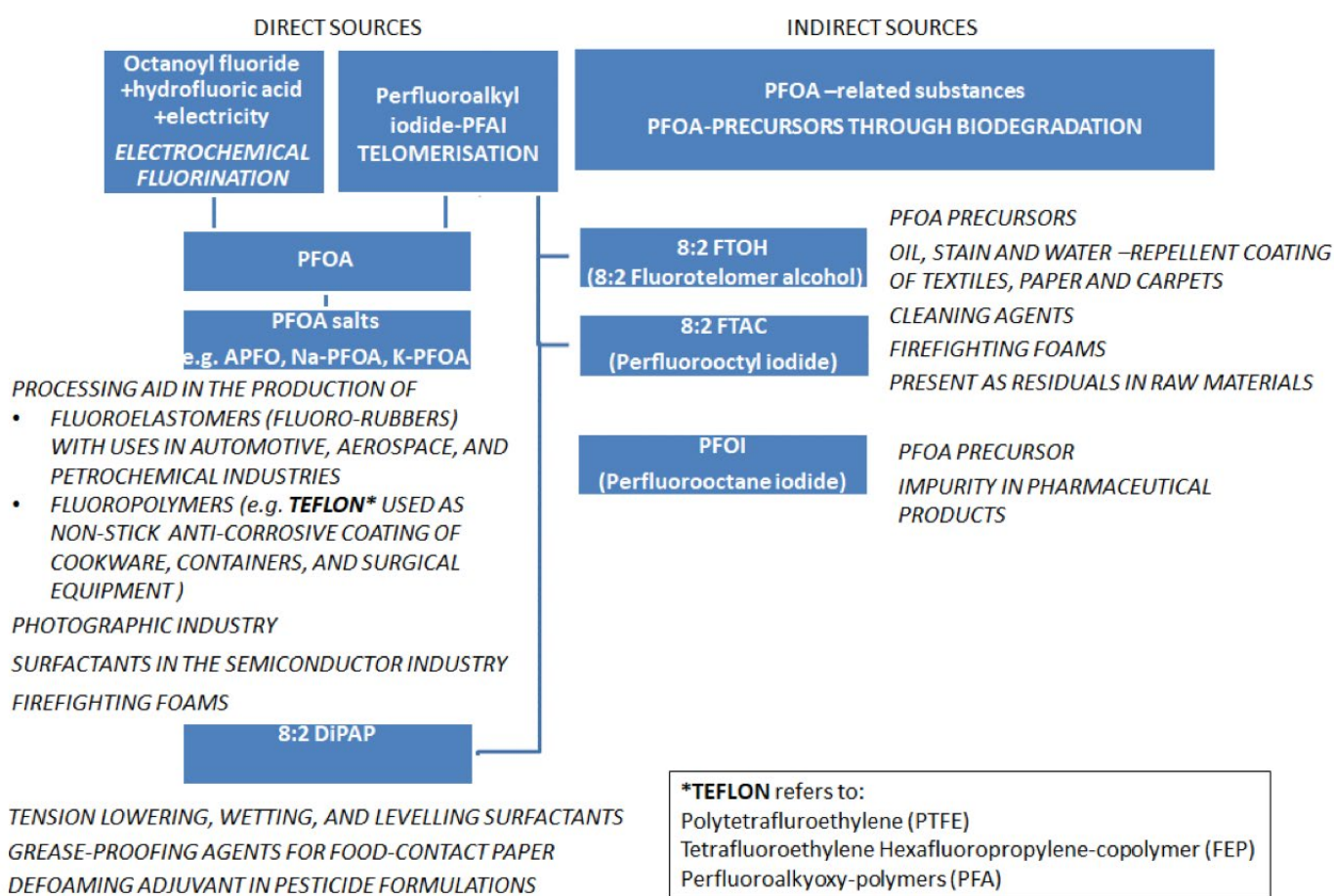


Figure III.3a. Sources, process scheme and major product categories and applications of PFOA, its salts and PFOA-related derivatives of biodegradation pathways. For specific applications see Figure 5. Source: Buck et al 2011; UNEP/POPS/POPRC.11/5 2015.

