



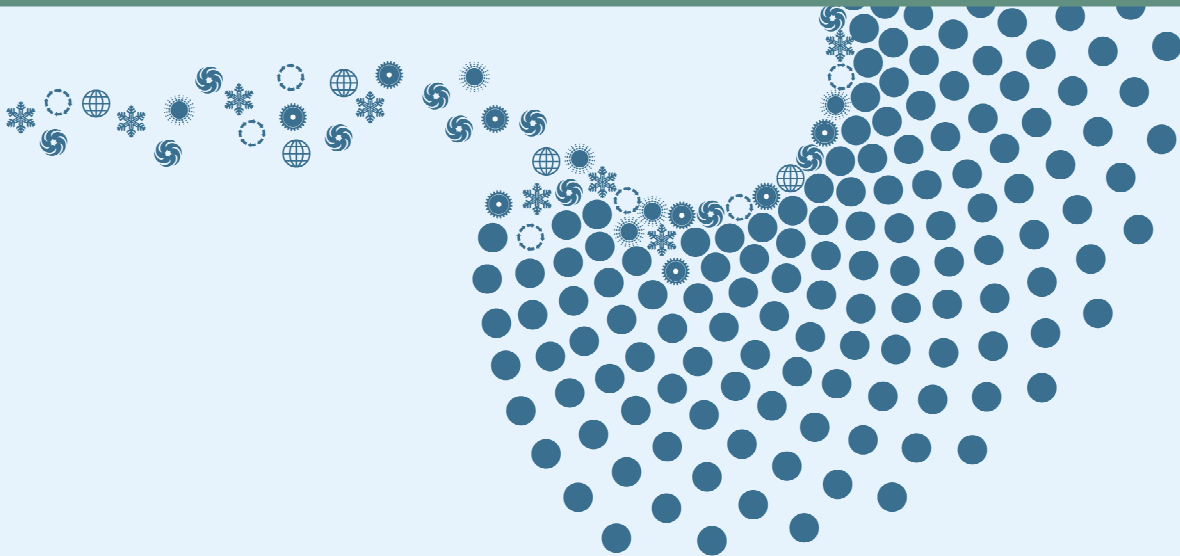
**Statens forurensningstilsyn**  
Norwegian Pollution Control Authority

Survey of national sources, 2007

# PFOA IN NORWAY

2354

2007



## **Preface**

The discharges of PFOA should be significantly reduced by 2010 and completely eliminated before 2020. Therefore, the Norwegian Pollution Control Authority (SFT) has commissioned IFP Research (Sweden), together with NILU (Norway) and FORCE Technology (Denmark), to perform a survey of national sources of PFOA and PFOA-precursors in Norway. The overall aim is to identify all possible sources of PFOA in Norway along the whole life cycle from production, use and disposal in industrial manufacturing and applications and other possible sources such as long range transport by air and sea currents.

This report outlines relevant perfluorinated products and possible user companies of these products, that were surveyed as potential sources of PFOA in Norway. PFOA is present in a low concentrations in consumer products such as coated and impregnated textiles, carpets, paper, paints and lacquers. Long range transport of PFOA, in estimated annual amounts from 130 kg to 380 kg, enters Norway mainly from the ocean and to a lesser extent from airborne particles, where the main part of PFOA has its origin from remote direct sources that manufacture polyfluorinated substances and polymers.

SFT, Oslo, December 2007

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## Table of content

<b>1.</b>	<b>Summary .....</b>	<b>5</b>
<b>2.</b>	<b>Background and purpose .....</b>	<b>8</b>
<b>3.</b>	<b>Chemistry .....</b>	<b>9</b>
<b>4.</b>	<b>Literature study on applications in products of PFOA and precursors.....</b>	<b>12</b>
4.1	PFOA related substances .....	12
4.2	Global use of PFOA.....	12
4.3	PFOA in products .....	17
4.3.1	Perfluorochemical treated products for food contact.....	17
4.3.2	Waterproofed textiles and impregnation fluids/agents .....	19
4.4	Precursors in products.....	20
4.5	Conclusions.....	22
<b>5.</b>	<b>Literature study on transport of PFOA and precursors.....</b>	<b>23</b>
5.1	Transport of PFOA .....	24
5.1.1	Local sources .....	24
5.1.2	Ocean currents .....	24
5.1.3	Wet and dry deposition .....	25
5.1.4	Long-range air transport .....	26
5.1.5	Sediment sorption .....	26
5.2	Transport and fate of PFOA precursors.....	27
5.2.1	Atmospherical transformation .....	27
5.2.2	Biological transformation .....	28
5.2.3	Waste incineration .....	28
5.2.4	Long range air transport.....	29
5.3	Transport models for PFOA and volatile precursors.....	30
5.4	Long range transport assessment of PFOA in Norway .....	33
<b>6.</b>	<b>Mapping of the use of PFOA in Norway .....</b>	<b>35</b>
6.1	Search in the Norwegian Product Register for use of PFCA-precursors.....	35
6.1.1	Identifying substances that potentially can degrade to PFOA (PFOA precursors) .....	36
6.1.2	Search in the Norwegian Product Register .....	36
6.2	Contact to companies.....	37
6.2.1	Identifying sectors and products that may be sources to PFOA.....	37
6.2.2	Companies to contact.....	39
6.3	Estimations based on statistics.....	39
<b>7.</b>	<b>Source information on relevant product groups for PFOA.....</b>	<b>41</b>
7.1	Impregnation agents.....	41
7.1.1	Impregnating agents for textiles .....	41
7.1.2	Waxes .....	42
7.1.3	Polish .....	43
7.2	Impregnated products .....	44
7.2.1	Paper (food paper, microwave paper, paper cups) .....	44
7.2.2	Textiles (out-door clothing, apparel, tarpaulins, tents).....	44
7.2.3	Foot wear .....	45

7.2.4	Carpets .....	46
7.3	Paint and lacquer.....	46
7.4	Fire fighting foams .....	48
7.4.1	Oil and gas industry .....	49
7.5	Other product categories.....	49
7.5.1	Herbicides .....	49
<b>8.</b>	<b>Results: Emissions, sources and trends in Norway.....</b>	<b>50</b>
8.1	Previous results; PFAS emissions in Norway (2002 - 2006) .....	50
8.2	Norwegian industrial sources of PFOA in 2007.....	52
8.3	Norwegian consumer product sources of PFOA in 2007 .....	55
8.4	Emissions of PFOA in Norway, 2007 .....	57
8.4.1	Sewage and landfill related environmental release of PFOA.....	57
8.4.2	Emission of PFOA from consumer products from the Norwegian marked ..	58
8.5	Trends of PFOA use in Norway .....	59
8.6	Conclusions.....	61
<b>9.</b>	<b>Discussion .....</b>	<b>61</b>
9.1	Knowledge and data gaps .....	61
9.2	Physico-chemical properties of fluorine chemistry .....	62
9.2.1	Physico-chemical properties of fluoropolymers and their treated surfaces...	62
9.3	Alternatives to C8-fluorotelomer chemistry .....	64
9.3.1	Fluorinated polyether.....	65
9.3.2	Fatty alcohol polyglycolether sulphate.....	65
9.3.3	Silicone polymers .....	65
9.3.4	Sulfosuccinate.....	66
9.3.5	Propylated aromatics .....	66
9.3.6	Conclusions.....	67
9.4	Future work.....	67
<b>10.</b>	<b>Reference List.....</b>	<b>69</b>
<b>11.</b>	<b>Appendix A – terminology and definitions.....</b>	<b>74</b>
11.1	Fluorochemical .....	74
11.2	Fluorinated chemical .....	74
11.3	Fluorotelomer .....	74
11.4	Fluorotelomer alcohol (FTOH).....	74
11.5	Fluoropolymer .....	74
11.6	Fluorinated (organic) polymer.....	74
11.7	Perfluoro- / perfluorinated .....	74
11.8	Perfluoroalkylated substance (PFAS).....	74
11.9	Fluoro surfactant.....	75
11.10	Perfluorocarboxylic acid (PFCA).....	75
11.11	Polyfluorinated substances .....	75

## 1. Summary

Perfluorinated products are extremely versatile and are used in a variety of industrial and consumer applications and products. Some of these perfluorinated products contain or release perfluoroalkyl carboxylates, among them perfluorooctanoic acid (PFOA) and its salts, that are documented as being persistent and widely spread in man and environment. Among the polyfluorinated products, some of them are known as PFOA-precursors, as they can be degraded to PFOA in the environment. PFOA will be classified by EU as a carcinogenic and reproductive toxicant according to the formal classification from of the advisory committee of hazardous substances of the European commission.

In context to Norwegian policy, the discharges of PFOA should be significantly reduced by 2010 and completely eliminated before 2020. Therefore the Norwegian Pollution Control Authority (SFT) has commissioned IFP Research (Sweden), together with NILU (Norway) and FORCE Technology (Denmark), to perform a survey of national sources of PFOA and PFOA-precursors in Norway. The overall aim is to identify all possible sources of PFOA in Norway along the whole life cycle from production, use and disposal in industrial manufacturing and applications and other possible sources such as long range transport by air and sea currents.

This report outlines relevant perfluorinated products and possible user companies of these products, that were surveyed as potential sources of PFOA in Norway. Since the Norwegian market is complex, priorities in the selection of relevant products and companies for a survey were necessary for the source identification and quantification of PFOA in Norway.

<b>Estimate of the annual maximal emission of PFOA from a range of consumer products identified in this survey as major contributors of PFOA</b>	
Carpets	12 kg
Coated and impregnated paper	1,3 kg
Textiles	0,5 kg
Paint and lacquer	1 kg
<b>Estimate of annual long range transport of PFOA into Norway</b>	
Total of direct and indirect sources	130 to 380 kg

For the determination of the quantities of relevant products on the Norwegian market and the companies that trade these products, a substantial search in the Norwegian Product register was performed. The search was based on a version of the OECD preliminary list of Perfluorinated Substances from April 2006, that cover around 1000 fluorinated substances. From this list 362 substances that may degrade into PFOA have been identified. In addition SFT has provided 2 substances from an earlier search, which resulted in a total search for 364 substances in the Norwegian Product Register. This search resulted in 29 of these substances being identified as fluorinated substances used in products in Norway. However, because of confidentiality reasons, no total amount for each of these 29 substances were reported.

A parallel search was conducted in the Nordic Business Key by D&B ([www.nordicbusinesskey.com](http://www.nordicbusinesskey.com)) updated in May 2007. This database covers all

registered companies in Norway, and among these 26.945 companies in various sectors were identified to be potential users of perfluorinated products. Since a direct contact with all these companies was a time consuming and unrealistic task, a strategy for a limited but effective contact activity was proposed to SFT. This strategy was based on the search in the Norwegian Product register in context to the size (turnover and number of employees) of the relevant companies, or if they already were known from previous studies as users of perfluorinated products. It was also recommended to identify companies that are positioned early in the Norwegian distribution chain, such as importers and distributors.

In this survey a range of impregnated consumer products and cleaning agents have been identified as potential sources of perfluorinated substances and consequently as potential sources of PFOA into Norwegian society and environment.

Coated and impregnated textiles are estimated in this study to emit approximately 0.5 kg pure PFOA in one year based on statistics of textile production for 2006 in correspondence to analytical results of PFOA from recent studies in Norway initiated by SFT.

Carpets have not been a target for chemical PFOA analyses in Norway yet, but according to correspondence with the chemical manufacturers, the PFOA concentration in factory treated carpets is about 5 times higher and for home treated carpets 25 times higher as in treated clothing. Since carpets are used in much higher amounts than clothing according to statistics from SSB in 2006, between 10 and 50 times higher amounts of PFOA are expected to enter the Norwegian market applied on carpets compared to textiles. With the perspective that 0,5 kg pure PFOA could have its origin from textiles in one year, an estimate that around 12 kg PFOA from carpets could enter the Norwegian society in one year.

Since paper-treatment products containing perfluorinated compounds are believed not to be in use anymore, only fluorotelomer-type treatment, containing trace amounts of PFOA (less than 0.1%) is considered in this report. Recent literature reported that the raw material for paper coating contains 88-160 µg PFOA/kg product. The author used a dilution factor of 300 for the application of the raw material to the finished product, resulting in an average PFOA concentration of 1860 µg/kg or <0.0002% PFOA in coated paper and cardboard.

Since the predominant manufacturer of coated and impregnated paper in Norway declared that the company didn't anymore use perfluorinated substances, the focus in this report will be on importation to Norway. Calculations based on statistics from SSB on imported coated paper into Norway for the year 2006, in combination with analytical results from recent literature sources on coated and impregnated paper, resulted in an estimate of 1.3 kg PFOA entering Norway applied on paper each year with increasing tendency.

According to calculations and estimations carried out in this survey impregnating agents for textiles, waxes and polishes, contribute to very low amounts of PFOA in comparison to impregnated carpets and impregnated textile goods, Amounts of PFO in impregnating agents are in the range of a few grams to almost 100 grams content of PFO. Paints and lacquers amount between 0.4 kg to 1.7 kg of PFO based on annual

statistics from SSB in context to reported total amounts of PFO from literature. This would mean that paints and lacquers are in the same range of PFO contribution as coated paper? and impregnated textiles.

A literature survey was conducted of recent literature data, that describe the latest knowledge about sources and transport of PFOA in society and environment. Many potential pathways for human exposure to PFCAs must be considered, including food, drinking water, air as well as consumer products. Food, surface waters and air are impacted by local, regional, continental and global PFCA emissions.

In 2004 SFT estimated a Norwegian emission of about 17 tons perfluoroalkylated substances per year (2002 data; SFT, 2004). No information about the portion of PFO in this number was given. With the conservative estimation of a 10% portion, 1700 kg PFO would have been emitted in 2002. With an 80% decline, according to literature, 340 kg PFO will be emitted in Norway during the period 2005 – 2050, less than 1/1000 of the estimated minimum emission globally. However, since consumption of PFO treated products still apparently increases in Norway, no decrease of emissions caused via the consumer product route can be expected soon.

This survey describes a range of analytical results concerning consumer products and historical data of indirect and direct emissions of PFOA from manufacturing use and processing of perfluorinated products in Norway. Due to the lack of sufficient data, this study cannot fully assess the relative importance of consumer articles in comparison to water, food and air to PFOA or PFCA exposure in Norway. It is strongly recommended that a priority be put on future studies, building on work which has been recently published, to make such an assessment. This data achieved in this study can be supportive in the future work for the identification of relevant perfluorinated chemicals and their importance as sources of PFOA in Norway.

## 2. Background and purpose

The Norwegian policy and goal is a significant reduction of the discharges of PFOA by 2010, and to eliminate use and emissions completely before 2020, the so-called one generation target report (Report to the Storting No. 14 (2006-2007), "Together for a Non-toxic Environment – Preconditions for a Safer Future").

In focus of this policy is, among other things, the protection of consumers from consumer products that have a direct impact on health, but also to regulate and reduce the amounts of prioritised hazardous substances from sources that contribute to the greater part of emissions to man and the environment. Among these prioritised hazardous substances are perfluorooctanoic acid and its salts (PFOA) and related compounds.

SFT (Norwegian Pollution Control Authority) has therefore commissioned a survey that aims to identify possible sources of PFOA along their life cycle in Norway from use and exposure in industrial manufacturing and applications and other possible public and industrial sources such as long range transport by air.

Perfluoroalkylated substances (PFAS) are used in several industrial branches such as paper, plastic, engineering, leather and textile industry, but they also occur in a large range of consumer product preparations, for example in cleaning and polishing products, but also as surfactants in fire fighting foams.

The present knowledge of the exact chemical compositions in preparations of perfluorinated compounds is very limited. This makes it difficult to evaluate the exposure and the impact of the presence of PFAS in the environment. Additionally, new studies suggest that environmental sources and food may be important in fully understanding human exposure to PFOA (Fromme et. al, 2007; Horowitz et al., 2007).

The methodology used for the product survey was to

- Identify relevant business sectors and relevant companies within these sectors.
- Perform a search in the Norwegian Product Registry and other databases.
- Contact the most relevant companies operating in Norway. The contact were primarily by personal contact (phone interviews) supplemented by e-mail correspondence.
- Contact international producers of polyfluorinated compounds (PFCs) such as DuPont and 3M.

Results of the project are

- An assessment of different sources and their relative contributions to emissions of PFOA in Norway along its life cycle from production, use and disposal in industrial manufacturing and consumer applications and other possible sources like long range transport by air and water currents.
- Identifying gaps of knowledge for future research within this area.



### 3. Chemistry

Polyfluoroalkylated substances belong to a large and complex group of organic substances that are extremely versatile and used in a variety of industrial and household applications.

The main characteristics of these polyfluorinated compounds are the replacement of most hydrogens by fluorine in the aliphatic chain structure. Some of these organic fluorine compounds are known as perfluorinated, which means that all hydrogens have been replaced with fluorine.

The chemical structure of PFOA (perfluorooctanoic acid, pentadecafluorooctanoic acid) contains a perfluorinated carbon chain with seven carbons connected to a carboxylate group (RCO<sub>2</sub><sup>-</sup>). Commonly the abbreviation PFOA describes both the perfluorooctanoic acid and its salts. Only the ammonium, sodium, potassium and silver salts of PFOA are in industrial use.

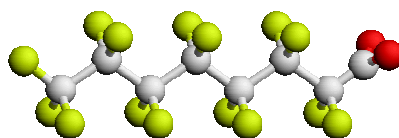
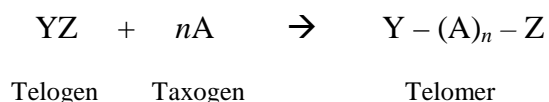


Figure 1: Perfluorooctanoate.

Perfluorinated carboxylic acids (PFCAs) do not occur naturally. They have been manufactured in 50 years (Kissa, 2001). The principal process for industrial manufacture of PFCAs, and specifically PFOA, is by electrochemical fluorination (ECF) (Kissa, 2002, Prevedouros et al, 2006).

In the ECF process, all hydrogens are replaced with fluorine in a radical reaction. The ECF process yields 10-30% branched isomers as well as a homologous series of acids. For instance, acids with 4-9 carbons result from ECF production of PFOA. The majority of PFOA, largely as its ammonium salt, made has been manufactured by the ECF process. The major global PFOA manufacturer, the 3M Company, announced in 2000 that they would stop their production. Other producers have continued to manufacture PFOA by the ECF process.

A second process used to manufacture PFOA is oxidation of perfluorooctyl iodide, which is made by the telomerisation process. The telomerisation process reacts a molecule, called telogen, with two or more unsaturated molecules in the ethylene family, called taxogens. The principal reaction is:



When pentafluoroethyl iodide (telogen) as an example is reacted with tetrafluoroethylene (taxogen) straight, short chain “telomers” are with the general formula: F[CF<sub>2</sub>CF<sub>2</sub>]<sub>n</sub>I where *n* is between 4 and 8 (Kissa, 2001). The telomerization process yield only linear, even carbon number substances as it is practiced commercially (Kissa, 2001). Perfluorooctyl iodide, where *n*=4, is a raw material used

to manufacture the ammonium salt of PFOA (Grottenmuller, 2002). PFOA is not used or added in the manufacture of telomers, however it may be formed as an unintended by-product from oxidation of perfluoroalkyl iodide raw material (Prevedourous, 2006). PFOA is also a by-product in the manufacture of perfluorooctyl sulfonyl-based products (Prevedourous, 2006).

Residual raw materials in the manufacture of fluorotelomer and perfluoroalkylsulfonyl-based products such as fluorotelomer alcohols and perfluoroalkyl sulfonamido alcohols have been shown to be potential indirect sources, “precursors”, of PFOA and PFCAs from their degradation in the environment (Prevedourous, 2006). Recently, the global manufactures of fluorotelomer-based products have entered in to a voluntary programme whereby they have committed to reduce by 95% the levels of PFOA and PFOA precursors in emissions and products by 2010 (US EPA, 2006).

The major historic industrial use of ammonium salt of PFOA since the 1950s has been as a processing aid in the manufacture of fluoropolymers such as polytetrafluoroethylene (PTFE) (Kissa, 2001). PFOA and other PFCAs have also been used in fire fighting formulations and a wide array of industrial and consumer products (Prevedourous, 2001). The direct emission of PFOA to the environment from its manufacture and use are the largest (>90 %) historic sources of PFOA in the global environment.

The different chemical-physical properties of PFOA versus volatile precursors will result in differing transport pattern of the two compound groups. Directly emitted to water in the environment PFOA is expected to dissolve and dissociate almost entirely to the perfluoroactanoate but the protonated form of PFOA is always present. Recent studies suggest that the pKa of PFOA at environmentally relevant concentrations is 3.5, one order of magnitude higher than previously reported (Ellis, 2007).

