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Polyfluoroalkyl substances (PFASs) in textiles for children

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Polyfluoroalkyl substances (PFASs) in textiles for children

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Contents

Foreword

This study is part of the Danish Environmental Protection Agency's programme for surveys of chemicals in consumer products, which in 2014 had a special focus on chemical substances in products for children.

The purpose of this study is:

- To survey which consumer products of textiles for children contain PFASs.
- To analyse the PFASs to be found in the materials and investigate the extent to which PFASs may be released during wear and washing of textiles.
- To assess whether the release of the substances poses health and environmental risk.
- To assess relevant waste streams, waste amounts and fates of the substances in waste treatment.

The project was conducted from March to December 2014, in a collaboration between COWI (project management, mapping, environmental assessment and quality control), NIPSECT (health assessment), Danish Technological Institute (analysis of total fluorine, washing and migration tests) and Aarhus University (analysis of PFASs in textiles, laundry water and migration fluid).

The project was overseen by a steering committee with the following members:

- Maiken Guldborg Rasmussen, Danish Environmental Protection Agency
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- Rossane Bossi, Aarhus University
- Allan Astrup Jensen, NIPSECT
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Minor corrections have by October 2022 been made to the calculations in Tables 14 and 15.

Summery and conclusion

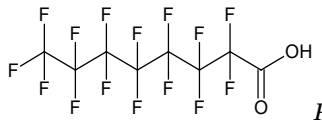
Background and purpose
Polyfluoroalkyl substances (PFASs) is a large family of surfactants with different uses and environmental and health properties. There are major difference in how thoroughly the substances are tested for hazardous effects on human health and the environment. Among the substances studied most thoroughly, carcinogenic, toxic for reproduction and acute toxic effects have been observed.

PFAS-based coatings are being used in garments and other textiles in order to make the materials water and dirt repellent. Previous studies have shown that fabrics treated with these coatings contain small amounts of a variety of PFASs. The presence of PFASs has given rise to concerns about whether users of these textiles are being exposed to the substances to a degree that may pose a health risk. Globally, coatings for textiles represent 50 % of the total use of PFASs. There is uncertainty about the extent to which the use of the substances in textiles contributes to the release of the chemicals to the environment.

The purpose of this study is:
- To survey which consumer products of textiles for children contain PFASs.
- To analyse the PFASs to be found in the materials and investigate the extent to which PFASs may be released during wear and washing of textiles.
- To assess whether the release of the substances poses health and environmental risk.
- To assess relevant waste streams, waste amounts and fates of the substances in waste treatment.

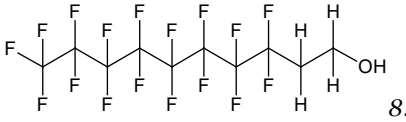
The substances investigated
PFASs - Polyfluoroalkyl substances (PFASs) represent a very large family of substances, which differ in the functional groups and the length of the carbon chains.

Perfluoroalkyl substances form a subgroup of PFASs of particular interest because of their persistence and toxicity. In the perfluoroalkyl substances all carbon atoms are fully fluorinated, as shown in the structural formula of perfluorooctanoic acid (PFOA) below.



Perfluorooctanoic acid (PFOA)

Another large subgroup of PFASs is the fluorotelomers. The most typically used fluorotelomers contain a perfluorinated carbon chain like the perfluoroalkyl substances, but the fluorotelomers also contain two more carbon atoms which are not fluorinated. An example of a fluorotelomer having a perfluorinated moiety of eight carbon atoms is 8:2 fluorotelomer alcohol (8:2 FTOH), as shown below. As the perfluorinated part is highly stable, 8:2 FTOH could be degraded to PFOA and perfluorononanoic (PFNA) in the environment.



8:2 fluorotelomer alcohol (8:2 FTOH)

Abbreviations for the substances and substance groups, given after the English substance names, are used throughout this report. The full names, CAS numbers, group names and chain lengths are shown in Annex 1.

Mapping

Marketed impregnating agents with PFASs - A number of PFAS-based coating agents for textiles are marketed under trade names such as SCOTCHGUARD®, Teflon® fabric protector, Nano-Tex® and Unidyne®. The products typically consist of a mixture of reactive PFASs, other reagents and solvents. The impregnating agents are applied as a thin film to the surface of the fabric, followed by polymerization and curing, whereby a thin polymer structure is formed on the surface, to which both polyfluoroalkylated and non-fluorinated side chains are attached.