Specific Uses of PFOA and related substances banned after 4 July 2022	Specific uses of PFOA and related substances banned after 4 July 2023	Specific uses of PFOA and related substances banned after 4 July 2032	Uses/products not restricted by the REACH Regulation
<ul style="list-style-type: none"> •equipment used to manufacture semi-conductors •latex printing inks 	<ul style="list-style-type: none"> •textiles for the protection of workers from risks to their health and safety •membranes intended for use in medical textiles •filtration in water treatment, production processes and effluent treatment •plasma nano-coatings 	<ul style="list-style-type: none"> •medical devices other than implantable medical devices within the scope of Directive 93/42/EEC (cited in Reach Regulation) 	<ul style="list-style-type: none"> •unavoidable by-products of the manufacture of $\leq C6$ fluorochemicals •photographic coatings applied to films, papers or printing plates; in photo-lithography processes for semiconductors; or in etching processes for compound semiconductors •fire-fighting foam mixtures that were placed on the market before 4 July 2020

Figure III.3b. Exemptions from restrictions in the use of PFOA and related substances under REACH.

III.2.4 The Water Framework Directive

Directive 2000/60/EC, hereafter reported as Water Framework Directive (WFD), lays down a strategy against the pollution of water, including measures against priority hazardous substances. Priority substances refer to pollutants presenting a significant risk to or via the aquatic environment, including risks to waters used for the abstraction of drinking water. The WFD as amended by Directive 2013/39/EU lists PFOS in Annex X. Member States are required to adopt measures aiming at the cessation or phasing-out of discharges, emissions and losses to the water environment of priority substances including PFOS (WFD:Article 16). The specified environmental quality standard (EQS) value for PFOS refers to surface waters and, in the case of inland surface waters (i.e. rivers, lochs, lakes), it is set at 0.65ng/l (annual average).

It is worth noting that the UK Technical Advisory Group (UKTAG) on the WFD has provided advice on the PFOS limit of quantification (LOQ) (i.e. 0.2ng/l) and groundwater concentration threshold (i.e. guideline value=1000ng/l) below which the danger of deterioration in the quality of the receiving groundwater is avoided (UKTAG 2016). Appendix X in UKTAG (2016) presents the approach to adjusting standards in an associated surface water to account for groundwater dilution. The UKTAG guideline threshold for PFOS in groundwater is presented in UKTAG (2016) as an action level for water suppliers based on 10% of the Total Daily Intake to 1L of drinking water consumed by a one year old 10kg child as per PHA guidance (DWI 2007; 2009).

The WFD has not listed PFOA or any other PFAS as priority substances. However, Directive 2014/80/EU amending the Directive 2006/118/EC¹⁷ (Groundwater Daughter Directive) identified the need to obtain and respond to new information on other substances posing a potential risk and thereby establish a groundwater watch list (GWWL) for pollutants of groundwater. This will increase the availability of monitoring data on substances posing a risk or potential risk to bodies of groundwater. The GWWL is a voluntary mechanism and focuses on substances that are currently not regulated under the WFD due to lack of information on their presence in the environment (i.e. lack of monitoring data) or their pollutant properties (GWWL 2017). On the basis of this mechanism, SEPA "selected the suite of perfluorinated substances based on considerations of likely significance in terms of usage and/or environmental risks as well as cost and ease of analysis. SEPA monitoring was a very limited assessment. It was undertaken as part of a more general regional groundwater quality assessment process rather than as an in-depth study particularly focussed on perfluorinated compounds." (Isla Smail, SEPA, pers. com. September 2018). The perfluorinated substances sampled in groundwater by SEPA are discussed in Section 4.2 of the main document.

III.2.5 Tolerable daily intake and exposure to PFOS and PFOA

As an official EU risk assessor, the European Food Standard Agency (EFSA) produces scientific opinions and advice that form the basis for European policies and legislation. In the context of PFAS, EFSA's Contaminants (CONTAM) Panel, established a Tolerable Daily Intake for PFOS at 150 nanograms per kilogram of body weight per day and for PFOA at 1.5 micrograms (1,500 nanograms) per kilogram of body weight per day (EFSA 2008). Based on limited monitoring data from food and environmental samples, the Panel estimated an indicative dietary exposure of 60 ng/kg b.w. per day in the European population. Despite uncertainty in the assessments, the Panel estimated¹⁸ that drinking water contributes less than 0.5% of the indicative exposure of the European population to PFOS and less than 16% of the indicative exposure to PFOA (EFSA 2008). EFSA noted that additional exposure to PFOS and PFOA could result from precursor substances transformed to PFOS and PFOA within the body.

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¹⁷ Directive 2006/118/EC establishes specific measures as provided for in Article 17(1) and (2) of the WFD in order to prevent and control groundwater pollution.

¹⁸ as of 2008, i.e. before restrictions on PFOS and PFOA came into force.

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