In addition, the undissociated acid has a very high vapour pressure lending it capable of atmospheric transport (Kaiser, 2005). Experiments have shown that PFOA can be measured in the air above an aqueous solution containing PFOA (Ellis, 2006, European commission, 2006)

A recent study investigating emissions from an industrial facility showed that indeed air transport over long distances of particle-bound PFOA directly emitted from a fluoropolymer manufacturing site is possible (Barton, 2007).

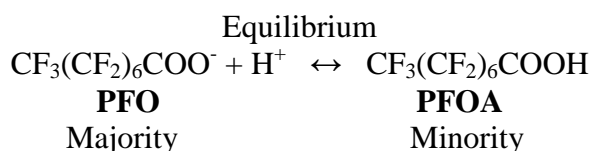


Table 1: Physico-chemical characteristics of PFOA and APFO ( ammonium perfluorooctanoate) ; (European Commission, 2006)

2.1 Physical form	PFOA and APFO are solids. (Kirk-Othmer, 1994).		
2.2 Molecular weight	PFOA: 414.09 APFO: 431.10		
2.3 Melting point/range (°C)	PFOA: 52 – 54 °C (Kirk-Othmer, 1994). PFOA: 54.3 °C (Lide, 2003).		
2.4 Boiling point/range (°C)	PFOA: 189 °C (Kirk-Othmer, 1994). PFOA: 189-192 °C/736 mm Hg (Boit, 1975) APFO: 130 (decomposition) (3M Company, 1987)		
2.5 Decomposition temperature	No data found PFAO and APFO.		
2.6 Vapour pressure (Pa(°C))	PFOA: 0.15 mm Hg at 25 °C /Estimated (Lyman WJ, 1985). PFOA: 10 mm Hg at 25 °C (approx.) (Exfluor, 198) APFO: 0.0081 Pa ( $6 \times 10^{-6}$ ) at 20 °C, calculated from measured data (Washburn et al., 2005). PFOA: 2.25 Pa (0.017 mm Hg) at 20 °C and 4.19 Pa (0.031 mm Hg) at 25 °C (Kaiser et al., 2005)		
2.7 Relative density ( $D_4^{20}$ )	Density/specific gravity. 1.792 (g/ml) (Kirk-Othmer, 1994).		
2.8 Vapour density (air = 1)	14.3 (calculated, the ratio of the molecular mass of PFOA , 414.09 g/mol to the "mean molecular mass" of air 29.0 g/mol)		
2.9 Conversion factor (11011 hPa at 25°C)	PFOA: 1 ppm = 17.21 mg/m <sup>3</sup> APFO: 1 ppm = 17.92 mg/m <sup>3</sup>		
2.10 Solubility	conc. at sat. (g/l)	solvent	Temperature (°C)
	PFAO: 3.4	water	20 °C (Merck, undated)
	PFOA: 9.5	water	25 °C (Kauck and Diesslin, 1951)
	APFO: > 500	water	20 °C (3M Company, 1987)
	PFOA: 4.14	water	22 °C (Prokop et al., 1989)
2.11 Partition coefficient (log Pow)	Experimental No data Calculated No data.		
2.12 Flammability	No data found.		
Flash point (°C)	open cup:		closed cup:
Explosivity limits (% v/v)	lower limit:		upper limit:
Auto-flammability temp. (°C)			
2.13 Explosivity	No data found.		
Danger of explosion at result of:	shock:	friction:	ignition:
Explosive properties at high temperature			
2.14 Oxidizing properties	No data found.		
2.15 Other physico-chemical properties	Dissociation Constants: pKa = 2.80 in 50% aqueous ethanol (Brace, 1962). pKa = 2.5 (Ylinen et al., 1990)		

## 4. Literature study on applications in products of PFOA and precursors

PFOA has been classified as toxic (T; R48/23), carcinogenic (Carc Cat2, R45), and a reproductive toxicant (Repr Cat2, R61) by the EU on the 31<sup>st</sup> of August 2007 (SFT, 2007). Presently, no national or European regulation of the use of PFOA exists. In the SFT publication: Impact assessment of a proposal for prohibition on certain hazardous substances in consumer products, dated 29 May 2007, it is proposed to ban PFOA, its salts and esters in consumer products with a threshold level of 0.005 weight % and 1 µg/m<sup>2</sup> in textiles and other coated materials.

### 4.1 PFOA related substances

The Organization for Economic Co-operation and Development (OECD) has published a preliminary list of PFOS, PFC, PFOA and related compounds, and chemicals that may degrade to PFCA (OECD, 2006a). Annex III in this publication contains a list of PFOA and its salts, and polymers that contain PFOA as a part of the entire polymer. Both linear and branched polyfluorinated octanoic acid derivatives, telomers and polymers are included. However, in general, only chemicals/fluorotelomers containing a linear perfluorinated C<sub>7</sub>-chain connected to a carbon atom have the potential to form PFOA and be PFOA precursors. Since PFOA contains a linear C<sub>8</sub>-backbone, only chemicals/fluorotelomers, mentioned on that list, containing a linear perfluorinated C<sub>7</sub>-chain connected to a carbon atom have the potential to form PFOA and be PFOA precursors.

In a second publication, the OECD addressed results of a 2006 survey on the production and use of PFOS, PFAS, PFOA, PFCA, their related substances and products/mixtures containing these substances (OECD, 2006b). The questionnaire circulated was returned by 17 OECD countries, 6 non-OECD countries and 2 companies. The responses showed that 4 countries did manufacture/import 4 PFOA related substances. These substances were PFOA ammonium salt (CAS No. 3825-26-1), PFOA sodium salt (CAS No. 335-95-5), the linear pentadecafluorooctanoic acid (PFOA acid, CAS No. 335-67-1) and a mixture of the linear and branched forms (branched pentadecafluorooctanoic acid, CAS No. 90480-55-0). In 2005 the manufacture/import volumes of the PFOA as acid, PFOA ammonium salt and PFOA sodium salt were up to 10, 260 and 10 tons, respectively.

### 4.2 Global use of PFOA

In 2005 almost all PFOA and its ammonium salt were used as polymerisation aids in the manufacture of fluoropolymers and as a component in aqueous fluoropolymer dispersions (OECD, 2006b). While the majority of fluoropolymers are sold as solids, where the PFOA (and PFNA) processing aid is removed during industrial processing, some fluoropolymer dispersions, which still contain PFOA, were used in coatings for non-stick cookware, paint formulations, as photographic film additives, as breathable barrier films for textiles and in the textile finishing industry. These fluoropolymer dispersions were mainly for industrial uses, but some paint products could be for consumer use as well. The fluoropolymer resin was used for glass fibre impregnation in tubes, cables, tape for industrial use as well as in the coating of non-stick cookware.

In 2005 four countries manufactured/imported 22 precursors of perfluorocarboxylic acid (PFCA) (OECD, 2006b). The volumes of the perfluorinated precursors and polyfluorinated precursors of PFCA were between 384 and 2 580 tons and between 344 and 3 440 tons, respectively. Only some of them have the potential to form PFOA and notably in much smaller amounts than the addition of precursors themselves since PFOA is a much smaller molecule than the precursors. The 22 PFCA precursors included:

- 1 perfluoro-amine compound 10-100 tons;
- 2 perfluoro-ether compounds 240-1 140 tons;
- 4 perfluoro-iodide compounds 112-1 120 tons;
- 4 partial perfluoro compounds 22-220 tons;
- 3 fluoro-alcohol compounds 21-210 tons;
- 5 fluoro-ester compounds 212-2 120 tons;
- 1 fluoro phosphate compound 10-100 tons;
- 2 poly fluoro compounds 101-1 010 tons.

PFCA precursors were also reported in coating formulations or cleaning/washing products for textile, leather, paper, glass, masonry and concrete surfaces, in anti-foam formulations, in silicone rubber products, in ink cartridges of printers and analytical laboratory uses. All these uses were at low volumes (OECD, 2006b).

Prevedouros et al. (2006) summarized the recent and the historical global use of PFCAs, mainly PFOA, in the last 50 years, distinguishing between direct and indirect sources. Amounts and time periods are given in Figure 2 (Prevedouros *et al.* 2006b):

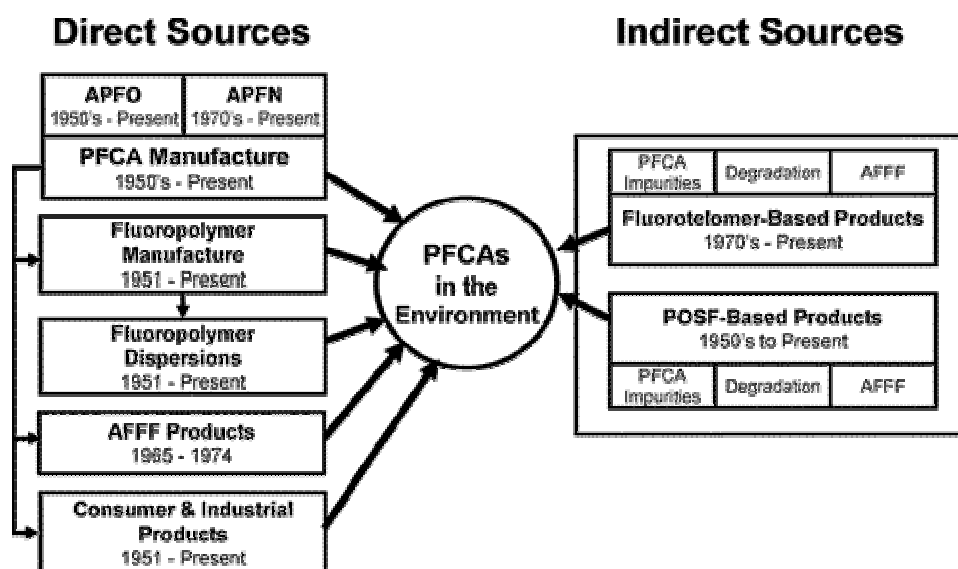


Figure 2: Potential sources of perfluorocarboxylates (PFCAs); APFO = ammonium perfluorooctanoate; APFN = ammonium perfluorononanoate; AFFF = aqueous fire-fighting foam; POSF = perfluorooctylsulfonyl fluoride, taken from (Prevedouros *et al.* 2006b).

Table 2: Global Historical PFCA Production and Emissions Summary, taken from (Prevedouros et al., 2006b)

Environmental input source	Historical time period (years)	Estimated total global historical PFCA emissions (metric tons)	Estimated total global production (metric tons)
<b>Direct PFCA Sources:</b>			
<b>1) PFCA manufacture</b>			
PFO/APFO	1951-2004	400-700	3600-5700
PFN/APFN	1975-2004	70-200	800-2300
<b>Total manufactured</b>		<b>470-900</b>	<b>4400-8000</b>
<b>2) Industrial and Consumer Uses</b>			
Fluoropolymer manufacture (APFO)	1951-2004	2000-4000	
Fluoropolymer dispersion processing (APFO)	1951-2004	200-300	
Fluoropolymer manufacture (APFN)	1975-2004	400-1400	
Fluoropolymer processing (APFN)	1975-2004	10-20	
Aqueous fire fighting foams (AFFF)	1965-1974	50-100	
Consumer and industrial products	1960-2000	40-200	
<b>Total direct</b>		<b>3200-6900</b>	
<b>Indirect PFCA Sources:</b>			
<b>1) POSF-based products</b>			
PFCA residual impurities	1960-2002	20-130	
POSF-based precursor degradation	1960-2002	1-30	
POSF-based AFFF	1970-2002	3-30	
<b>2) Fluorotelomer-based products</b>			
PFCA residual impurities	1974-2004	0.3-30	
Fluorotelomer-based precursor degradation	1974-2004	6-130	
Fluorotelomer-based AFFF	1975-2004	< 1	
<b>Total indirect</b>		<b>30-350</b>	
<b>Total source emissions (direct + indirect)</b>		<b>3200-7300</b>	

<sup>a</sup> Low and high estimated values as well as the period of use/production for each source are based upon publicly available information cited in the text.

Direct sources with up to 6 900 tons emitted PFCA were representing the vast majority of PFCAs emitted to the global environment compared to indirect sources which contributed up to 350 tons.

The same research group published a model for the long-term (1950-2050) global fate of PFOA by composing an initial global-scale mass balance model to evaluate, whether identified direct emissions of PFOA from manufacture and use could account for observed concentrations of PFOA in the environment. Table 3 lists both direct and indirect PFOA emission sources to the global environment. This table also presents estimated minimum and maximum projected cumulative emissions between 2005 and 2050 for each source together with the contribution of each source to the total PFOA emissions for the time period. Projected cumulative emissions between 2005 and 2050 (480-950 tons) are forecasted to be at least 80% lower than the estimated cumulative emissions between 1950 and 2004 (Armitage *et al.* 2006).

Table 3: Estimated Historical (1950-2004) and Future (2005-2050) PFOA Emissions (Armitage et al. 2006).

PFOA emission source	1950-2004 min-max (metric tons)	% of total PFOA emission (average)	2005-2050 min-max (metric tons)	% of total PFOA emissions (average)
<b>Direct Sources</b>				
FP manufacturing (APFO)	2060-4090	72.3%	410-815	86.0%
APFO manufacturing	370-590	11.8%	20-40	4.2%
FP dispersion (APFO)	215-340	6.8%	45-75	8.7%
AFFF-ECF	50-100	1.8%	0	0%
FP manufacturing (APFN)	3-10	0.1%	<1-2	0.1%
Consumer & Industrial Products	2-10	0.1%	0	0%
APFN manufacturing	1-2	0%	<1	0%
PVDF (APFN)	<1	0%	<1	0%
<b>Direct sources:</b>	<b>2700-5140</b>	<b>92.9%</b>	<b>475-932</b>	<b>99.0%</b>
<b>Indirect Sources</b>				
POSF raw material degradation	4-585	5.0%	0	0%
POSF impurities	14-110	1.2%	0	0%
POSF-AFFFs	2-23	0.2%	0	0%
FT raw material degradation	3-60	0.6%	1-14	0.8%
FT impurities	<1-17	0.1%	<1-4	0.2%
<b>Indirect sources:</b>	<b>23-795</b>	<b>7.1%</b>	<b>1-18</b>	<b>1.0%</b>
<b>Direct and indirect sources:</b>	<b>2723-5935</b>	<b>100.0%</b>	<b>476-950</b>	<b>100.0%</b>

<sup>a</sup> AFFF = aqueous film forming foams (also aqueous fire fighting foams); APFN = ammonium perfluorononanoate; APFO = ammonium perfluorooctanoate; ECF = electrochemical fluorination, a process used to produce fluorinated chemicals; FP = fluoropolymer; FT = fluorotelomer; POSF = perfluorooctanesulfonyl fluoride; PVDF = polyvinylidene fluoride.

The contribution of indirect sources is expected to decrease both in absolute numbers and relative to direct sources within the year 2050 (Armitage, 2006).

Following processes occurring during the life cycle of a general PFS-containing consumer product can cause emission and transfer of PFS into the environment:

- Offgassing
- Washout
- Binding to particles



- Abrasion
- Cleaning/Washing
- Disposal
- Further processing
- Migration
- Uptake and excretion into the human organism
- Uptake from plants and animals
- Leachate

Which of these processes dominate for PFOA is yet unknown.

### 4.3 PFOA in products

The chemical resistance, non-stick properties and thermal stability of fluoropolymers such as PTFE have lead to a variety applications in consumer products, which may be potential PFOA sources. Additionally, PFOA can be found as impurity and unintended by-product in consumer products treated with polyfluorinated chemicals such as fluorotelomers and perfluoroalkylsulfonic acid derivatives e.g. perfluorooctane sulfonyl fluoride (PFOSF).

#### 4.3.1 Perfluorochemical treated products for food contact

Polytetrafluoroethylene (PTFE) is, for example, used to coat cookware intended for stovetop cooking and baking. Other polyfluorinated chemicals, such as fluorotelomer and perfluoroalkylsulfonic acid derivatives (e.g. PFOSF), are or have been used to treat paper to improve its moisture and oil barrier properties. In particular, papers used in contact with high-fat content foods may be treated with fluorotelomer or perfluoroalkylsulfonyl-based paper additives/coatings to prevent oil stains or oil soak through the paper. Typically, these paper coatings/additives are phosphate esters or acrylic polymers containing polyfluoroalkyl functionality (Begley *et al.* 2005).

Larsen *et al.* (2005) detected small amounts of PFOA (up to 140 ppb) in extracts of polytetrafluoroethylene (PTFE) resins, obtained after applying pressure and increased temperatures to the material. Subsequent studies of cookware, coated with PTFE dispersions have shown no-detectable-levels of PFOA extractable from cookware under normal use conditions (Powley *et al.*, 2005). A later study by the Norwegian Institute of Public Health (2007) went in further details about these findings. In a worst case scenario the new study showed that an adult human would be exposed to 66 ng PFOA /kg bw, when drinking 100 ml water cooked in a PTFE coated pan. It was concluded that, even at an assumption of 100% uptake of PFOA, these extremely low levels will not be an essential intake route for humans. According to Horowitz, 98% of the PFOA intake is contributed to by food (Horowitz, 2007).

Begley *et al.* (2005) analysed several consumer products for PFOA and concluded that fluorotelomer-based paper coating/additive formulations before application onto paper have the highest PFOA content, but during normal application rates this amount of PFOA will be diluted by about 300 times on the final paper product (see Table 4). Therefore, the PFOA content in finished paper should be in the few hundred mg/kg range, which is consistent with the data in Table 4.

Table 4: Summary of PFOA analysis in product, (Begley *et al.* 2005).

Consumer products	Concentration of PFOA ( $\mu\text{g/kg}$ )
PTFE cookware	4–75
Dental floss (PTFE based)	3
Dental tape (PTFE based)	4
PTFE film/sealant tape	1 800
FEP (fluoro-ethylene-propene copolymer) tubing	nd
Popcorn bags	6–290
French fry box	nd
Paper plates (soak-proof shield)	nd
Hamburger and sandwich wrapper	nd
Perfluoro paper coatings (not applied)	88 000–160 000

nd = non detects

The residue content of PFOA in PTFE-products is directly related to the processing temperatures used to make the products. Cookware and dental products use a high temperature sintering process that should volatilize PFOA, while production of PTFE film used as e.g. sealant tape does not use that sintering process. Begley *et al.* (2005) conclude that fluoropolymer food-contact materials do not appear to be a significant source of human exposure to perfluorochemicals (e.g. PFOA). In particular, the coated cookwares tested did not appear to be a significant source of PFOA. Furthermore, an extreme heating test (abusive) of the cookware did not appear to increase the residual amount of PFOA in the cookware. Additional PFOA did not appear to be formed during the normal use or misuse of these products.

This results were in contrast to the results of a more recent study of Sinclair *et al.* (2007), where gas-phase release of PFOA, 6:2 FTOH, and 8:2 FTOH were measured from heating non-stick frying pans and microwave popcorn bags. Gas-phase PFOA was measured in all four non-stick frying pan brands. PFOA was reported to vaporize at 189 °C and decompose at temperatures higher than 234 °C .

The authors suggest that residual PFOA is released from the PTFE coating to the gas phase under the normal cooking temperatures. Gas-phase concentration of PFOA varied depending on the frying pan brand, which suggests that the sintering conditions (temperature, pressure, and duration) used in the coating of fluoropolymers may have an influence on the release of PFOA. In addition, PFOA was detected in water boiled for 10 min. in three brands of non-stick frying pans (Table 5).

Table 5: Released amounts (ng) and concentrations ( $\mu\text{g}/\text{m}^2$ ) of PFOA and fluorotelomer alcohols from non-stick frying pans of 4 different brands (Sinclair *et al.* 2007).

Brand	Surface temperature	Area	PFOA		6:2 FTOH		8:2 FTOH	
	$^{\circ}\text{C}$		ng	$\mu\text{g}/\text{m}^2$	ng	$\text{g}/\text{cm}^2$	ng	$\mu\text{g}/\text{m}^2$
1	180	640	12	19	16	25	73	114
2	229	477	32	67	97	204	298	625
3	190	670	192	287	36	54	28	42
4	205	659	40	61	<10	<15	40	61
Stainless steel	230	670	<5	<7	<10	<15	<10	<15

In the same study, PFOA was found in the vapours produced by microwave heating of pre-packed popcorn bags. Furthermore, mg quantities of both PFCAs and FTOHs were calculated to coat the entire surface of the package (Sinclair *et al.* 2007). The authors noted that they were not able to explain the origin of the FTOHs from the cookware, because FTOHs are not used to manufacture cookware, and no plausible way for FTOH to be formed from PTFE is known.