The side chains, which extend as small hairs from the material, aid in providing the dirt and water repellent effect. The fluorinated side chains are similar in structure to PFASs, and have a perfluorinated moiety of varying length. The non-polymeric PFASs, which can be extracted from the cured fabric, consist partly of unreacted intermediates and partly of PFASs released by hydrolysis of the ester bond between the side chains and the polymer.

Besides the occurrence in PFAS-based water impregnation agents, PFASs also occur in the membranes of the plastic polytetrafluoroethylene (PTFE), which is used to make clothes waterproof. The best known of these membranes is Gore-tex®, but there are also others on the market. PTFE may contain residues of PFOA and PFNA from manufacturing whereby the sodium salts of these substances traditionally have been used as a processing aid. Today, alternative processing aids are used to a great extent, but it has not been possible to find information on which materials are being used. As Gore-tex® and similar membranes are rarely used in children's clothing (however, they are used in footwear for children), focus on textiles with PFAS-containing coatings in the outer fabric is prioritized in this study.

Prevalence of PFAS-based impregnating agents in articles for children – Fifty-five distributors of children's clothing and items for children have been contacted in this study. The majority replied that they do not sell relevant products without filling out a questionnaire. Out of the 17 companies that have returned a completed questionnaire, six companies indicated that they sell products treated with PFAS-based coatings. Two of these companies informed the authors that their products were treated with Teflon®, while the others informed that they did not know which PFAS-based impregnating agents the products are treated with. An Internet search revealed that it is only the Teflon® Fabric Protector trademark, which is used in the advertising, and that the other brand names were not referred to in advertising for the products.

As a result of the questionnaire and an Internet search on products with water and dirt repellent properties for children, the following product types where it is specifically stated that PFAS-based coatings (typically by specifying Teflon® fabric protector) have been used, were found:

- Widespread occurrence: Snowsuit [Danish: flyverdragter], rainwear, skiwear, gloves and mittens, infant sleeping bags [Danish: køreposer] and rain cover for prams.
- Occasional examples: backpacks/school bags, soft-shell jackets, hats, sun canopies for prams, strollers, baby carriers, baby changing bags (not nursing pillows).

Several Internet shops sell one or more products indicated to contain the trademark Teflon® fabric protector, but it has not been possible to find products where it is indicated that they are treated with other trademarks. A study of 3,770 products for children, sold by eight Internet shops that all indicate that one or more of their products is treated with Teflon® fabric protector, revealed that between 3 and 5 % of snowsuits, gloves/mittens, ski pants, rain cover for prams and infant sleeping

bags were reported to be treated with Teflon® fabric protector. The calculated percentages may underestimate the true prevalence of PFAS-impregnated products, since the brand name is probably not indicated for all products with PFAS-based impregnating agents.

A trend in decreasing use of PFAS-based impregnating agents in clothing for children is currently observed. In the responses to the questionnaire, four companies indicated that they have previously used fluorine-based technology to achieve water and/or dirt-repellency, but they have now switched to a PFAS-free, alternative polymer technology (dendrimer technology). Products treated with this technology are water-resistant, but not oil and dirt repellent to the same extent as products treated with PFAS-based agents.

On the basis of this survey, it is estimated that the total share of snowsuits, gloves/mittens, ski wear, and rain cover for prams treated with PFAS-based impregnating agents, probably will be around 10-30 %, while that of rainwear, jackets and infant sleeping bags will probably be somewhat lower. For the other product types mentioned, the estimated percentage of products treated with PFAS-based impregnating agents is less than 10 %.

Given the size of a snowsuit and the large number being sold, these are presumed to constitute the largest tonnage of textiles with PFAS-based impregnating agents. It is estimated that annually 200,000-300,000 snowsuits are sold, of which 20,000 to 90,000 suits will contain PFASs.

Waste Disposal - The impregnated garments for children will (possibly after being reused several times) ultimately end up in waste incineration plants or exported for reuse abroad. The exposure conditions, when the garments are reused, are deemed not to be significantly different from the conditions by the first use of the garment. Other surface-treated products are assessed primarily to end up in waste incineration, as export for reuse of the products abroad is estimated to be modest. Treated clothing is deemed to constitute the largest source of PFASs to incinerators, as clothing worldwide (and probably also in Denmark) constitutes about 50 % of the consumption of fluorotelomer and fluorotelomer-based polymers.

The main issue in relation to the disposal of textiles with PFAS-based impregnation agents in Denmark is whether the substances are degraded by the waste incineration. The available results indicate that PFASs is effectively destroyed in modern incinerators. PFASs which are not destroyed by the incineration are likely to be captured by flue gas treatment and end up in the flue gas cleaning products, similar to, for example, dioxins and furans. It is therefore assumed that the incineration of textiles with PFAS-based impregnation agents is not a source of significant releases of PFASs to the environment – and this assumption is shared in foreign studies.