Table 6: Released amount (ng) of PFOA of fluorotelomer alcohols at making popcorn in a bag of three different brands. (Sinclair *et al.* 2007)

Brand	PFOA	6:2 FTOH	8:2 FTOH
1	16	223	258
2	17	<20	<20
3	<2,5	<20	<20

#### 4.3.2 Waterproofed textiles and impregnation fluids/agents

For many applications, easy maintained textiles are desired (upholstery, table-cloths, car seats), or the textiles must be waterproof but breathable (tents, ski clothes, rain jackets, shoes). PFCs repel both dirt, water and grease, and are therefore ideal chemicals for surface treatment of such textiles. Furthermore, polytetrafluoroethylene (PTFE)-based membranes are often used in rain coats due to their water resistance and ability to “breathe”. In this context PFOA may be found in the products as an impurity as noted earlier and not as an deliberately added active component.

The Norwegian Pollution Control (SFT) and Friends of the Earth Norway reported results of the analyses of perfluorinated compounds in textiles in 2006 (Schulze *et al.* 2007; SFT, 2006). The content of PFOA in the analysed textiles varied between 0.4 and 127  $\mu\text{g}/\text{m}^2$  textile (Table 7). The highest values were found for a jacket produced by Bergans with 127  $\mu\text{g}/\text{m}^2$ . The levels of PFOA were considered low and in the same order of magnitude as findings in biota from remote locations.

Table 7: PFOA concentrations in water- and stain proofed textiles in  $\mu\text{g}/\text{m}^2$  of single samples (Schulze *et al.* 2007; SFT, 2006)

<b>Results from SFT report</b>							
Kmk 3.08	Colour Kids 0.42	Marmot 13.7	Sandvika 34.2	Head 1.21	Ultima 9.26	Reima 1.82	H&M 2.26
Stormberg 1.73	Stormberg 8.23	Fjell Råven 32.7					
<b>Results from Friends of the Earth, Norway report</b>							
PolarnOPYret 4.76	Peak performance 24.6	HellyHansen 20.4	Stadium 0.80	Bergans 127	Bergans 23.2		

Recently, Friends of the Earth, Norway, published a report about PFAS in impregnation fluids, covering PFOA as well. Thirteen commercial products were analysed for a variety of PFAS. Seven of the investigated products contained PFOA, varying between 45 and 692 ng/mL (Naturvernforbundet, 2007). Waterstop from Collonil classic contained the highest amounts of PFOA with almost 700 ng/mL. Additionally, other PFCA as well as up to 1000-times higher amounts of 8:2 FTOH were detected.

A risk characterization from the potential exposure to PFOA in consumer articles has been published (Washburn *et al.*, 2005). The authors investigated potential human exposure to PFOA in a wide variety of consumer articles, including treated textiles and concluded that the trace levels of PFOA present would not be expected to cause adverse human health effects not contribute to quantifiable levels of PFOA in human blood. The authors noted that PFOA was present in a number of consumer articles, which were not treated with fluorinated products. This may result from the presence of PFOA contamination globally (Yamashita *et al.*, 2005).

#### 4.4 Precursors in products

The formation of PFOA from fluorotelomers and perfluorooctyl sulfonamide raw materials and polymers may occur during use of treated products, as a result of atmospheric photolytic processes or via metabolism in humans and biota. Potential global environmental concentrations of PFOA from these indirect sources has been assessed and estimated to be small (Prevedouros, 2006). None the less, in Norway, precursors to PFOA may potentially arise from local source and from long-range transport.

A study performed by Dinglasan-Panlilio *et al.* (2006), indicates the potential for a significant amount of residual fluorinated alcohols to be released from a suite of fluorinated materials that are industrially applied and commercially available and hence contribute substantially to the atmospheric burden of FTOHs (Figure 3).

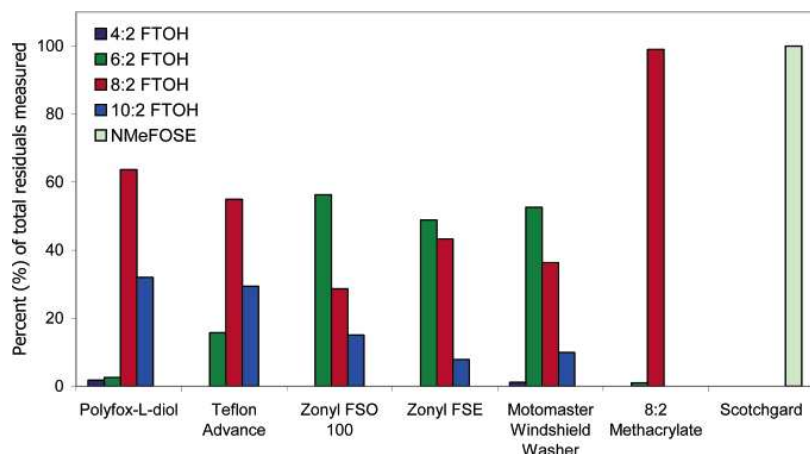


Figure 3: Profiles of residual unbound polyfluorinated alcohols detected in the seven fluorinated materials tested. Values are expressed as percent of total residuals measured, taken from (Dinglasan-Panlilio *et al.* 2006).

The examined fluorinated materials contained 0.04-3.8% residual free fluorinated alcohols on an applied fluorinated alcohol basis (dry mass basis). The ability of these compounds to undergo long-range atmospheric transport provides means to the production of PFCAs in the environment, as they degrade. This study also suggests that direct exposure of the general population to these compounds is plausible, if these materials are applied in homes and are outgassing after treatment of surfaces, including carpet, textiles, or paper products. Metabolism of these volatile precursors could then lead to the perfluorinated acids detected in human blood samples worldwide (Dinglasan-Panlilio *et al.* 2006).

Two patterns of PFCAs, were found (Figure 4 and Figure 5). They are hypothesized to represent the two main production processes of PFCAs. Figure 4 shows a “common” almost symmetric distribution of PFCA homologues around PFNA, which leads to the conclusion that this PFCA mixture originates from an electrochemically produced PFNA.

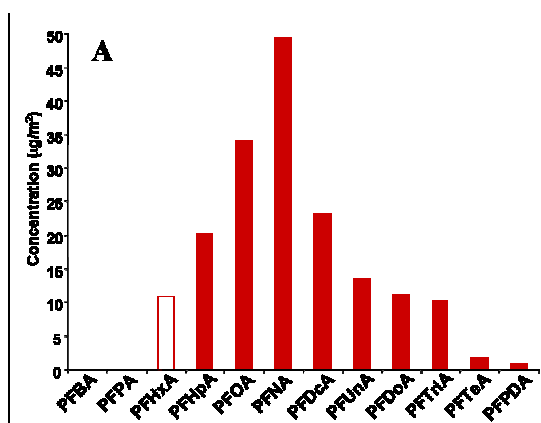


Figure 4: PFCA pattern in the extract from a textile (A) (Berger and Herzke, 2006).

In contrast, Figure 5 shows predominantly even carbon numbered homologues, dominated by PFOA, as could be expected from a telomerisation production of PFOA. Surprisingly, in both textile extracts PFCAs up to C15 could be detected, which might point to direct sources of long-chain perfluorocarboxylates (Berger and Herzke, 2006). So far it was hypothesised, that these must be degradation products of

other long-chain fluorochemicals, such as FTOHs (Martin et al., 2004a). Further research is needed to better understand these patterns and to be able to more confidently assign their origin.

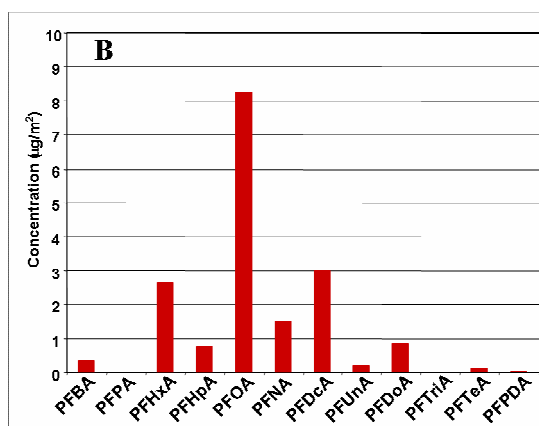


Figure 5: PFCA pattern in the extract from a textile (B) (Berger and Herzke, 2006).

## 4.5 Conclusions

PFOA can be present in consumer products treated with fluorinated compounds due to intentional application, in form of an unintended residues or due to degradation of precursor compounds like FTOHs. It is not always possible to distinguish between these cases, since recipes of technical applications are mostly confidential or the actual composition of the used mixture of active compounds confidential. Products intended for contact with food seem to contain small PFOA amounts, but since almost all available data origins from authors related to fluoropolymer manufacturing companies, the interpretation of these data should be done carefully. The same is the case for research on metabolism of PFOA in organisms. Data from independent research groups are needed in order to confirm these potentially prejudiced findings.

## 5. Literature study on transport of PFOA and precursors

PFOA is present in wildlife, humans, air and waters throughout the world. It has been emitted for over 50 years to the global environment. PFOA is water-soluble and waters are believed to be the main global environmental reservoir for PFOA (Prevedouros *et al.* 2006; Armitage *et al.* 2006; Wania, 2007). PFOA is believed to be transported globally by coupled water-air mechanisms. Findings of PFOA in air samples (particulate phase) and in remote regions as the Arctic confirm the potential of PFOA to get transported over long distances (Jahnke *et al.* 2007a; Shoeib *et al.* 2006; Smithwick *et al.* 2005; Smithwick *et al.* 2006).

No consensus exists in the scientific community regarding transport pathways and environmental fate of PFOA. Although it is clear that both water and air are the means by which substances move globally, there is no common agreement about what is most important.

Several possible explanations of PFOA findings in remote regions are discussed by the scientific community:

- Direct sources during production, use and discharge of PFOA and PFOA-containing products (Armitage *et al.* 2006; Prevedouros *et al.* 2006; Wania, 2007)
- PFOA may be emitted from primary sources in association with particulate matter, and be directly transported long-distances in the atmosphere attached to particles (Simcik, 2005).
- Emissions of the more volatile PFOA precursors (telomers) in industrial areas may be transported by air to the arctic and degraded to PFOA there by atmospheric photolysis of (D'Eon & Mabury 2007; Ellis *et al.* 2004 Martin *et. al* 2006).

Direct sources of PFOA include its application in industry and as impurity in fluoropolymer manufacture, aqueous fire fighting foams, and other industrial products, with total global production estimated to be 4 400-8 000 tons. These may undergo subsequent long-range transport through the atmosphere and oceans.

Direct emissions of PFOA can occur during its production, product use and as product impurities. In 2000 about 20 tons of PFOA was emitted from the largest PFOA production plant in the US which used the electrochemical fluorination (ECF) process. The estimated historical global emissions by industry from PFOA production (1951-2004) are estimated between 400 and 700 tons, with the main part emitted via water (Prevedouros *et al.*, 2006). As a result of the termination of PFOA production using the ECF-based technology, global manufacturing emissions have decreased from 45 tons in 1999 to about 15 tons in 2004 and to an expected 7 tons in 2006 (Prevedouros *et al.*, 2006).

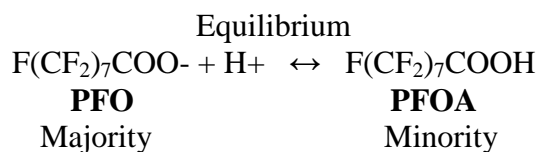
Sewage treatment plants (STP) may be important vectors of diffuse releases of PFCs into the aquatic environment. The particulate phase of the influent contributes significantly to the overall influent concentration of PFCA. STP sludge was shown to contain high amounts of PFOA and STP can serve as point sources for PFC both for the aquatic ecosystem (effluent discharges) and the terrestrial (sewage sludge

application) (Kallenborn *et al.*, 2004; US EPA, 2004; de Voogt and Saez, 2006; Schultz, 2007; Sinclair *et al.*?, 2007). The range of PFOA in leachate water from Norwegian landfills and effluents from sewage plants was 5-115 ng/l and 6-78 ng/l in the particle phase (SFT, 2005).

## 5.1 Transport of PFOA

The different chemical-physical properties of PFOA versus volatile precursors will result in differing transport pattern of the two compound groups. Directly emitted PFOA is expected to dissociate in the environment almost entirely to the ionic PFOA. One author postulated that with its negligible vapour pressure, high water solubility and moderate sorption to solids, accumulation in surface waters and to particulate matter is likely (Mabury, 2004).

Recent studies suggest that the pKa of PFOA at environmentally relevant concentrations is 3.5, one order of magnitude higher than previously reported (European commission, 2006). In addition, the protonated acid has a very high vapour pressure lending it capable of atmospheric transport (Kaiser, 2005). Experiments have shown that PFOA can be measured in the air above an aqueous solution containing PFOA (Ellis, 2006) A recent study investigating emissions from an industrial facility showed that indeed air transport over long distances of particle-bound PFOA directly emitted from a fluoropolymer manufacturing site is possible (Barton, 2007).



### 5.1.1 Local sources

In order to better understand the pathways for transport of ammonium perfluorooctanoate (APFO) from a point source, a focused investigation of environmental media (water and soil) near a fluoropolymer manufacturing production site was undertaken by Davis *et al.* (2007). Environmental media at and near a fluoropolymer manufacturing facility in Parkersburg, West Virginia, USA, which uses APFO, were analysed for perfluorooctanoate. The data from the groundwater flow and air dispersion modelling, the Ohio River sampling program, and the surveying and sampling program indicate that air transport is the most likely transport pathway from point sources to off-site environmental media and that APFO from the point sources is transported via air emissions by wind and is deposited on the well field surface soils. The investigation results further support the hypothesis, or transport conceptual model, in which precipitation then leaches the APFO downward through the unsaturated zone to the aquifer. Dissolved APFO, as perfluorooctanoate, then migrates with groundwater within the aquifer (Davis *et al.* 2007). Additional studies building on this work have also been conducted (Barton 2006; Barton *et al.*? 2007).

### 5.1.2 Ocean currents

Yamashita *et al.* (2005) detected PFOA as the major perfluorinated compound in oceanic waters. Wania (2007) and Armitage *et al.* (2006) have confirmed by ocean transport modelling that oceanic currents are a global transport vehicle for PFOA and likely the major pathway for PFOA transport to Arctic waters.



Kaiser *et al.* (2006) carried out experiments in laboratory scale indicating that perfluorooctanoate is concentrated in the surface foam whether alone in an aqueous solution or with another co-surfactant. This result suggests that marine aerosol transport should be considered as a potentially important transport mechanism. The authors calculated an enrichment factor of up to 3 in foam produced from bubbling through a dilute PFOA solution.

Based on the concentrations measured in open ocean water, it is possible to calculate an annually flux of between 2 and 12 tons of perfluorooctanoic acid (PFOA) to the Arctic (Prevedouros *et al.* 2006). Subsequent refined calculations using the Globpop model confirm this finding (Wania, 2007; Armitage *et al.*, 2006). However, how this or other transport mechanisms contribute to biota that are part of the Arctic food chain originating with water exposure is still to be determined. Alternatively, or perhaps additionally atmospherically deposited PFAS originating from indirect sources such as fluorotelomer alcohols and perfluoroalkyl sulfonamidoalcohols and -amides have been proposed to have a greater influence on the biologically productive near-surface waters (Smithwick *et al.* 2006). In addition, ocean transport of PFOA to the Arctic is highly dependent upon the rate of water, air and water-couple-with-air processes. Ocean water transport alone has been estimated by one researcher to take on the order of decades (Li *et al.* 2006) and a recent modelling study suggests that doubling times of PFCAs in biota may indeed be explainable by ocean transport (Armitage *et al.* 2006). More monitoring and modelling research is needed to be able to elucidate the importance of these potential transport processes.

### 5.1.3 Wet and dry deposition

Air can carry and precipitation deposit particle bound PFOA over considerable distances. The potential for airborne emissions to undergo long-range transport or to be removed from the atmosphere is influenced by their physical-chemical properties. When perfluorooctanoates enter the environment, their physical-chemical properties can vary significantly, depending on whether it exists as acid, salt, or dissociated ions. The most volatile species, the undissociated acid, has a pH-dependent air-water partitioning coefficient ( $K_{aw}$ ). The variability of  $K_{aw}$  with pH influences the potential for vapour formation from aqueous environments and later deposition. Using the pH-dependent  $K_{aw}$ , and measured rain and air concentrations. Barton *et al.* (2007) showed that PFOA exists primarily in the particle phase in ambient air near direct sources of emissions, and that it is efficiently scavenged by rain droplets, making wet deposition an important removal mechanism for emissions originating from either PFOA or APFO. Washout ratios of particle-associated PFOA were determined to be in the same range as other semi-volatile compounds, for which wet deposition is an important mechanism for atmospheric removal and deposition onto soils and water bodies.

Additionally, Scott *et al.* (2006) found PFOA as a major component in precipitation samples across North America. Mega cities like Toronto are possible sources for PFOA in precipitation, shown by frequencies of 100% for PFOA findings in Toronto rain/snow samples.

The results from Scott *et al.* (2006) generally support the hypothesis by Ellis *et al.* (2004) that FTOHs are oxidized, in part, in the atmosphere to ultimately yield PFCAs.

This is also consistent with observations of highest concentrations of polyfluorinated alcohols and –sulfonamides in the North American troposphere found near urban areas. However, the highest concentrations, fluxes, and proportions of PFOA were seen in urban influenced sites and, not as predicted, in more remote sites. The air mass trajectory results also point to highly populated urban areas as the main sources, with low PFOA in the coastal sites associated with air masses coming from the East and southeast Atlantic. However, there was considerable overlap in the air mass directions implying multiple sources and possibly local sources could also have a strong influence on the concentrations (Scott *et al.* 2006).

Ellis *et al.* (2004) estimated, based on prevailing air concentrations, that atmospheric oxidation of FTOHs gives rise to a (C<sub>8</sub>-C<sub>10</sub>) PFCA flux in the range 1-100 t/yr in North America or a deposition of 40-4100 ng/m<sup>2</sup>.

Young *et al.* (2007) investigated high Arctic ice caps to determine seasonal cycles, temporal trends, and atmospheric fluxes of PFOA. They found high fluxes for PFOA, averaging 271 kg/yr, in 2004 and 2005, confirming the estimates by Ellis and co-workers (2004). These fluxes agree with those determined through modelling of FTOH degradation as well (Wallington *et al.* 2006). According to these findings, atmospheric transport and degradation of 8:2 FTOH is the most plausible explanation for the fast response to changes in production and changing concentrations of PFOA in ice cores. Additionally, PFOA levels are too high to be explained by ocean transport. The findings of the non-commercial PFDA and PFUnA in similar concentrations argues against the deposition of direct transported PFOA to the Arctic but for contamination by atmospheric oxidation of precursors as well.

#### 5.1.4 Long-range air transport

Only sparse information is available describing the long-range air transport properties of PFOA, due to its low volatility in the dissociated state. Barber *et al.* (2007) collected air samples from 4 field sites in Europe: rural, semi-rural and urban.

Additionally, air samples were taken from indoor locations in Norway. There are also ongoing outdoor air sampling in Birkenes and Ny Ålesund in Norway but the final results are not available at present.

Perfluorooctanoate was often the predominant compound found in the particulate phase at concentrations ranging from 1–818 pg/m<sup>3</sup>. PFOA was ubiquitous in air samples. The repeated findings of non-volatile ionic PFAS in air samples raises the possibility that they might directly undergo significant atmospheric transport on particles away from source regions (Barber *et al.* 2007). A recent study suggests that direct emissions from a fluoropolymer manufacturing site do have the potential to undergo transport in air over long distances (Barton *et al.*, 2007).

#### 5.1.5 Sediment sorption

Sediment sorption experiments of PFCA, conducted by Higgins & Luthy (2006), were influenced by both sediment-specific and solution-specific parameters. Sediment organic carbon, rather than sediment iron oxide content, was the dominant sediment-parameter affecting sorption, indicating the importance of hydrophobic interactions. However, sorption also increased with increasing solution concentration of the

calcium ion ( $\text{Ca}^{2+}$ ) and decreasing pH, suggesting that electrostatic interactions are also important. Perfluorocarbon chain length was the dominant structural feature influencing sorption, with each  $\text{CF}_2$  moiety contributing 0.50-0.60 log units to the measured distribution coefficients. These data should prove useful for modelling and to evaluate the potential environmental fate of this class of contaminants.

## 5.2 Transport and fate of PFOA precursors

Unlike PFOA, volatile and semi volatile precursors, like 8:2 FTOH, will be distributed in the atmosphere available for long-range transport prior to degradation and deposition as the final product PFOA (Young *et al.* 2007). Both photodegradation, radical initiated oxidation as well as biotransformation of suitable precursor compounds can lead to the formation of PFOA far away from the original emission.

### 5.2.1 Atmospheric transformation

Several studies investigated the chemical reactions leading to PFOA as a final product (Andersen *et al.* 2006; Wallington *et al.* 2006). In general, the 8:2 fluorotelomere alcohol seems to be the most important precursor with a high potential of forming PFOA and PFNA in the atmosphere by radical initiated oxidation (Andersen *et al.* 2006; Wallington *et al.* 2006):

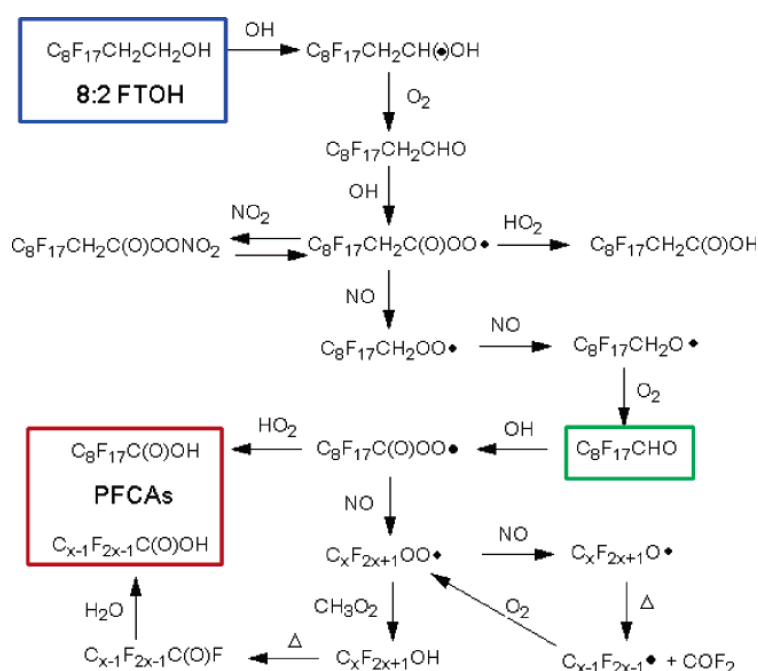


Figure 6: Simplified mechanism for the atmospheric degradation of 8:2 FTOH (blue box) illustrating its conversion into  $\text{C}_8\text{F}_{17}\text{CHO}$  (green box) and the competition between  $\text{NO}$  and either  $\text{HO}_2$  or  $\text{CH}_3\text{O}_2$  radicals that limits the formation of perfluorocarboxylic acids (red box), taken from Wallington *et al.* (2006).

Testing that, Gauthier *et al.* (2005) photodegraded 8:2 fluorotelomere alcohol (8:2 FTOH) in aqueous hydrogen peroxide solutions, synthetic field water systems, and Lake Ontario (Canada) water samples. It was found to undergo indirect photolysis with half-lives of 8:2 FTOH ranging between 0.83 and 163 hours in the different

experiments applied. No significant loss of the parent compound by direct photolysis could be observed. The major monitored products were the 8:2 fluorotelomer aldehyde, the 8:2 fluorotelomer acid (8:2 FTCA), and PFOA.

The results of Gauthier *et al.* (2006), confirm the chemistry model, developed by Wallington *et al.* (2006), indicating that 8:2 FTOH degrades in the atmosphere to yield PFOA and other perfluorocarboxylic acids (PFCAs).

### 5.2.2 Biological transformation

Biologic transformation of precursor compounds in source near regions can result in PFOA as well. Dinglasan *et al.* (2004) examined the aerobic biodegradation of the 8:2 telomer alcohol using a mixed microbial system. The initial measured half-life of the 8:2 FTOH was similar to 0.2 days/mg of initial biomass protein. Telomer acids and PFOA were identified as metabolites during the degradation, the unsaturated telomer acid being the predominant metabolite measured. Biological transformation may be a major degradation pathway for fluorinated telomere alcohols in aquatic systems, followed by transport of the end-products, among others PFOA, via water currents and oceans to remote regions (Dinglasan *et al.* 2004). elaborated further on the biodegradation pathway for 8:2 FTOH in activated sludge for ninety days, achieving mass balance and showing that yield of PFOA were modest 5-10%.

Sinclair *et al.* (2006) found significantly increased mass flows of PFOA after secondary treatment by activated sludge in a waste water treatment plant, in New York State, USA. The observed increase in mass flow of several PFOA may have resulted from biodegradation of precursor compounds such as fluorotelomer alcohols.

In a recent report, D'Eon & Marbury (2007) investigated the potential of polyfluoroalkyl phosphate surfactants (PAPS), nonpolymeric fluorinated surfactants approved for application to food contact paper products, to get metabolized to PFOA by mice. The ingestion of PAPS and the *in vivo* production of perfluorinated acids seems to be linked, stressing the issue of many yet uninvestigated potential PFOA precursors used in considerable amounts in consumer products.

### 5.2.3 Waste incineration

Yamada *et al.* (2005) investigated the fate of a fluorotelomer-based polymer under incineration conditions to determine whether PFOA is formed as a thermal degradation product. The main aim was to investigate the thermal degradation of a fabric treated with a fluorotelomer-based acrylic polymer under laboratory conditions conservatively representing typical combustion conditions of time, temperature, and excess air level in a municipal incinerator. Thermal testing was initiated at 600 °C. The decomposition of the "Telomer" ( $\text{CF}_{2n+1}\text{CH}_2\text{CH}_2\text{-X}$ ) functionality resulted in the formation of compounds containing the  $\bullet\text{CF}_2\text{CH-CH}_2$  fragment in greater amounts with increasing temperature (Yamada *et al.* 2005).

Additionally, the authors report that analysis for PFOA in combustion tests of the treated and untreated article at 1000 °C showed no detectable level of PFOA. It can therefore be concluded that under typical municipal waste incineration conditions no significant quantity of PFOA would be formed from the incineration of a textile or paper substrate treated with a fluorotelomer based acrylic polymer even without

consideration of post-combustion pollution control equipment for acid gas scrubbing in place at municipal incinerators (Yamada *et al.* 2005).

In Norway, combustion of waste is always carried out at temperatures greater than 850°C, ensuring the destruction of PFOA.

#### 5.2.4 Long range air transport

The potential for airborne emissions of precursor compounds to undergo long-range transport or to be removed from the atmosphere is influenced by their physical-chemical properties.

Several studies were carried out in order to investigate the fate of PFOA precursors in remote regions. Shoeb *et al.* (2006) analysed twenty high-volume air samples collected during a crossing of the North Atlantic and Canadian Archipelago in July 2005 and found high concentrations (sum of gas- and particle-phase) of FTOHs; most for 8:2 FTOH (perfluorooctyl ethanol) followed by 10:2 FTOH (perfluorodecyl ethanol) and 6:2 FTOH (perfluorohexyl ethanol).

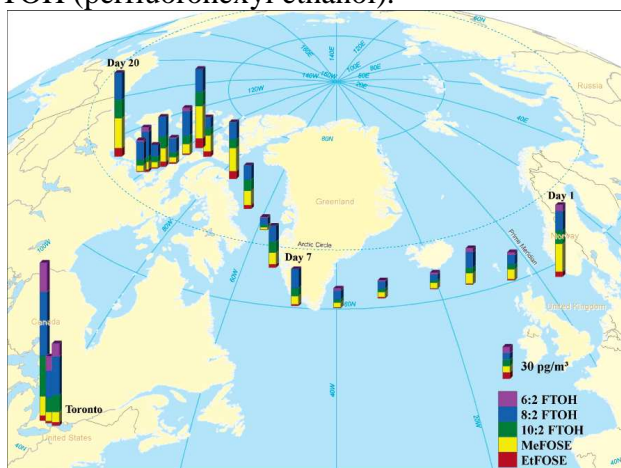


Figure 7: Total air concentrations (sum of gas phase and particle phase) for FTOHs and PFASs across the North Atlantic Ocean and Canadian Archipelago and in Toronto, Canada, taken from (Shoeb *et al.* 2006).

Barber *et al.* (2007) detected 8:2 FTOH and 6:2 FTOH in air samples from 4 field sites in Europe as the prevailing analytes found in the gas phase, at 5–243  $\text{pg}/\text{m}^3$  and 5–189  $\text{pg}/\text{m}^3$ , respectively. These volatile PFCs were ubiquitous in air samples. Concentrations of neutral PFCs were several orders of magnitude higher in indoor air than outdoor air, making homes a likely important diffuse source of PFCs to the atmosphere.

Jahnke *et al.* (2007b) carried out a study on the occurrence of airborne PFAS in the gaseous and particulate phase of German air samples taken at a metropolitan site (Hamburg city centre) as well as in a rural area (Waldhof). Furthermore, they focused on the influence of different ambient temperatures on airborne PFAS concentrations as well as the distribution of analytes between the gaseous and particulate phase and the two sampling sites. A wide distribution of FTOHs, fluorinated sulfonamides, and sulfonamidoethanols (FOSAs/FOSEs) air was found. All of the detected compounds belong to the group of volatile precursor compounds of PFCA/PFOA. A significant correlation was found with the ambient temperature for the partitioning of airborne

FOSEs between the gaseous and particulate phase, whereas FTOHs and FOSAs were almost exclusively found in the gaseous phase.

### 5.3 Transport models for PFOA and volatile precursors

In order to predict and better understand to some extent the extremely complex mechanisms of the transport of PFOA and volatile precursors several recent developed transport models have been applied in global scale in order to predict future exposure and transport of PFOA to remote regions. No study calculated the transport of PFOA to Norway directly, but similar conclusions can be drawn:

Results for the calculation of the globally distribution of PFOA, obtained using the IMPACT 3-D chemistry/transport model analysis are obtained by Wallington et al. (2006) The results suggest that telomers and their degradation products are ubiquitous in the Northern Hemisphere. The sum of 8:2 FTOH and its degradation products are typically  $(0.5-5) \times 10^5$  molecule  $\text{cm}^{-3}$  in remote ocean and Arctic locations in the Northern Hemisphere. This is lower than the concentrations in source regions by just a factor of 5 and is consistent with an atmospheric lifetime of 20-40 days for the group as a whole. Telomer species in the remote Northern Hemisphere in the model are one-third primary 8:2 FTOH, one-third long-lived fluorine-containing aldehydes, and one-third terminal reaction products. PFOA is also ubiquitous in the Northern Hemisphere. Using estimated emissions of FTOHs based on air concentrations and the three-dimensional model, 0.4 t/yr of perfluorooctanoic acid was calculated to be deposited at latitudes north of 65°N via atmospheric oxidation. Concentrations are often higher at remote locations than in source regions, with the highest values occurring over the Atlantic and Pacific Oceans, North Africa, and the Arctic during the summer. This is consistent with the chemistry described in Figure 6. The present modelling results do not prove that the atmospheric oxidation of 8:2 fluorotelomer alcohol is the source of PFCAs observed in remote locations. However, the present modelling results show that with current estimates of chemistry and flux the atmospheric oxidation of 8:2 FTOH can provide a quantitative explanation for the presence of PFCAs in remote regions.

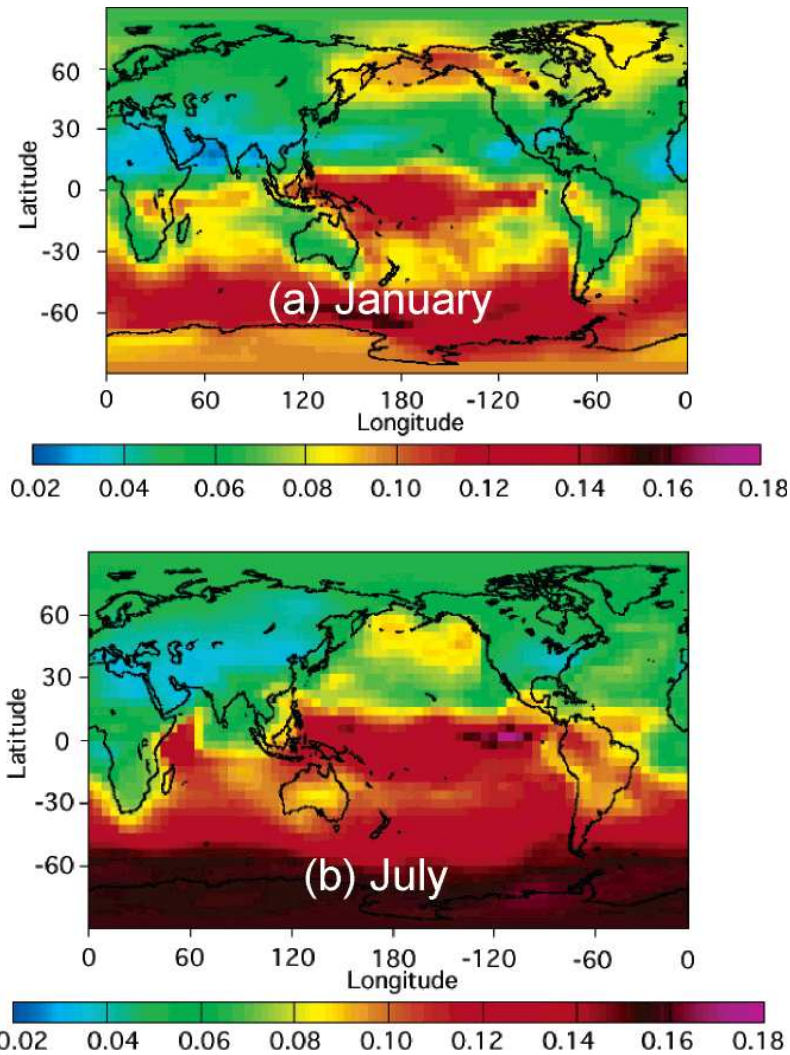
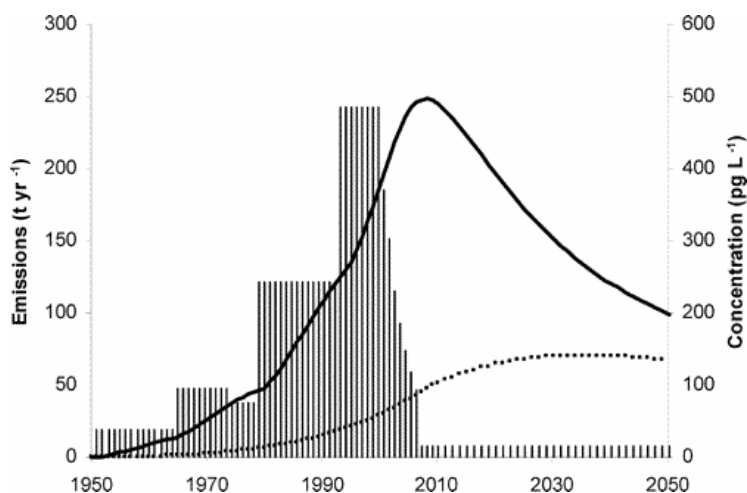


Figure 8: Ratio of PFOA to (PFOA + COF<sub>x</sub>) at 50 m in altitude for (a) January and (b) July. The colour scale extends from 0.02 to 0.18. taken from (Wallington *et al.* 2006).

Armitage *et al.* (2006) estimated that between 2 700 and 5 900 tons of PFO have been emitted between the years 1950 and 2004. The largest single source contribution (~72% of the total) originated from fluoropolymer (FP) manufacturing, followed by ammonium perfluorooctanoate (APFO) manufacturing (~12% of total) and FP dispersion processing (~7% contribution). Direct sources are estimated to be approximately an order of magnitude larger than indirect sources. However, the model analysis indicates that a general pattern of a 20-30 year delayed response to emission reductions will be observed. This pattern is a result of the time required for extremely persistent chemicals such as PFO to redistribute throughout the oceans. Oceanic rates of exchange between northern temperate oceans and the Arctic are likely to be different for the Atlantic and Pacific Oceans which could lead to varying time lags depending on the major oceanic transport routes for PFO to the Arctic. Depending on the scenario considered, a net flux of 8-23 tons per year of PFO was estimated to flow into the Northern Polar zone of the model in 2005. It is noteworthy that these preliminary estimates are 20-60 times greater than the amount estimated to be deposited from the global emission, distribution, and degradation of 8:2 fluorotelomer alcohol. If concentrations in the primary exposure media (i.e., surface water) continue to increase due to direct sources, it follows that concentrations in

wildlife will also continue to increase long after emissions have been drastically reduced or even eliminated.

Figure 9 shows the modelled change in concentration in the ocean water of the northern hemisphere of the model for the simulated Long-Term Fate of PFO from 1950 to 2050 for one representative model scenario. The absolute values vary between scenarios, but the general patterns and features do not. Ocean water concentrations in the Northern Polar zone of the model are estimated to have increased steadily from commencement of emissions in 1951 until the present day with an estimated doubling time between 1975 and 2004 of approximately 7.5-10 years. After 2005, ocean water concentrations in the Northern Temperate, Boreal, and Subtropical model zones are predicted to decline as a result of redistribution to other zones and emission reductions, although there is a time lag of 5-10 years between emission reductions and concentration declines. Modelled concentrations in the Northern Polar zone are projected to increase until about 2030 and then gradually decline as ocean concentrations adjust to lower emission rates and PFO degradation and/or burial processes exert their continued influence (Armitage *et al.* 2006).



*Figure 9: Modelled change in concentration in the ocean water of the northern hemisphere for the period 1950-2050. The scenario illustrated assumes maximum historical emission levels, a mixed mode of entry, negligible degradation (i.e., maximum half-life), and a minimum value of  $K_{OC}$ . Vertical bars represent annual emissions, the solid line represents modelled ocean water concentration in the Northern Temperate zone, and the dotted line represents modelled ocean water concentration in the Northern Polar zone, taken from (Armitage *et al.* 2006).*

In 2007, Wania carried out a simulation with the zonally averaged global fate and transport model Globo-POP, in combination with historical emission estimates for FTOHs and PFOA, to evaluate the relative efficiency and importance of the two transport pathways of direct and indirect PFOA sources (Wania, 2007). The author used a global modelling study that considers and contrasts both transport hypotheses with the objective to evaluate their relative efficiency and importance, building on the model used by Armitage *et al.* (2006). It also needed to be confirmed that reductions of the emissions of volatile precursor compounds would indeed result in fast declines in Arctic seawater concentrations. Estimates of the emission-independent Arctic Contamination Potential reveal that the oceanic transport of directly emitted PFCAs is



more than 10-fold more efficient than the atmospheric degradation of FTOHs in delivering PFCAs to the Arctic, mostly because of the low yield of the reaction. The cumulative historic emissions of FTOHs are lower than those estimated for PFOA alone by a factor of 2-3, further limiting the contribution that precursor oxidation (Wania, 2007).

Armitage and Wania concluded that PFOA from direct emissions to the environment will most probably be transported to the Arctic via ocean currents for several decades ahead, even with a considerable reduced industrial production, potentially giving rise of PFOA levels in wildlife and humans. A similar assumption can be applied for Norway as well.

More temporal and spatial multi-media monitoring of water and biota in food chains along with modelling efforts is needed to fully determine time trends and be able to begin to elucidate the relative contributions of direct and indirect sources and air and water transport pathways.

#### **5.4 Long range transport assessment of PFOA in Norway**

PFOA can reach Norway in different ways.

- i) Long range transported dissolved in ocean waters
- ii) Resolving from ocean surface waters (foam etc.)
- iii) Long range transported via air currents adsorbed to particles
- iv) Atmospheric degradation of 8:2 FTOH

i) and ii) will play a role for the shoreline of Norway alone whilst iii) and iv) will influence the PFOA burden of the terrestrial and the coastal ecosystem.

The SFT commissioned NILU in 2007 to measure among others also PFOA in air samples, investigating long range transport properties (Miljøovervåkning av utvalgte kjemiske forbindelser, P-820). Three samples from two locations were sampled (Ny Ålesund and Birkenes). The results are expected at the end of year 2007 and will give an indication of the contribution of long range transport to the PFOA burden in Norway.

However, data of the air measurements of PFOA cannot distinguish between route ii), iii) and iv). Only an overall assumption will be possible for Norway, based on very rough estimations and very few data points (6 samples from two locations are not representative for the whole country).

Until then only literature data can be used in order to assess the possible PFOA amounts entering Norway via long-range transport.

Using estimated emissions of FTOHs based on air concentrations and the three-dimensional model, 0.4 t/yr of perfluorooctanoic acid was calculated to be deposited at latitudes north of 65°N via atmospheric oxidation. Concentrations are often higher at remote locations than in source regions, with the highest values occurring over the

Atlantic and Pacific Oceans, North Africa, and the Arctic during the summer (Wallington *et al.* 2006).

Armitage *et al.* (2006) estimated that between 2 700 and 5 900 tons of PFO have been emitted between the years 1950 and 2004 (Armitage *et al.* 2006). Direct sources are estimated to be approximately an order of magnitude larger than indirect sources.

According to model calculations and measured background levels of surface ocean water concentrations (e.g., located away from direct sources and coastal regions) >80% of global inventory of PFO is present in the world's oceans in 2005. After 2005, ocean water concentrations in the Northern Temperate, Boreal, and Subtropical model zones are predicted to decline as a result of redistribution to other zones and emission reductions, although there is a time lag of 5-10 years between emission reductions and concentration declines. Regarding Norway however, modelled concentrations in the Northern Polar zone are projected to increase until about 2030 (but to a maximum at much lower levels than in the Northern temperate zone, 150 pg/L and 500 pg/L, respectively) and then gradually decline as ocean concentrations adjust to lower emission rates and PFO degradation and/or burial processes exert their continued influence.

Depending on the scenario considered, a net flux of 8-23 tons per year of PFO was estimated to flow into the Northern Polar zone of the model in 2005.

Estimates of the emission-independent Arctic Contamination Potential reveal that the oceanic transport of directly emitted PFCAs is more than 10-fold more efficient than the atmospheric degradation of FTOHs in delivering PFCAs to the Arctic, mostly because of the low yield of the reaction. The cumulative historic emissions of FTOHs are lower than those estimated for PFOA alone by a factor of 2-3, further limiting the contribution that precursor oxidation (Wania , 2007).

$$\begin{aligned} \text{Long range transported PFOA} &= \text{direct sources} + \text{indirect sources} \\ &= 8\text{-}23 \text{ tons /yr} + 0.4 \text{ tons/ yr} \\ &= 8\text{-}23.4 \text{ tons/yr for the Northern Polar zone} \end{aligned}$$

The Norwegian terrestrial area of the Northern Polar zone correspond to ca 90 000 km<sup>2</sup> plus 40 000 km<sup>2</sup> for Svalbard plus ca. 300 000km<sup>2</sup> ocean area. Together it sums up to ca. 430 000 km<sup>2</sup>.

The Arctic area covers 26 000 000 km<sup>2</sup>. The Norwegian part corresponds to approximately 1.6% of that area. If 8 – 23.4 ton/yr will be transported via long range transport to the Northern Polar zone in total, the Norwegian part would correspond to 0.13 – 0.38 tons of PFOA per year, estimated from data for 2005. We stress that the numbers are illustrative and are subject to large uncertainties.

However, the estimated long range transported amounts are in the same order of magnitude as the estimated direct PFOA emissions.

## 6. Mapping of the use of PFOA in Norway

PFOA is neither manufactured nor used in productions in Norway. The PFOA released into the Norwegian environment is imported either in the form of technical chemical products or as components (impurities) in finished products (e.g. as a constituent in impregnated textiles). The PFOA found in the Norwegian environment may be caused by impurities in compounds, products with content of certain fluorinated substances or by degradation products from certain fluorinated substances (PFOA precursors). Certain perfluorocarboxylic acids (PFCAs) and certain PFAS compounds – all with a chain length of seven or eight may be degraded to PFOA in the environment. The mapping of PFOA in Norway therefore includes the mapping of several hundred different fluorinated substances – all with the potential to degrade to PFOA in the environment.

In this report we have only investigated the substances with the potential to degrade to PFOA. Degradation to higher PFCAs (i.e. longer chains e.g. C<sub>10</sub> or C<sub>12</sub> etc.) has not been included in this study.

In order to map the use of PFOA in Norway, we have used the following sources of information:

- Search in literature and Internet
- Search in the Norwegian Product Register
- Interview with different companies (mainly Norwegian)

The search carried out in the Product Register and the contact to companies is described in more details below.

### 6.1 Search in the Norwegian Product Register for use of PFCA-precursors

In order to find products with a content of PFOA or substances that may potentially degrade to PFOA in the environment, a search has been performed in the Norwegian Product Register for different fluorinated substances.

The search in the Product Register does not identify all products with a content of fluorinated compounds as only chemical compounds that are classified as dangerous should be registered. Articles/products such as impregnated consumer products (clothes, carpets etc.) are not registered in the Product Register. As the fluorinated compounds are not classified as dangerous, the products registered with a content of fluorinated compounds must also contain other substances which are classified as dangerous.

The search in the Product Register can therefore not be used as the only source to products containing fluorinated products that potentially can degrade to PFOA (PFOA precursors). It must be supplemented by information gathered in a different way. The information from the Product Register is therefore with a high degree of probability

incomplete, and the actual import will be much larger. A considerable amount will also be imported to Norway as a constituent in fabricated products, which are not governed by a compulsory registration in the Product Register.

The information gathered from the Product Register was used as a guide to identify products that contains substances that potentially can degrade to PFOA and as a guide to which companies and sectors that were interesting to contact in order to find information about substances that potentially can degrade to PFOA.

### **6.1.1 Identifying substances that potentially can degrade to PFOA (PFOA precursors)**

OECD has in April 2006 (OECD 2006a) published their preliminary list of fluorinated substances, i.e. PFOS, PFAS, PFOA and related compounds and chemicals that may degrade to PFCA. This preliminary list contains about 1,000 CAS numbers in total. It is divided in the following categories:

- Annex 1: List of perfluorooctanesulfonate (PFOS) and related
- Annex 2: List of perfluoroalkylsulfonate (PFAS) and related compounds
- Annex 3: List of perfluorooctanoic acid (PFOA) and related compounds
- Annex 4: List of perfluoro and fluoro chemicals that potentially degrade to perfluorocarboxylic acid (PFCA)

After the start of this project, OECD published in August 2007 “Lists of PFOS, PFAS, PFOA, PFCA, related compounds and chemicals that may degrade to PFCA” (OECD, 2007). The differences between this list and the preliminary lists used in this project for the Product Register Search have not been examined closely.

A list of substances that potentially degrades to PFOA was for the purpose of this project compiled in the following way:

- None of the substances from annex 1 (PFOS) are included since these compounds only can form PFOS which are not within the scope of this study.
- Substances with a chain length of 7 or 8 from annex 2 (PFAS) are included as substances with this chain length potentially can degrade to PFOA. Furthermore, substances that are polymers are included regardless of the chain length as they potentially can degrade to PFOA.
- All substances from annex 3 (PFOA and related compounds) are included.
- Substances with a chain length of 7 or 8 from annex 4 (fluoro chemicals potentially degrading to PFCA) are included as substances with this chain length potentially can degrade to PFOA. Furthermore, substances that are polymers are included regardless of the chain length as they potentially can degrade to PFOA.

This result in a list of 362 substances that are either PFOA related compounds or compounds that potentially may degrade to PFOA because of their chain length. Furthermore, SFT has information about two substances that also are relevant with respect to PFOA. In total a search for 364 substances have therefore been carried out in the Norwegian Product Register.

### **6.1.2 Search in the Norwegian Product Register**

All registered substances in the Norwegian Product Register were compared to the 364 potential PFOA-precursors CAS numbers from the OECD list from April 2006. There were 24 PFOA-precursors from the OECD list that gave a match of CAS numbers, but since most of the information is confidential only a few quantities of these identified precursors were presented from this search. Also the uses of some of the PFOA-precursors found were confidential. Categories of chemical products that were identified in the search as potential sources to PFOA could be identified on their use on the Norwegian market as

- Polish
- Fire fighting foam
- Hydraulic fluids
- Paints and lacquers
- Cleaners
- Waxes

In context to this search described above, a few other product groups were earlier identified but not covered by the Norwegian Product Register since they neither represent chemical products nor are expected to contain any classified chemicals at that time they were registered.

## **6.2 Contact to companies**

To learn more about the amounts used, and thereby the amounts of PFOA or PFC present in the products, it is necessary to contact the most relevant Norwegian companies that either produce, sell or import the relevant products listed above. It was not possible to contact all relevant companies, but the largest companies within each sector have been contacted.

The method used to identify the most relevant companies is described in more details below.

On the basis of knowledge about products that may be sources to PFOA, relevant sectors were identified, and from this a list of relevant Norwegian companies was generated.

### **6.2.1 Identifying sectors and products that may be sources to PFOA**

Several investigations show that fluorinated compounds are used in the products listed in the table below (Poulsen *et al.*, 2005; SFT, 2004; OECD, 2006b; Swedish Chemicals Agency, 2006).

Table 8: List of products that may be sources to PFOA

<b>Product group</b>	<b>Products</b>
Impregnating agents	Impregnating agents for textiles
	Impregnating agents for carpets
	Impregnating agents for leather
Impregnated products	Grease proof paper (for fast food)
	Paper cups
	Microwave popcorn bags
	Water proof clothing
	All-weather clothes
	Skiwear
	Golf clothes (windbreakers, caps)
	Carpets
	Shoes and boots (for children)
	Tents
	Umbrellas
	Bags of leather and textile
	Caps
	Shower curtains
	Furniture
	Table cloths
Degreasing agents	
Cleaning agents (for glass)	
Wax	Wax for cars and skis
Polishing agents (for floorings)	
Paints	
Fire fighting foams	
Electronics	Semiconductors

The relevant Norwegian companies were identified by performing a search in the database called Nordic Business Key by D&B ([www.nordicbusinesskey.com](http://www.nordicbusinesskey.com)). The latest update of the database over the Norwegian companies is from May 2007. The search has been performed for the sectors that were found useful with respect to the above list of products that may be sources to PFOA in Norway.

The search for Norwegian companies within these relevant sectors resulted in 26 945 companies. Most of the companies lies within the fields of wholesale (8 296 companies), retail sail (4 366 companies) and industrial cleaning (5 460 companies).

When all one-man business and field offices were excluded, the search resulted in 11 305 companies in stead. Companies with 10 employees ore more within the above mentioned sectors only account for 1 557 companies. This list was still too long, why the companies within each sectors were sorted after the number of employees (as information of turnover is not available for all companies).

### 6.2.2 Companies to contact

Still having excluded the one-man businesses, more than 11.000 potential relevant Norwegian companies were to be contacted, in order to cover a substantial part of the Norwegian market. Since this is a time consuming and unrealistic task, there was a need for a strategy for a limited but effective contact activity with the aim to find important sources of PFOA in the Norwegian society. This company selection strategy were applied in the following way

- The search in the Norwegian Product Register found important perfluorinated products put into the Norwegian market by companies that trade and use these products. Some of these companies were contacted, if the company information were available and not hindered by secrecy.
- For the prioritization of companies within one sector, number of employees and turnover were considered in order to concentrate efforts on the largest Norwegian companies.
- Companies that have known uses of perfluorinated products through previously conducted studies were also contacted.

All in all, 25 different companies were contacted during the preparation of this project. A list of the contacted companies and their response is found in Appendix 1. As the survey proceeded, it was realised that the method of contacting companies was in many cases unsuccessful. Many companies did not want to give detailed information on this subject. In some cases the companies did not have a knowledge of fluorinated compounds (never heard of this problematic chemical group) or in many other cases, they knew about the compounds, but would not give us any detailed information.

Therefore, this project is mainly based on calculations from statistics supplemented with literature data. Useful information from the interviews is of course added.

### 6.3 Estimations based on statistics

In order to quantify the amounts of PFOA or PFOA precursors in all categories of products mentioned in Table 8 and identified in the Norwegian Product Register, these product categories were processed through statistics from Norwegian Statistics Bureau (SSB) on manufacture, export and import volumes where the net consumption could be calculated through the following formula:

Net consumption of products in Norway (NC) = manufacture + import – export.

Detailed statistics were received from SSB with respect to foreign statistics (import and export) for different product codes. These statistics were given in amounts in kilograms as well as Norwegian currency (NOK) calculated in export prices. Statistics on Norwegian manufacture of products are not on the same detailed level as import and export statistics. Furthermore the production statistics does not necessarily follow the same product codes and only cover 90% of the Norwegian companies in total. In addition, the manufacturing statistics are only available in currency (NOK) meaning that a conversion to the amount (kg) must be made with the assumption of the cost per kg for the manufacture equals the cost per kg for the import/export. Finally, for some areas in manufacture statistics the data are confidential (= non-available). All in all

this means that the calculations based on the statistics must be regarded as rough estimates.

Authentic product information that was received from the interviewed major companies in each relevant sector is expected to be of a higher quality and might support the quantitative information from the statistics. In order to reform calculated net consumption quantities into possible emissions of PFOA, the range of reported calculated amounts and analytical results of PFOA from several products, will be used to assess the quantities of the total amount of PFOA that theoretically could be emitted in Norway.

It is important to note that, this study does not assess the relative importance of consumer articles in comparison to water, food and air to PFOA or PFCA exposure in Norway as insufficient data is available to assess these sources.



## 7. Source information on relevant product groups for PFOA

### 7.1 Impregnation agents

#### 7.1.1 Impregnating agents for textiles

One of the companies on the Norwegian market was interviewed concerning their impregnation products that they sell to non professional consumers in Norway. Mainly two of their products, an aerosol and trigger, are used for impregnating shoes, tents, clothes, furniture etc. The company further mentioned that these two products may be applied on all kinds of textiles and most kinds of leather. The third product is applied during domestic washing of table cloth, sportswear or similar products. These products are sold in various shops and stores where furniture, clothes paints etc are sold. The company declared that they are a small player on the Norwegian market, but are growing steadily in comparison to main players such as Boston, Kiwi, Imprenex, Nikwax and Granger that dominate the Norwegian market. Further they mentioned that they have sold around 30 000 to 40 000 units to the consumer market during 2006 and expect to double that amount during 2007.

The company products were analysed and there were findings of 0,73 mg PFOA/kg and 4 mg PFOA/kg, but there is no correlation available to the amounts in one unit of these products in order to quantify the exposure of PFOA during one year from these particular products.

*Table 9: Amounts of fluorinated substances extracted from several commercial impregnation agent products*

<b>Product name</b>	<b>Sum FTOH (ng/mL)</b>	<b>Sum PFAS (ng/mL)</b>	<b>Sum PFCA (ng/mL)</b>	<b>Sum FTS+ FTCA (ng/mL)</b>	<b>Sum FOSA+ FOSE (ng/mL)</b>
Ecco Universal waterproofing spray	224 310	0	2 783	0	0
Armour	3 480	0	0	0	0
Nikwax TX Direct wash-in	760	0	244	0	0
Boston Raingard all over	585 490	12,1	223	0	0
Kiwi select all protector	664 250	12,5	6 286	58,8	0
Imprenex plus	0	81,3	0	0	0
Nikwax nubuck & mocka proof	0	0	0	0	0
Springyard Waterproofer	1 144 630	22,4	91	0	0
XT	4 649 520	17,7	298	0	0
Boston protector	203 300	0	0	0	0
Nikwax TX Direct Spray-on	0	0	0	0	0
Atsko Waterguard	9 419 980	5,1	1 075	0	0
Collonil classic waterstop	882 250	0	8 372	0	0

Presuming that one unit contains 250 g, the annual sale from this company would be 10000 kg. With a content of maximum 4 mg PFOA/kg this would amount to 40 g PFOA annually. We do not know their market share other than they are small player

on the Norwegian market. If assumed they have between 1-5% of the Norwegian market, then the total annual PFOA content from impregnating agents for textiles sold in Norway would be between 800 g and 4 kg PFOA, assuming that the content of PFOA in the other impregnating agents on the market is the same.

### 7.1.2 Waxes

Different types of waxes exist, for example waxes for cleaning (e.g. car waxes) and ski waxes. Waxes are known to contain fluorinated compounds in small amounts. A search in the Nordic Product Register (SPIN) for PFCA-precursors showed that 29 fluorinated substances are used in the Nordic countries. For half of these substances the uses are confidential, but among the rest, one fluorinated compound is reported to be used in wax in Norway.

DuPont produces fluorinated products to be used in wax. According to DuPont, the fluorotelomer-based surfactants are added to wax in order to improve wetting and levelling (important for spreading and gloss), and re-wetting characteristics (e.g. the second coat is smooth and level). The fluorotelomer-based surfactant product is added in an amount between 100 and 500 mg of product per kg wax. (Washburn et al., 2005)

The search in the Norwegian Product Register on PFCA-precursors showed a consumption of zero for the years 2002 and 2003, and a confidential use for the years 2004, 2005 and 2006. This indicates that waxes used in Norway contain PFCA-precursors.

Swix, a Norwegian producer of ski wax, was contacted. They produce more than 100 tons of ski wax annually, and the main part is based on a fluorine free paraffin wax. They are producing one type of wax based on nearly 100% of a perfluorinated alkane (C<sub>16</sub>F<sub>34</sub> perfluorohexadecane). This specific fluorinated compound is not on OECD's list of substances that can be degraded to PFCA (is actually not on OECD's list at all). This type of wax is very expensive and is therefore only produced in an amount of about 1 tonne yearly. 50% is being exported.

Three other wax producers, which primarily produce car waxes, were contacted. Two of the companies do not use any fluorinated compounds in the waxes at all. The third company gave the impression that this applies to their products as well, but when talking to the main production site in Holland, they would not give any information at all.

Surveys in Denmark and Sweden also indicate that the use of fluorinated substances in wax is a minor area (Jensen *et al.*, 2008; Anfält *et al.*, 2006). The Swedish survey state that fluorinated substances are used in waxes, but not in all types of waxes. Products that maintain hard wax surfaces, and also have a cleaning effect, may however contain fluorinated compounds (Anfält *et al.*, 2006).

The calculation of estimated PFO content in wax is based on statistical data. The calculations are complicated by the fact that production statistics have not been available. Furthermore, the export has for some years been larger than the import, indicating that some production in Norway must occur, even though the production is not known or at least not included in the calculation received from Statistics Norway.

The calculations are therefore solely based on import and export statistics, which means that the net consumption in reality is higher. However, when taking these assumptions into account, the calculated net consumption of wax in Norway for 2004-2006 varies between 930 tons and 1,098 tons.

The consumption of wax is calculated for artificial and synthetic wax. The product codes for wax based on chemically modified lignite and wax based on polyethyleneglycol is not considered to contain fluorinated compounds<sup>1</sup>:

The estimated amount of PFO in wax is calculated by using the information from ENVIRON International Corporation and DuPont of the total content of PFO in consumer products (Washburn *et al.*, 2005). In this report it is stated that the total calculated concentration of PFO per litre Industrial Floor Waxes and Wax Removers is between 0.0005 mg/litre and 0.06 mg/litre. This results in a calculated estimated amount of PFO between 0.6 g and 66 g for 2006, if it is assumed that all wax products contain fluorinated substances.

This is however, not the case. Precisely, how large a percentage of the total wax products in Norway that contains fluorinated substances, is not known, but an educated guess is that the percentage is lower than 50%. This is based on the fact that some car waxes and that most ski waxes probably not contain fluorinated compounds.

When assuming that a maximum of 50% of the waxes in Norway are based on fluorinated substances, the total yearly estimated amount of PFO in waxes in Norway is between 0.3 g and 33 g.

This amount only covers impurities of PFOA in the fluorinated surfactants used in the wax products. As it is not known exactly which kind of fluorinated compounds that are used for waxes, the actual amount could be much higher if fluorinated compounds that can be degraded to PFCAs are used. The search in the Norwegian Product Register carried out in this project indicates that such fluorinated compounds are used for waxes in Norway. The use is however confidential.

### 7.1.3 Polish

Fluorinated substances are known to be used in polish products, e.g. in floor polish. A search in the Nordic Product Register (SPIN) for PFCA-precursors showed that 29 fluorinated substances are used in the Nordic countries. The use areas of half of the substances are confidential, but for the rest of the fluorinated substances, at least one of the substances is used in floor polish and other polish products. A search in the Norwegian Product Register on the same substances shows that the use of these PFCA-precursors in polishes was 20 kg in 2006 (a use of zero before 2006). Only compounds classified in Annex 1 as dangerous must be reported to the Product Register, meaning that the actual use could be much higher.

According to Du Pont<sup>2</sup> almost every acrylic/wax floor polish on the market contains a fluorosurfactant (100 to 500 mg of product per kg). The fluorotelomer-based

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<sup>1</sup> These two product areas are also very small compared to the total import/export of wax (below 10% of the import and below 1% of the export).

<sup>2</sup> DuPont website: [http://www2.dupont.com/Zonyl\\_Foraperle/en\\_US/uses\\_apps/industrial.html](http://www2.dupont.com/Zonyl_Foraperle/en_US/uses_apps/industrial.html)

surfactants are added to floor polish formulations in order to improve wetting and levelling (important for spreading and gloss), and re-wetting characteristics (e.g. the second coat is smooth and level). (Washburn *et al.*, 2005).

Information was received from a polish producing company. A maximum of 50 000 litres of polish is sold annually in Norway. However, no information about the content of fluorinated substances was received.

Another Norwegian polish producer, produces polish products with a content of maximum 0.02% (200 mg/kg) fluorinated surfactants. The fluorinated surfactants in their products have a chain length that is shorter than C<sub>8</sub>. No information was received on the amount of polish they produce and sell in Norway.

No statistics on polish was received from Statistics Norway.

We must therefore rely on the information from the Norwegian Product Register that 20 kg of PFCA-precursors were used in Norwegian polish products in 2006. The potential amount of PFOA will therefore be significantly lower than the 20 kg.

## **7.2 Impregnated products**

### **7.2.1 Paper (food paper, microwave paper, paper cups)**

Nordic paper is the only manufacturer in Norway that produces impregnated paper internationally and for the domestic food industry market in Norway. Nordic Paper has three major plants whereof two are located in Norway namely in Greker and Geithus. Nordic paper declared in a survey in 2006 that they do not apply any perfluorinated substances to their impregnated paper. They also stated that impregnated or coated paper is not manufactured anywhere else in Norway. However Nordic paper declared that there are possibilities that imported impregnated paper that may contain up to 1 kg perfluorinated substances per tonne paper. If we assume, based on import statistics, that all imported impregnated paper is treated with 1 kg perfluorinated substances per tonne paper, this results in an average of 500 to 700 kg in total per year. Unfortunately there is presently no specific data available to quantify the specific PFOA content in these imported impregnated papers.

### **7.2.2 Textiles (out-door clothing, apparel, tarpaulins, tents)**

Some major chemical companies such as Huntsman and Clariant are well known distributors of fluortelomer containing impregnation products for industrial manufacture of working and sports clothes in Norway. These companies provide the majority of products to the Norwegian domestic market. However, the majority of impregnation products are used in the textile producing regions of the world and as such are present in treated articles imported in to Norway. Imports are substantially greater than the volume of textile manufactured domestically in Norway. When interviewing the largest chemical company, they reported that they export 13 000 kg fluortelomer product into the Norwegian textile industry every year. With this quantity they cover around 75% of the Norwegian market. They assumed that the total distribution to the Norwegian textile industry was around 20 tons of fluorotelomer product every year, where an average content of fluortelomer is in average around

18,5 %, which would mean that around 3 700 kg of fluorotelomer active ingredient composed of approximately 50 wt% C<sub>8</sub>-based of acrylate type, would be added to fabrics used for working and sports clothes manufactured in Norway in one year. With ordinary application methods during textile manufacture, this would mean that the finished fabrics would contain around 3,5 %<sup>3</sup> of reacted C<sub>8</sub> based perfluorinated products on an average working and sports clothes manufactured in Norway.

The calculation of estimated PFOA content in impregnated and coated textiles are based on statistical data. The calculations are complicated by the fact that manufacturing statistics are not easily available, as described in the previous chapter. The manufacturing statistics are only available in currency (NOK) meaning that a conversion to the amount (kg) must be made with the assumption of the cost per kg for the production equals the cost per kg for the import/export. However, with these assumptions the calculated net consumption of textiles in Norway for 2004-2006 varies between 5 530 tons and 6 876 tons.

The estimated amount of PFO in impregnated and coated textiles are calculated by using the information from Washburn *et al.*, 2005. In this report it is stated that the total calculated concentration of PFOA per kg textiles is between 0 mg/kg and 0.034 mg/kg. This results in a calculated estimated amount of PFOA between 188 g and 234 g for 2004 and 2006 respectively, if it is assumed that all impregnated and coated textiles from the statistics have a content of fluorinated substances.

### 7.2.3 Foot wear

The domestic Norwegian foot wear industry is dominated by small companies that have their production abroad. They are mainly importers to Norway through own factories or as distributors for foreign foot wear manufacturers. In order to get some information on the foot wear consumption in Norway that has any relevance to use and emissions of fluortelomers and PFOA, statistics from SSB on manufacture, import and export has been analysed. Only foot wear that was made of wood, plastic non water proof were excluded from the statistical assessment.

Since there are no data available concerning possible amounts of fluortelomer products applied from authentic market data, estimated amounts have been used in the calculations as for technical textiles in a range between 0 and 0,034 mg PFO per kg material (Washburn *et al.*, 2005).

It is important to notice that the manufacture statistics from SSB is registered as a substantial part of the whole Norwegian foot wear consumption, which is not the signal given through interviews with some of the major foot wear companies in Norway. The mass data of manufacture was recalculated from Norwegian currency (NOK) export prices over the years 2004 to 2006 to kilograms through advisory information from SSB. With the data available the calculated amounts of PFOA are estimated to 0,5 kg per year. Due to uncertainties in volumes and market data, there

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<sup>3</sup> An estimate of an ordinary applied impregnation product for textiles on the Norwegian market is done as follows: A recipe in textile production contain 50g/l of fluortelomer product. Pick-up is estimated to maximum 70%. Since the density of the impregnation bath is close to 1 kg/dm<sup>3</sup> then for 1kg finished textile the amount "dry" impregnation product is: 1 x 0,70 x 1 x 0,050 = 0,035 kg = 3,5 %

are good reasons to view these data with critical eyes but at this point it is the best assessment available to us.

#### 7.2.4 Carpets

Carpets are considered as an important source for fluorinated compounds in society. This has been verified through findings of fluorinated compounds (PFOS and PFOA) vacuum cleaner dust in private households and offices could be an indication of use of fluorinated compounds in carpets, but other products in the homes are of course also possible sources (Moriwaki *et al.* 2003).

The manufacture of carpets in Norway is believed to cover only 5% of the total Norwegian consumption of carpets, so the main flow is imported. There are two main categories of carpets on the Norwegian market, one category is made of wool and similar natural fibres and the other category is made of synthetic or man-made fibres.

A substantial part of the carpets from synthetic fibres are impregnated with fluorinated compounds during manufacturing. Carpets made of natural fibres such as wool and similar natural fibres are according to industry<sup>4</sup> normally not impregnated with these compounds. According to an earlier SFT study of the use of PFAS substances in Norway, 5-10% of the wool carpets are impregnated, but none of the Norwegian producers are impregnating their wool carpets. A few customers ask for an impregnation of the wool carpets. In these cases Teflon® is used. (SFT, 2004).

Impregnating products used for carpets are for example Zonyl® and Teflon® products from DuPont, Baygard® products from Bayer, or Scotchgard® products from 3M. However, which fluorinated products that are used in Norway is not investigated in this project.

The net consumption of carpets in kilos in Norway is found by using statistics from SSB, Norway. The calculations are based on the amounts from the SSB statistics of imported synthetic carpets, which is the main stream of consumed carpets in Norway. According to ENVIRON International Corporation and DuPont (Washburn *et al.*, 2005), the total calculated concentration of PFO per kg carpet fibres is between 0.2 mg/kg and 2 mg/kg. When assuming that 100% of the synthetic carpets are impregnated with fluorinated compounds, the calculated estimated amount of PFO is between 1.5 and 20 kg in carpets per year.

### 7.3 Paint and lacquer

Paints, lacquers and varnishes are known to contain fluorinated compounds. According to DuPont the fluorotelomer-based surfactants are added to paints to

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<sup>4</sup> Oral information from carpet industry in Denmark.

improve flow, wetting and levelling, and for reducing the surface defects and craters. The fluorotelomer-based products are added in an amount between 300 and 500 mg of product/kg of paints. (Washburn *et al.*, 2005).

A search in the Nordic Product Register (SPIN) for PFCA-precursors shows that 29 fluorinated substances are used in the Nordic countries. For half of the fluorinated substances, the use areas are confidential, but between the rest (15 substances), six fluorinated compounds are used within paint, lacquers and varnishes.

DuPont produce a range of Zonyl products for paint and lacquer systems, such as aqueous systems and printing inks. Going through the available MSDS of the Zonyl products<sup>5</sup> for paints and coatings, shows that the fluorinated compounds in general are fluorotelomers with a multiple chain length (n).

The search in the Norwegian Product Register for PFCA-precursors showed a consumption of zero for all years (2002 to 2006). This number does, however, not necessarily mean that no fluorinated substances are used, as only compounds classified as dangerous should be reported to the Norwegian Product Register.

Three Norwegian paint producers were contacted. According to the largest producer of paint in Norway, fluorinated substances are not used in their water-based paints (70% of the market). Some kind of Teflon wax is used in their powder coatings, but no detailed information was received. Another company informed that they are only using fluorinated substances in their marine coatings. The fluorinated substances are used for painting the bottom of the ships, as the very low surface tension of the fluorine agent has the advantage that nothing sticks to the bottom of the ships. They use a siliconefluoropolymer for this purpose. Less than 5 000 litres of this paint product is sold yearly in Norway. No information about the percentages of the content of the siliconefluoropolymer was received.

The calculation of estimated PFOA content in paints and lacquers are based on statistical data. The calculations are complicated by the fact that manufacturing statistics are not easily available, as described in the previous chapter. The manufacturing statistics are only available in currency (NOK) meaning that a conversion to the amount (kg) must be made with the assumption of the cost per kg for the production equals the cost per kg for the import/export. However, with these assumptions the calculated net consumption of paint in Norway for 2004-2006 varies between 96 071 tons and 108 564 tons.

The consumption of paints is calculated for the entire product group of paint, varnishes, and printing inks.

The estimated amount of PFOA in paints and varnishes are calculated by using the information from ENVIRON International Corporation and DuPont of the total content of PFOA in consumer products (Washburn *et al.*, 2005). In this report it is stated that the total calculated concentration of PFOA per litre Latex paint (water-based paints) is between 0.02 mg/litre and 0.08 mg/litre. This results in a calculated

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<sup>5</sup> Can be found on <http://msds.dupont.com/NASApp/msds/Mediator> by searching after German MSDS's for Zonyl products, that for some reason contains more information about the fluorinated substances used in the products.

estimated amount of PFOA between 2.1 kg and 8.7 kg for 2006, if it is assumed that all paint products have a content of fluorinated substances.

This is however, not the case. Precisely, how large a percentage of the total paint products in Norway that contain fluorinated substances, is not known, but an educated guess is that it is lower than 20%. This is based on the fact that 70% of the Norwegian production of water-based paints does not contain fluorinated substances. According to SSB (statistics Norway) 44% of the paint and varnishes are imported from Sweden and 18% from Denmark. The Swedish Chemicals Agency has carried out a survey of perfluorinated substances and their uses in Sweden. Paint and varnishes are not treated in details – the report just mentions that products such as paint and varnishes are known to contain fluorinated compounds. The report also mentions that paint and varnish is one of many minor areas (such as fire fighting foams, hydraulic oils, wetting agents, lubricants etc.) that together account for approximately 10% (2.4 tons) of the use of perfluorinated substances. (Anfält *et al.*, 2006). Similarly, a survey of the use of fluorinated substances in Denmark has been carried out. This survey shows that about 20% (3.5 tons) of the use of fluorinated substances in Denmark are used within paint and varnishes alone (according to the Product Register). However, when the Danish trade organisation for paint and varnishes asked their members, this resulted in an actual amount which was much lower, as most paint producers in Denmark do not use fluorinated substances.

When assuming that a maximum of 20% of the paint and varnish products in Norway are based on fluorinated substances, the total yearly estimated amount of PFO in paint and varnishes in Norway is between 0.4 kg and 1.7 kg.

This amount only covers impurities of PFOA in the fluorinated surfactants used in paint and varnish products. As it is not known exactly which kind of fluorinated compounds that are used for paint and varnish, the actual amount could be much higher if fluorinated compounds that can be degraded to PFCAs are used. However, the search in the Norwegian Product Register carried out in this project – only on relevant fluorinated substances, that can be degraded to PFCAs, indicate that no such fluorinated compounds are used within paint and varnish products in Norway.

## 7.4 Fire fighting foams

There are several different types of fire fighting foams on the market that both are fluorine free and those that contain fluoroprotein substances such as polyfluoralkyl betaine substances.

Fire fighting foams, the so called AFFF based on PFOS, have been used since the early 1970's to today but do not dominate the market anymore due to restrictions that came in force both in USA and EU. The major global manufacturer, 3M halted manufacture in 2002, but other suppliers remain. Recently, the EU has implemented a use ban for PFOS-based substances which has resulted in a new generation of fire fighting foams mainly based on C<sub>6</sub>-fluortelomers, where C<sub>8</sub>-fluortelomers only occur in trace levels as impurities. This means that the new generation of fire fighting foams, both for domestic and professional use only might contain traces of PFOA.



The C<sub>6</sub>-fluorotelomer products used in fire fighting foams cannot degrade to PFOA in the environment since the main components are not based on C<sub>8</sub> chemistry. The degradation products may include 6:2 fluorotelomer sulfonates.

The largest supplier of fire fighting in Norway, NOHA Norway AS, reported through one of their major suppliers of fire fighting foam concentrates that perfluorinated fire fighting foams are not used in consumer products at all but only specialized industrial products that are handled by specialist fire-fighters. Fluorotelomers used in fire fighting foam concentrates have trace amounts of PFOA created as a by-product of the fluorotelomer manufacturing processes, but since no fire fighting foam production occur in Norway these emissions that occur during production are of no relevance for emissions of PFOA in Norway.

The distributors of fire fighting foams did not provide typical amounts, sales volumes and suppliers since this is confidential information and therefore it is not possible to quantify emitted trace amounts of PFOA from use of fire fighting foams in Norway.

#### **7.4.1 Oil and gas industry**

The oil and gas industry is one of the largest industrial sectors in Norway. Statoil, and since 1<sup>st</sup> of October Statoil/Hydro, is one of the largest users of fire fighting foams in Norway. Through our contact, Statoil did their own search among 7000 chemical products that they use. In total their chemical products contain around 6000 individual substances. They got two hits on the OECD list from April 2006, where CAS no. 70969-47-0 is a "fluoroalkyl non ionic surfactant" (an fluoro amine compound) and CAS no. 34455-29-3 is a polyfluoroalkyl betaine (a fluoro sulfonate/sulfonamide/sulfonyl compound). Furthermore, Statoil use polyfluoroalkyl betaine, which is a component in fire fighting foams.

### **7.5 Other product categories**

#### **7.5.1 Herbicides**

The Norwegian Food Safety Authority is a governmental body that is responsible for healthy and safe food and safe drinking water. Among these responsibilities the Norwegian Food Safety Authority keep statistics of pesticides used in Norway. A recent search in their database initiated by SFT on CAS numbers from the OECD report from 2006 gave a hit on the following CAS numbers: 68412-68-0 or 68412-69-0. Based on statistics of the use of these possible imported C<sub>8</sub>-precurcor-substances, around 65 kg were emitted into the Norwegian environment 2006 according to the Norwegian Food Safety Authority. They also mentioned that there are only slight variations of these herbicides so looking back a number of years before 2006 we might expect similar amounts of emissions of these substances some years back in time.

## 8. Results: Emissions, sources and trends in Norway

### 8.1 Previous results; PFAS emissions in Norway (2002 - 2006)

SFT carried out a study regarding the consumption of PFOS-related substances and other perfluoroalkylated substances in Norway in 2003 (Material flow analysis) (SFT, 2004).

The report concluded with:

All PFCs sold in Norway is imported, either as technical product or as part of a consumer product. The dominating field of PFACs use reported was in fire fighting foams. PFOA is not used in fire fighting foams, and will not be distributed in the environment via this route except as an impurity.

The remaining fields of usage involving PFCs were listed with consumption per year of (tons, 2002 numbers):

Impregnation of textiles	7 – 10
Impregnation of carpets	0.4
Impregnation products for home use	0.2
Floor polish and wax	0.2

The amount of PFOA in these applications is unknown.

The SFT reports estimated PFCs emissions in Norway in 2004 as (SFT, 2004):

- Annual emission from offshore installations in the North Sea 7 – 8 tons.
- Annual emission to water/sediment from airports, industry and the like 7 – 8 tons
- Waste < 1 ton

In addition, the SFT commissioned a study on emissions of PFCs from PTFE-coating industry that exist in Norway. During autumn 2006, emissions were analyzed for PFOA at following plants by NILU (SFT, 2007):

- Norwegian Coating Technology (NCT), Notodden
- Iittala, Moss
- Belegningsteknikk, Drammen
- Otto Olsen, Lillestrøm

Both particle bound PFCs and volatile PFCs were analysed, representing the only study done on air emissions of PFOA in Norway.

PFOA was the dominating compound in almost all particle samples. The highest PFOA-concentration was found at Iittala in Moss (115 to 320 ng/Nm<sup>3</sup>). The annual emission for this plant was calculated to an amount of around 5 g/year.

Table 10: Concentration (ng/Nm<sup>3</sup>) of ionic PFCs in air filter (SFT, 2007).

Bedrift	Punkt	ng/Nm <sup>3</sup>											Avgass- mengde Nm <sup>3</sup> /h <sub>101</sub>	g/år
		PFHxA	PFHpA	PFOA	PFNA	PFDCa	PFUnA	PFHxS	PFOS	PFDCs	PFOSA	6:2FTS		
NCT, Notodden	Sprøyteb.	0,00	0,00	0,00	0,00	0,00	0,00	0,06	1,22	0,00	0,00	0,00	22300	0,0
	Sprøyteb.	0,00	0,00	0,00	0,00	0,00	0,00	0,03	0,47	0,00	0,00	0,00	22300	0,0
	Ovn 1	0,00	0,00	0,55	0,00	0,00	0,00	0,01	0,11	0,01	0,00	0,29	484	0,0
Iittala, Moss	Pipe 1	0,00	1,73	115	0,00	0,00	0,00	0,00	0,05	0,00	0,00	0,00	2880	1,2
	Pipe 2	0,00	2,43	137	0,00	0,00	0,00	0,00	0,02	0,01	0,00	0,00	1890	0,9
	Pipe 3	0,71	8,62	320	1,60	0,00	0,65	0,00	0,03	0,00	0,02	0,00	2270	2,6
Belegnings- teknikk, Drammen	Sprøyteb.	0,00	0,69	109	0,00	0,00	0,00	0,00	0,03	0,00	0,00	0,05	6590	2,5
	Innbr.ovn	0,00	0,00	8,02	0,00	0,00	0,00	0,00	0,00	0,00	0,00	2,68	263	0,0
	Sprøyteb.	0,00	0,00	2,03	0,00	0,00	0,00	0,00	0,01	0,01	0,00	0,13	6590	0,0
Otto Olsen, Lillestrøm	Sprøyteb.	0,00	0,00	3,26	0,00	0,00	0,00	0,00	0,11	0,00	0,00	0,00	6640	0,1
	Sprøyteb.	0,00	0,00	15,5	0,00	0,00	0,00	0,00	0,26	0,00	0,00	0,00	6620	0,4
	Innbr.ovn	0,00	0,00	14,2	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	440	0,0

The total emitted amount of PFOA from all four investigated plants together was estimated to be 7 to 8 g/a.

Table 11: Concentration (ng/l) of ionic PFCAs in coating products sampled at Iittala (Primer &amp; Topcoat, Greblon, Weilburger Coatings GmbH)

Prøve		ng/L				
		PFHxA	PFHpA	PFOA	PFNA	PFDCa
13	Primer (resirkulert)	0,68	4,03	42,0	0,48	0,00
14	Top Coat (resirkulert)	0,05	1,81	48,6	0,37	0,00
15	Primer (Produkt)	0,00	6,80	427	0,00	0,00
16	Top Coat (Produkt)	18,7	355	1 591	15,9	20,2

Coating products produced/applied at Iittala (Primer & Topcoat, Greblon, Weilburger Coatings GmbH) were analysed for ionic PFCAs as well. Again PFOA dominates the product composition with concentrations up to 1.6 µg/l coating product leading to an elevated emission potential for this compound.

## **8.2 Norwegian industrial sources of PFOA in 2007**

The data given in table 12 reflect maximum data when using data from SSB in calculations of estimates. Maximum values were used in order to give a conservative assessment on the situation and compared with the reported amounts of applied fluorinated compounds, based on interviews with companies/authorities.

Table 12: Summary consumption of fluortelomers in consumer products and from industrial production based on statistics and interviews with companies, associations and authorities in kg/a.

Article/preparation	No. of companies contacted	Reported amounts of applied fluorinated compounds #	Calculated maximum amounts "consumed" PFO *	Comments
<b><i>Impregnated products</i></b>				
Textiles, work and sportswear manufactured in Norway	2	3700 kg of C <sub>8</sub> -fluortelomer product	No data available	Industrial use where approximately 99% of the C <sub>8</sub> -fluortelomer remain chemically bound to the fabric. <sup>6</sup>
Textiles, impregnated clothes for outdoor use.		No data available	0.19 to 0.24	
Foot wear	3	No data available	0.5	There is less than 5% manufacture in Norway and therefore the amounts of fluorinated compounds reported are based on import- statistics and interviews
Impregnated/coated paper	3	Approx. 700	No data available	There is no manufacture in Norway and therefore the amounts of fluorinated compounds reported are based on import- statistics and interviews
Carpets	2 (outside Norway)	No data available	18 to 20	There is less than 5% manufacture in Norway and therefore the amounts of fluorinated compounds reported are based on import- statistics and interviews
<b><i>Cleaning agents</i></b>				
Waxes	4	None identified (about 1000 kg of a C <sub>16</sub> F <sub>34</sub> )	A max. of 0.033 kg per year +	According to Product Register, PFCA substances are used within wax, but the use

<sup>6</sup> Järnberg&Posner et.al (2004)

Table 12: Summary consumption of fluortelomers in consumer products and from industrial production based on statistics and interviews with companies, associations and authorities in kg/a.

Article/preparation	No. of companies contacted	Reported amounts of applied fluorinated compounds #	Calculated maximum amounts "consumed" PFO *	Comments
		compound identified).	confidential use.	is confidential.
Polish	2	No information from companies. Product Register: 20 kg of fluorinated compounds	No data available	Information from the Norwegian Product Register that 20 kg of fluorinated compounds that can be degraded to PFCAs are used in Norwegian polish products.
<b><i>Chemical preparation containing fluortelomers</i></b>				
Fire fighting foams	4	Main components are C6-fluortelomers and therefore not relevant in this study.	No data available	Traces of C8-fluortelomer impurities may occur
Paint and lacquer	4	0 kg according to the Product Register Is used in powder coatings and marine coatings, but the amounts are unknown.	1.5 to 1.7 kg per year	
Impregnation agents	1	No data available	0.8-4 kg per year	
Herbicides	1**	65 kg per year	No data available	

# : based on interviews with companies/authorities

\* : based on statistics from SSB between 2004 and 2006

\*\* The Norwegian Food Safety Authority

After compiling the answers given in the questionnaires/interviews by the companies, two conclusions can be drawn:

- data from SSB can supply missing information but can give a contradicting picture
- no data on use of PFOA were available for textiles (impregnated clothes for outdoor use), foot wear, carpets, impregnation agents

Detailed statistics were received from SSB with respect to foreign statistics (import and export) for different product codes. These statistics were given in amounts in kilograms as well as Norwegian currency (NOK) calculated in export prices. Statistics on Norwegian manufacture of products are not on the same detailed level as import and export statistics. Furthermore the production statistics does not necessarily follow the same product codes and only cover 90% of the Norwegian companies in total. In addition, the manufacturing statistics are only available in currency (NOK) meaning that a conversion to the amount (kg) must be made with the assumption of the cost per kg for the manufacture equals the cost per kg for the import/export. Finally, for some areas in manufacture statistics the data are confidential (= non-available). All in all this means that the calculations based on the statistics must be regarded rough estimates.

Confidentiality or the lack of knowledge about the identity of applied chemicals can lead to wrong interpretation of the data. Often only the amount of used fluorinated compounds is known to the company, but not the exact content of the mixture or the composition. As a result, for calculating annual use of PFOA containing products, SSB numbers were used. If no data were available from SSB on one line of business, answers from respective company-interviews were used.

### **8.3 Norwegian consumer product sources of PFOA in 2007**

PFO treated carpets represent the largest group of consumer products circulating on the Norwegian marked.

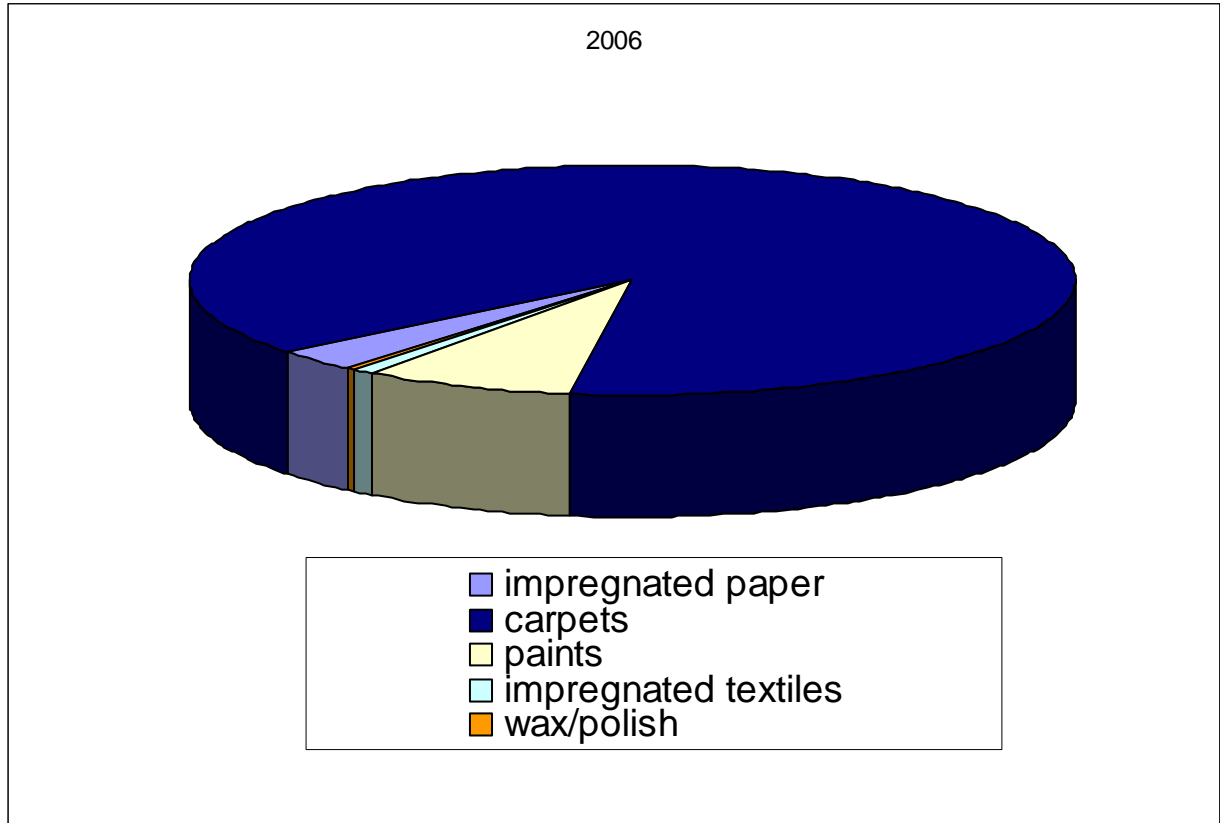
Numbers reported to the project team by the paper industry reflect only the use of fluorinated substances on imported paper, with no details on PFOA content in the applications. No PFOA is used in papercoatings produced in Norway according to the companies.

The portion imported paper on the Norwegian marked in 2006 was calculated by :

$$732489 \text{ kg import} / (732 \text{ 489 kg} + 30 \text{ 000 000 kg produced} - 467 \text{ 126 kg export})$$

As a result, 2.5% of all paper used in Norway is imported. Not 100% of that portion will be treated with fluorinated compounds. 732 kg fluorinated compounds were reported for imported paper for the year 2006. By using the conventional assumption of a 0.1 % PFOA content, 0.732 kg PFOA would have entered the Norwegian market via this route. Since none of the treated paper is produced in Norway (everything is imported to Norway), the only routes of potential PFO emission would be storage, use and discharge of the paper products.

Paints, impregnated textiles and wax applications play only a minor role in PFOA consumption in Norway. No data were reported for shoe- and impregnating agent production/application.



*Figure 10: Contribution to PFOA consumption in Norway in 2006 according SSB and answers to questionnaires by PFOA consuming industry; numbers assuming a 0.1% content of PFOA in the fluorinated compounds applied on paper*



Paints, impregnated textiles and wax applications play only a minor role in PFO consumption in Norway. No data were reported for shoe- and impregnating agent production/application.

*Table 13: Maximal PFO consumption in production of consumer products in 2006 (kg) by SSB ; \*: data for 0.1 % content PFOA in fluorotelomer products used*

*\*\* : not including possible impurities by applications similar as stated by Huntsman*

Consumer product	PFOA consumption in 2006 (kg) based on statistics calculations
Impregnated paper (import)	0.73*
Carpets	20.2
Paints	1.74
Impregnated textiles	0.23**
Wax/polish	0.03

*\*: data for 0.1 % content PFOA in fluorotelomer products used*

*\*\* : not including possible impurities by applications similar as stated by Huntsman*

The data reported by SFT in 2004 (2002 data), reflect the overall PFAS consumption in Norway with no possibility to distinguish between PFOA consumption and other PFAS and are therefore much higher than the data reported now. Paper coating was not mentioned in 2004, but impregnated textiles were stated as the major consumer of PFAS (7-10 tons/a), only a minor PFOA contributor in the current report. However, in the questionnaires the company Huntsman stated the import of 3 700 kg C8-fluorotelomer compounds, applied to work wear, to Norway each year. PFO can be a considerable part of that amount as an impurity, without being part of the data received from SSB.

## 8.4 Emissions of PFOA in Norway, 2007

Several sources for PFOA emissions in Norway were identified in the here presented study:

- Industrial emissions during application of PFOA containing products
- Use of PFOA containing consumer products
- Sewage and landfill related environmental release of PFOA
- Long range transport of PFOA (e.g., ocean currents, atmosphere)
- Source near atmospheric transport followed by wet/dry deposition

With its negligible vapour pressure, high water solubility and moderate sorption to solids, accumulation of PFOA in surface waters is likely (Mabury, 2004). In general, PFOA containing products will most likely emit PFO to the air adsorbed to particles or to the water due to the high water solubility of the compound.

### 8.4.1 Sewage and landfill related environmental release of PFOA

Already under the Nordic PFAS screening in 2004, sewage and landfill was identified as major anthropogenic sources based on measurements of PFOA concentrations in sewage sludge, sewage effluent samples and landfill effluents. In the analysed Norwegian effluent samples, PFOA was the predominant PFAS constituent. The concentration levels found for the two sample sets of landfill effluent are considerably higher than the levels for sewage

effluents (around 10 x higher, 200 ng/L) confirming the significant contribution of landfill effluent water as major anthropogenic source for PFAS in Nordic countries. PFOA was the predominant PFAS constituent in all landfill effluents measured (Kallenborn, 2004). In addition, STP sludge was shown to contain high amounts of PFOA and STP can serve as point sources for PFOA both for the aquatic ecosystem (effluent discharges) and the terrestrial (sewage sludge application) (US EPA, 2004);

in 2004 by SFT, PFOA was found in elevated concentrations in cleaned landfill and STP effluents and sediments, mainly as dominating compound, confirming STP and landfill effluents as main emission source for PFOA in Norway. No difference in PFOA concentrations could be observed between cleaned and non-cleaned effluents, indicating failing cleaning procedures, mainly the inefficiency of the sedimentation process used (SFT, 2005).

The interquartile range of PFOA in leachate water from landfills and effluents from sewage plants was 5–115 ng/l, and 5,8 to 78 ng/l in the particle phase.

The observed concentrations of sum PFAS in natural surface waters (lakes and rivers) revealed an interquartile range of 1–4 ng/l. There was no sign of any significant point sources of PFAS to any of the studied freshwater sites, and the concentrations were in general low as compared to those from other industrialized countries.

The observed concentrations of sum PFAS in sediments from landfills and in sewage sludge revealed an interquartile range of 2–4 ng/g d.w., and the highest concentrations (about 24 ng/g d.w.) were found in leachate sediments from a landfill. PFOS and PFOA dominated among the different perfluorinated alkylated substances.

The calculated interquartile range of sum PFAS in freshwater sediments was 0.1–0.5 ng/g d.w., and the highest concentration was about 1 ng/g d.w. Although there are few comparable data, the reported concentrations seem low compared to data from other industrialized countries.

The interquartile range of sum PFAS in marine sediments was 0.5–1,5 ng/g d.w., and the highest concentration was about 6 ng/g d.w. (SFT, 2005).

Due to the lack of data about yearly amounts/volumes of all landfill and STP effluents and sludge emitted in the Norwegian environment, annual amounts of PFOA emitted via this route cannot be calculated.

#### **8.4.2 Emission of PFOA from consumer products from the Norwegian marked**

##### Paper

Since paper-treatment products containing perfluorinated compounds are believed not to be in use anymore, only fluorotelomer-type treatment, containing trace amounts of PFOA (0.1%) is considered in this report. Begley *et al.* (2005), reported that the raw material for paper coating contains 88-160 µg PFOA/ kg product. The author used a dilution factor of 300 for the application of the raw material to the finished product, resulting in an average of 1 860 µg/kg or <0,0002 % PFOA in paper and cardboard.

The SSB reported 732 489 kg imported coated paper into Norway for the year 2006. When using the same assumptions as Begley et al. (2006), 1.36 kg PFOA enters Norway applied on paper each year and with increasing trend.

PFC-coated or impregnated paper, mainly from food packages, will most likely be deposited on landfills which would probably dissolve free bound PFOA into the water systems. If the paper is incinerated in waste incinerators nonpolar volatile PFCs will evaporate to the air and adsorbed to air bound particles. Some PFC will probably migrate into food and indirectly enter STPs through food consumption.

#### Textiles and carpets

According to the studies carried out by SFT and Friends of the Earth in 2006, PFOA concentrations varied between 2 and 171 µg/kg textile in the SFT study and up to 416 µg/kg textiles in the Friends of the Earth study (Schulze & Norin, 2007; SFT, 2006). The average PFOA concentration in both studies was 64.7 µg/kg and 157 µg/kg respectively. The SSB reports a net consumption of 6 876 257 kg impregnated textiles in 2006, corresponding to 0.445 kg pure PFOA for the SFT study and 1.08 kg pure PFOA for the Friends of the Earth study.

Carpets have not yet been analysed for PFOA in Norway but according to information from DuPont, the PFOA concentrations in factory treated carpets are about 5 times higher and for home treated carpets 25 times higher than in treated clothing (Horowitz, 2007). Carpet care solutions are assumed to add 500 µg/kg solution to the PFOA burden of carpet. Additionally, since carpet is used in much higher amounts than clothing (10 081 tons carpets compared to 6 876 tons clothing in 2006) between 10 and 50 times higher amounts of PFOA enter the Norwegian market applied on carpets compared to textiles.

Depending on the handling of these textiles during use for washing and discharge as waste to either landfill or incineration unbound water soluble PFOA or telomer, will be emitted to water and sewage treatment plants, and nonpolar volatile PFCs will evaporate to the air and to adsorbed to air bound particles.

## **8.5 Trends of PFOA use in Norway**

As shown in figure 11, the use of PFOA in Norway increased between the years 2002 and 2007 based on our assessment. It must, however, be noted, that the trends of the PFOA use in Norway solely is based on trends in statistics of products containing PFOA (and precursors?) from SSB for the years 2002 to 2007. We have not received any information in trends on the use of PFOA or PFOA-precursors from companies. The trends therefore indicate an increase or fall in the consumption of the different product groups. For the calculations we have used the following maximum amounts of PFOA in the products (Environ 2006):

0,06 mg PFOA/liter solution

Industrial Floor Waxes and Wax removers

0.034 mg PFOA per kg technical textile of fluortelomerproduct

0,08 mg PFOA/liter paint

Latex paint (water based paints)

2 mg PFOA/kg carpet fibre

Fluortelomerbased product - Mill treated carpeting

1000 mg fluortelomer/kg coated paper

Mainly the use of PFOA in imported paper increased considerable with about 150% during the last six years, but on low levels following the assumptions described above. The use of PFO in carpets increased as well with about 60% during the same period.

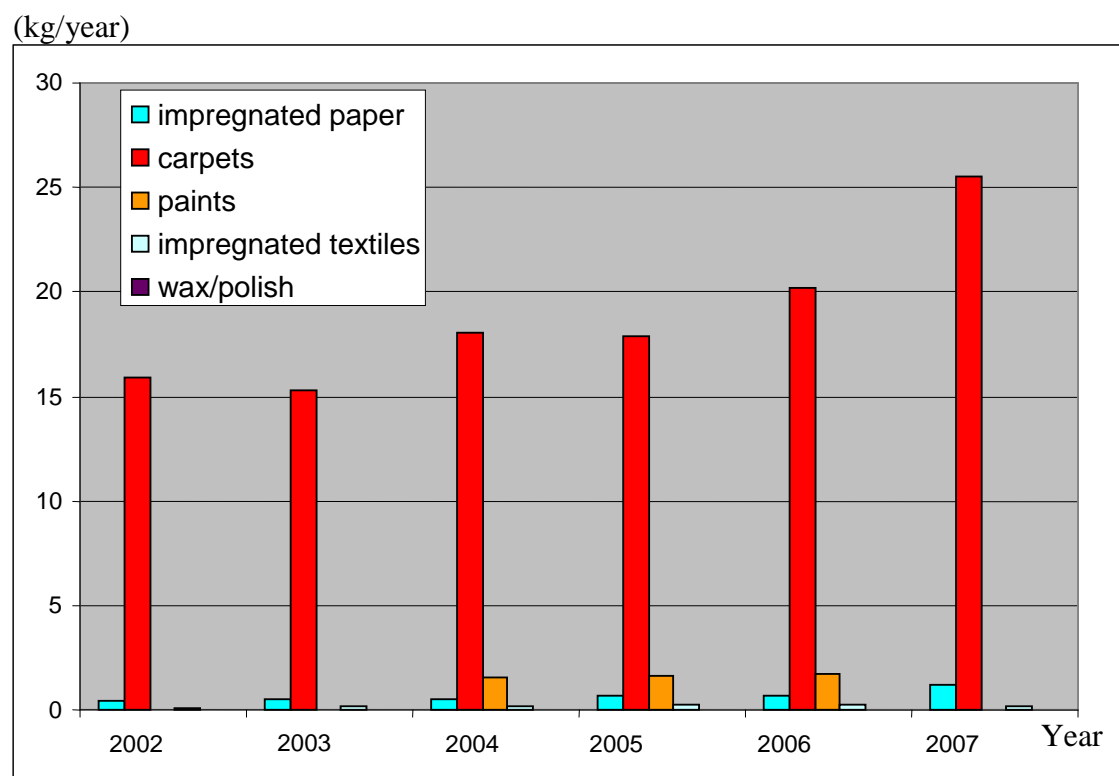


Figure 11: Time trend of maximum PFOA consumption reported by SSB in kg/year (numbers for 2007 are extrapolated by using twice the half-year consumption of 2007).

The consumption of PFOA in paints remained stable on a low level during the years 2003-2006, with no data for 2007.

## 8.6 Conclusions

The estimate from this survey of the annual maximal emission of PFOA from a range of consumer products identified in this survey as major contributors of PFOA could be summarized as shown below:

Carpets	12 kg
Coated and impregnated paper	1,3 kg
Textiles	0,5 kg
Paint and lacquer	1 kg

The long range annual transport estimate from this survey of PFOA into Norway from direct and indirect sources mainly from oceans and airborne particles is as follows

Total of direct and indirect sources	130 to 380 kg
--------------------------------------	---------------

However, in general and despite the high degree of uncertainty of the assessed data material, the amounts of PFOA circulating in the Norwegian society and has its origin from long range sources are rather low with some tens of kilograms as a maximum amount.

## 9. Discussion

### 9.1 Knowledge and data gaps

The globalisation of trade has moved the actual manufacture and production processes far away from the consumer markets in Europe. As a consequence we lack knowledge about chemicals applied deliberately or as impurities/degradation products in consumer products through the distribution chain into the Norwegian market. This is definitely a global problem. However, the Scandinavian countries have had a long tradition of close attention to flows of chemicals that enter the Scandinavian markets, either through legislation or by customer agreements with Scandinavian companies.

This project has clearly shown the complexity of mapping sources to PFOA in Norway, and the cause of PFOA emissions into the environment, especially from consumer products. Many different sources of information have been applied in this project, such as interviews with companies that manufacture and distribute articles that could be sources to PFOA, or search in different relevant databases and statistics of manufacture, import and export and through mathematics calculate the extent of PFOA occurrence and emissions into the Norwegian society and environment. There is, however, little or no knowledge available on how these information sources interrelates. Beside the lack of correspondence between these different important information sources, there are also knowledge gaps along the distribution chain of chemicals, where the article manufacturer or distributor could not give detailed answers during our interviews. There is also a commercial perspective, where the supplier of perfluorinated products does not distribute important information along the distribution chain, since it is considered as business secrets.

If amounts of landfill and STP effluents and sludge emitted in the Norwegian environment were fully known, the data collected in the studies by SFT and Nordic screening could be used in order to calculate annual amounts of PFOA emitted via this route.

## 9.2 Physico-chemical properties of fluorine chemistry

### 9.2.1 Physico-chemical properties of fluoropolymers and their treated surfaces

Surface energy and contact angles are the most critical physico-chemical parameters in the action of fluorochemical products. Therefore it is essential to define these surface properties in order to achieve the appropriate surface protective properties or otherwise the purpose of the surface treatment is lost.

Fluorinated chemicals can be used to provide water repellency, stain resistance and soil release properties to a treated surface is related to the physico-chemical properties of these fluorinated materials. The critical surface tension is the determining physico-chemical parameter why fluorinated chemicals can repel both water and oil substances ( J.F. Colbert et al 1983) (J. M. Corpart et al. 1997).

The critical surface energy  $\gamma_c$  of the  $CF_3$  and  $CF_2$  groups are much lower compared to the surface energy of the corresponding hydrocarbons (  $CH_3$  and  $CH_2$  ), which is described in table 14 below.

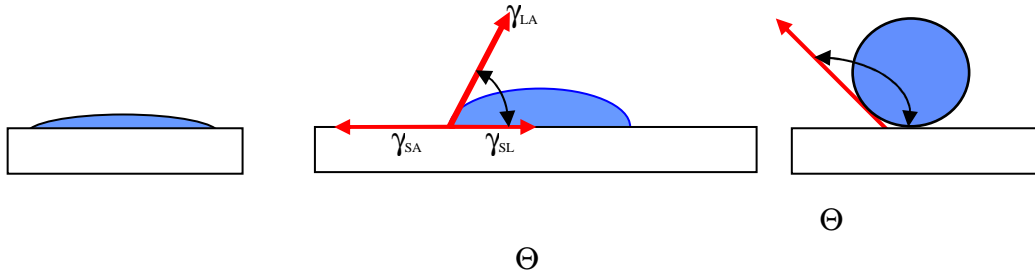
Table 14: Surface energies for characteristic polymer backbone structures

<u>Surface</u>	<u>Liquids</u>	<u>Surface energy : <math>\gamma_c</math> ( mN/m )</u>	<u>Surface tension : <math>\gamma_L</math> ( mN/m )</u>
- $CF_3$		6	
- $CF_2H$		15	
- $CF_2$ -		18	
- $CH_3$		22	
- $CH_2$ -		31	
- $CH_2CHCl$ -		39	
Polyester		42	
Polyamide		46	
Cotton		44	
	water		72
	n-Octane		22
	Olive oil		32

One of the fundamental laws of physics states that every system strives for a minimal surface energy. Therefore when a fluorinated chemical is coated on a textile substrate and exposed to water with its surface tension of 72 mN/m or oily substances with surface tensions of 20 mN/m and more, they will not spread on the textile surface. The consumer can observe this phenomenon as "water and oil repellency".

The spreading of a liquid on a surface is measured via contact angles and demonstrates well when a fabric is being wetted or not.

When : - angle  $\Theta$  is  $> 90^\circ$  : liquid will not wet the surface  
 - angle  $\Theta$  is  $< 90^\circ$  : liquid will wet surface partially  
 - angle  $\Theta = 0^\circ$  : complete spreading & wetting of the surface by the liquid.



$\Theta = 0^\circ$  Perfectly Wettable

$\Theta < 90^\circ$  +/- Wettable

$\Theta > 90^\circ$  Unwettable

Spreading occurs only if  $S > 0$

Spreading coefficient :  $S = \gamma_{SV} - (\gamma_{LV} + \gamma_{SL})$       S = solid, L = liquid, A = air  
 where :  $\gamma_{SA}$  = Surface energy of the substrate  
 $\gamma_{LA}$  = Surface tension of the liquid  
 $\gamma_{SL}$  = Interfacial tension

Figure 12: Contact angle versus wettability of a surface

As can be seen from the formula for spreading  $S = \gamma_c - (\gamma_L + \gamma_{cL})$  it is observed that if the surface energy of the substrate is lowered sufficiently, the liquid will not be able to wet the surface.

Practice shows that it is not sufficient to have only terminal  $CF_3$  groups in a fluorinated chemical. Optimum reduction of the surface energy  $\gamma_c$  is achieved with perfluorinated chains with a sufficient chain length to obtain a large enough density of fluorinated carbons on the surface.

This is demonstrated in literature ( J.F. Colbert et al 1983) (J. M. Corpart et al 1997) of fluorochemicals that there is a relationship of the chain length of the perfluorinated chains that is related to the critical surface energy of the surface as described in figure 13 below.

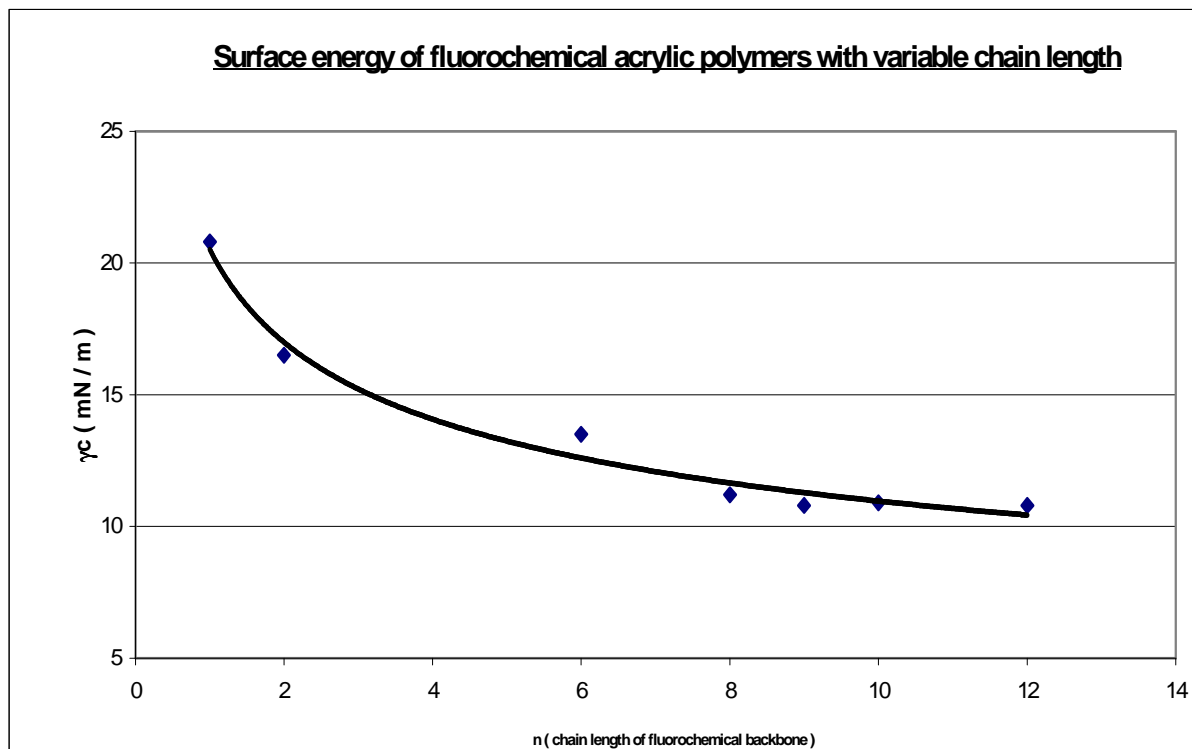


Figure 13 : Surface energy versus number of carbons in the fluorotelomer backbone structure, where 4 on the x-axis means C4-chemistry, 6 means C6- chemistry and 8 means C8- chemistry etc.

Based on the already explained surface energy properties of fluorinated chemicals, it is understood that for instance a non fluorine surface treatment, such as silicones on treated polymers, can provide rather good water repellency but no oil repellency due to the fact that the oil has lower surface energy than the silicone layer. The surface energy obtained with a silicone surface can not be lower than 22 mN/m, which is comparable to the surface tension of hydrocarbon oily substances. This means that fluortelomers are not always possible to replace with a non fluorine surface treatment if oil or soil repellance is required.

### 9.3 Alternatives to C8-fluortelomer chemistry

There are no current commercial alternatives to the C8 fluortelomer-chemistry, that cause PFOA emissions for impregnation of carpets and textiles. To achieve required soil and water repellence properties, leather is sometimes coated with fluoropolymers instead of being impregnated with C8-fluortelomers. Major chemical manufacturers<sup>7</sup> carries out extensive research at the moment to present commercial soil repellent properties for their chemical preparations based on C4 or C6 chemistry with acceptable quality properties as for the C8 chemistry. Forecast says that the first alternative fluorochemical preparations will be on the market earliest 2008.

<sup>7</sup> Oral information from Clariant



There are however surfactant alternatives that can provide similar wetting properties in coating formulations to C8 fluortelomers. They are described in the following chapters below,.

### 9.3.1 Fluorinated polyether

Fluorinated polyethers can be used as surfactants and as flow, level, and wetting additives for coating formulations. These formulations are currently being used as a surfactant in floor polish and are polymers with a molecular weight greater than 1,000. The polymers are based on ether links – both the polymer backbone linkages and the link between the backbone and the perfluoroalkyl pendant. The fluorosurfactants are synthesized from perfluoroalkyl starting materials with a fully fluorinated carbon chain length of C<sub>4</sub> or less (Poulsen et. al 2005).

The basic structure of these compounds is illustrated in the figures below.

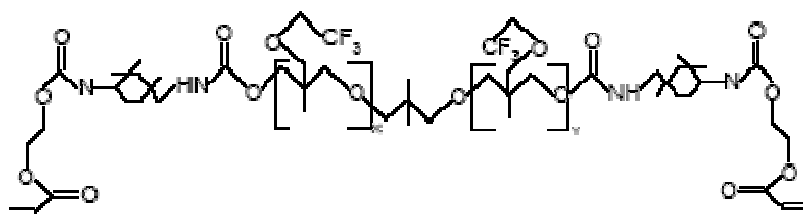


Figure 14: The basic structure of a fluorinated ether compound.  $x+y$  equals about 20.

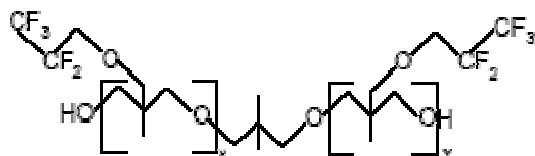


Figure 15: The basic structure of a fluorinated ether compound as the basic perfluoroalkyl group.

### 9.3.2 Fatty alcohol polyglycoether sulphate

BASF produces a fatty alcohol polyglycoether sulphate, which can be used as levelling and wetting agent in paints and coatings (Poulsen et al 2005).

### 9.3.3 Silicone polymers

Silicon polymers can be used as wetting agents as replacement for fluorinated surfactants in several cases in the paint and ink industry. Their products contain 3-(polyoxyethylene) propylheptamethyl trisiloxane with (CAS No 67674-67-3) (Poulsen et. al 2005).

### 9.3.4 Sulfosuccinate

Sulfosuccinate can be used as a wetting agent for paints and coatings. In one product the sulfosuccinate is mixed with water and ethanol, and in the other the sulfosuccinate is mixed with water and 2, 2-dimethylpropane-1, 3-diol (Poulsen et. al 2005).

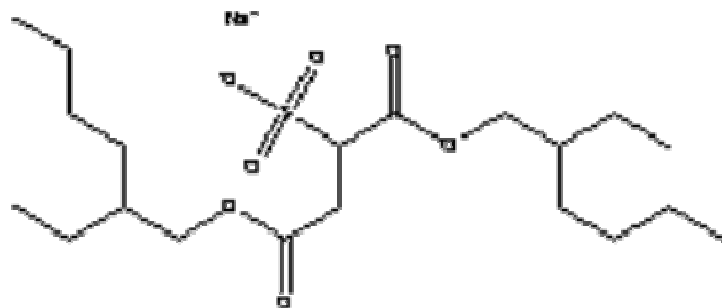


Figure 16: The chemical structure of the sodium salt of di(2-ethylhexyl) sulfosuccinate (CAS No. 577-11-7).

### 9.3.5 Propylated aromatics

Propylated aromatics, naphthalenes and biphenyls, can be used as water repelling agents for different applications, such as corrosion protection systems, marine paints, resins, printing inks, coatings, electrical, electronical and mechanical applications

The presented propylated aromatic products are all colourless liquids with a boiling point at about 300 °C. Their flash point lies all about 140 °C. The substances have a very low solubility in water. Common for the substances is that none of them is classified as hazardous substances (Poulsen et. al 2005).

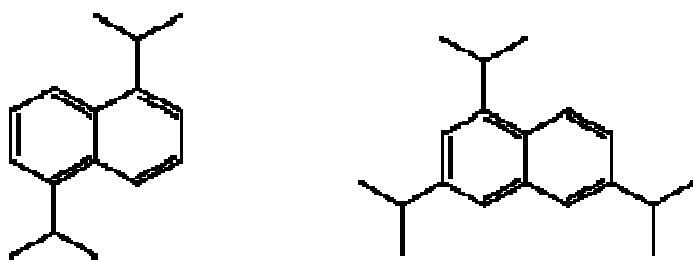


Figure 17: The chemical structure of propylated aromatics (CAS No. 38640-62-9) and (CAS No. 35860-37-8) – to the right.



Figure 18: The chemical structure of propylated aromatics, to the right (CAS No. 25640-78-2) – to the left, (CAS No. 69009-90-1) to the left. Both products are mixtures of isomers.

### 9.3.6 Conclusions

In general very little information about the specific substances above is available. It is therefore advisable to apply the substitution principle for these alternative substances as replacements to C8-fluortelomers, when we know more about their potential health and environmental hazard properties.

The alternative hydrocarbon surfactants seem to be ready biodegradable. The fatty alcohol polyglycoether sulphate is readily biodegradable and does not seem to be toxic to aquatic organisms. The sulfosuccinates are likewise easily biodegradable, do not seem to bio concentrate, but are harmful to aquatic organisms. The biphenyls and the naphthalene derivatives are potentially bio accumulative. The biphenyl moiety seems to be easily biodegradable, whereas the naphthalene moiety only slowly biodegrade. The sparse information suggests that the biphenyls are acutely toxic to aquatic organisms, whereas the naphthalene's have no acute toxic effects in the investigated fish species.

Of the alternatives, the silicone polymers seem to have the more adverse environmental effects. The specific compound investigated is classified as environmentally harmful (R51/53 "Toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment"), as the substance is toxic to aquatic organisms and is bio accumulative (Poulsen et.al 2005).

## 9.4 Future work

In order to improve the information flow in the Norwegian distribution chain, a number of actions are recommended to get the best use of existing chemicals information for the improvement of correlation between the national statistics with chemical information. This would mean that a supplier or manufacturer of a certain category of articles will describe the chemical content of an article and report it in an appropriate way to statistics. Business secrets need to be dealt with in such a way that existing chemical information could become available from the chemicals supplier along the distribution chain.

In this context it could be recommended that the Product Register should be notified all products containing PFCs and not only dangerous products. That would be the best way to get information about the national use in chemical products and if possible for consumer articles.

This correlated source of information from statistics and market information on articles on the Norwegian market in context to better knowledge about amounts of landfills and STP effluents, the mass balances of PFOA could easier be followed within Norway and consequentially could make it possible to calculate annual amounts of PFOA emitted to the Norwegian environment.

This survey describes a range of analytical results concerning consumer products and historical data of indirect and direct emissions of PFOA from manufacturing use and processing of perfluorinated products in Norway. Due to the lack of sufficient data, this study cannot fully assess the relative importance of consumer articles in comparison to water, food and air to PFOA or PFCA exposure in Norway.

The authors of this report recommend that a priority be put on future studies, building on work which has been recently published, to make such an assessment in strong cooperation on an international level, since the PFC issue is an international problem and not isolated to Norway.

This data achieved in this study can be supportive in the future work for the selection of relevant perfluorinated products and their magnitude as sources of PFOA in Norway.

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## 11. Appendix A – terminology and definitions<sup>8</sup>

### 11.1 Fluorochemical

A general, non-specific, term used for a broad description of all chemicals containing the element fluorine. Specifically, the term is used most commonly to describe small (1-8 carbon length) fluorinated molecules, which are most often used for refrigeration, as fire suppression agents and as specialty solvents.

### 11.2 Fluorinated chemical

A general, non-specific term used synonymously with "fluorochemical".

### 11.3 Fluorotelomer

A specific term used to describe an oligomer created by reaction of tetrafluoroethylene (TFE) with perfluoroethyl iodide  $\text{CF}_3\text{CF}_2\text{I}$  to produce  $\text{F}(\text{CF}_2\text{CF}_2)_n\text{-I}$  [ $n = 3-6$ , avg. 4], a linear, even carbon number chain length oligomer; the term "*telomer*" is often used synonymously with *fluorotelomer*.

### 11.4 Fluorotelomer alcohol (FTOH)

A general term which describes a class of alcohols of the general structure  $\text{F}(\text{CF}_2\text{CF}_2)_n\text{CH}_2\text{CH}_2\text{OH}$ , where  $n$  is an integer.

### 11.5 Fluoropolymer

A general term used to describe a polymer which has fluorine attached to the majority of carbon atoms which comprise the polymer chain backbone. Common fluoropolymers are: polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), fluorinated ethylene-propylene (FEP), etc. These are typically high molecular weight polymers used in high performance applications where chemical resistance and thermal stability are essential.

### 11.6 Fluorinated (organic) polymer

A general term used to describe a polymer which has a hydrocarbon backbone (polyamide, polyester, polyurethane, etc.) to which a fluorinated carbon chain is appended, also known as a fluorinated alkyl chain; an example would be a polymer such as -  $[\text{CH}_2\text{CH}(\text{C}(=\text{O})\text{OCH}_2\text{CH}_2(\text{CF}_2)_8\text{F})]_n\text{-}$ .

### 11.7 Perfluoro- / perfluorinated

Describes specifically a substance where *all* hydrogen atoms attached to carbon atoms are replaced with fluorine atoms –  $\text{CF}_n$ .

### 11.8 Perfluoroalkylated substance (PFAS)

<sup>8</sup> Poulsen, P. B, Jensen, A. A., and Wallström, E. More environmentally friendly alternatives to PFOS-compounds and PFOA. 20-5-2005. Danish Ministry of the Environment. Environmental project No. 10132005., Appendix A

A general term, which describes a substance that bears a perfluorocarbon unit  $F(CF_2)^n-R$  where n is an integer and R is not a halogen, or hydrogen. Examples include  $F(CF_2)_6CH_2CH_2OH$ ,  $F(CF_2)_6SO_2N(CH_3)CH_2CH_2OH$ , and p- $F(CF_2)_6-C_6H_4OH$ .

*Note: OECD uses the abbreviation (PFAS) for perfluorinated alkylated sulphonates (OECD 2006)*

### 11.9 Fluoro surfactant

A non-specific, general term used to describe a surface active, low molecular weight (<1000), substance where carbons bear fluorine in place of hydrogen. Examples would include  $CF_3(CF_2)_7SO_3^-K^+$ ,  $H(CF_2)_7COO^-NH_4^+$ ,  $F(CF_2CF_2)_3CH_2CH_2SO_3^-NH_4^+$ ,  $CH_3CH_2CF_2CF_2CH_2COO^-NH_4^+$ , etc.

### 11.10 Perfluorocarboxylic acid (PFCA)

A general term which describes a class of fully fluorinated carboxylic acids and their salts of the general structure  $F(CF_2)^n-COOX$  ( $X = H, NH_4^+, \text{Group I alkali metal, Group II alkaline earth}$ ). These are generally manufactured by the electrochemical fluorination process or from fluorotelomer-based raw materials. The most common industrially used fluoropolymer polymerization aids are: ammonium perfluorooctanoate (CAS# 3825-26-1) and perfluoro fatty acids, C<sub>7-13</sub>, ammonium salts (CAS# 72968-3-88).

### 11.11 Polyfluorinated substances

A term for substances with more fluoro-substituents.



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<p>Forfattere Stefan Posner, IFP Research (SE), Dorte Herzke, NILU (NO), Pia Brunn Poulsen, Force Technology (DK), Allan Astrup Jensen, Force Technology (DK)</p> <p>Tittel - norsk og engelsk PFOA i Norge - Kartlegging av nasjonale kilder, 2007 PFOA in Norway - Survey of national sources, 2007</p> <p>Sammendrag – summary This report outlines potential sources of PFOA in Norway. PFOA is present in low concentrations in consumer products such as coated and impregnated textiles, carpets, paper, paints and laquers. Long range water and air transport from direct sources outside Norway is probably the most important source of PFOA. Rapporten beskriver potensielle kilder for PFOA i Norge. PFOA finnes i lave konsentrasjoner i forbrukerprodukter som belagte og impregnerte tekstiler, tepper, papir, maling og lakk. Sansynligvis er langtransporterte foruresninger fra luft- og havstrømmer den viktigste kilden til PFOA i Norge.</p>
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4 emneord Perfluoroktylsyre (PFOA), perfluorete forbindelser, fluorpolymer, fluortelomer	4 subject words Perfluorooctanoic acid (PFOA), perfluorinated substances, fluorpolymer, fluortelomer.
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SFT skal bidra til å skape en bærekraftig utvikling. Vi arbeider for at forurensning, skadelige produkter og avfall ikke skal føre til helseskade, gå ut over trivselen eller skade naturens evne til produksjon og selvfornyelse.

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