PFAS-containing clothes exported for recycling abroad are mainly disposed of to landfill sites, which are uncontrolled in many cases. The further fate of the PFAS-based impregnation agents has not been investigated, but it is reasonable to assume that in the longer term, decomposition of textiles may result in release of substantial amounts of PFASs to the environment. Regardless of the time perspective, the fact that textiles account for almost half of the global consumption of PFASs indicates that textiles could potentially cause major impacts on the environment in countries without waste incineration in the long term.

Selection of products and substances

Selection of products - On the basis of the survey, 22 products, which included the product groups snowsuits, jackets, gloves, rain suit and infant sleeping bags, were purchased.

Prior to purchasing the products, several exposure scenarios were prepared with the aim of ensuring that the study includes the applications representing the greatest risk of exposure to children.

Furthermore, samples of the fabric part of eight car seats were taken. The products were purchased as part of another study for the Environmental Protection Agency.

Selection of substances - A total of 39 PFASs was selected within the following groups of PFASs:

- Perfluoroalkyl substances: Perfluoroalkane sulfonic acids (PFSAs), perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkane sulfonamides and perfluoroalkane sulfonamidoethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs).
- Fluorotelomers: Fluorotelomer carboxylic acids (FTCAs), fluorotelomer acrylates (FTACs) fluorotelomer methacrylates (FTMACs), fluorotelomer alcohols (FTOHs) and fluorotelomer sulfonic acids (FTSAs).

These substances were selected on the basis of the results of previous studies of PFASs and the knowledge of which substances most likely would occur in the textiles.

Chemical analyses

Content of total fluorine – The analysis program was initiated with the analysis of total fluorine in 22 products with a proven content of fluorine for the (more expensive) analyses of PFASs. Total fluorine in concentrations above the detection limit in the outer material was found in 19 of the 22 (~ 86 %) examined samples of children's clothes and infant sleeping bags. The concentration varied in those products where the concentration was above the detection limit, from 8,000 to 365,000 µg/m², with an average of approximately 72,700 µg/m². It is assumed that PFASs constitute the vast majority of the fluorine content of the products. In the group of products which only indicates that the products are stain and water resistant, the concentrations of total fluorine were above the detection limit in 8 of 10 products (80 %). Therefore, for outerwear a claim of being "dirt and water repellent" may be a good indication that the clothes have been treated with PFAS-based impregnating agents.

In all of the eight car seats that were tested for total fluorine, the concentration of total fluorine in the textiles was below the detection limit of 20 mg/kg. On this basis, it is concluded that none of the car seats contained textiles treated with PFAS-based impregnating agents.

Contents of PFASs - Based on the results of the analysis of total fluorine, 15 products were selected for analysis of PFASs content. The products were selected from among those products with the highest concentrations of total fluorine. In the analysis, the quantity of non-bound PFASs that could be extracted from the textiles was measured. It is not possible to conduct an analysis of the polyfluorinated side chains making up the bulk of the fluorine content in the textiles.

Concentrations of PFASs above the detection limit were found in all 15 products. The total concentration of the examined PFASs varied from 18 to 407 µg/m². Only a very weak correlation between the content of total fluorine and the total content of the analysed PFASs was found. This finding illustrates that there are many factors that affect the amount of unreacted intermediates and PFASs formed from hydrolysis of the bond between the side chains and polymer backbone. The content of total fluorine was, on average, 1,600 times greater than the total content of non-polymer PFASs (by weight).

The results of the analyses showed that fluorotelomer alcohols (FTOHs) could be found in the highest concentrations in all products, followed by perfluoroalkyl carboxylic acids (PFCAs). Combined, these two groups represented 94 % of the total content PFASs in each of the samples.

In five of the products, the concentration of PFOA was above 1 µg/m², which is set as a limit value for PFOA content in textiles in Norway (with a transitional period for stored goods).

Besides PFOA, the following perfluoroalkyl carboxylic acids were found in concentrations above 1 µg/m² in some of the products: PFHxA (C_6), PFHpA (C_7), PFNA (C_9), PFDA (C_{10}), PFDoDA (C_{12}), PFTrDA (C_{13}) and PFTeDA (C_{14}).

Relatively low concentrations of perfluoroalkane sulfonic acids (PFSAs) were found and the concentration of PFOS was below 0.21 µg/m² in all samples. Consequently, PFOS was below the EU limit value of 1 µg/m² in all samples.

In recent years, much debate on whether a change from fluorine chemistry, based on substances with chains of 8 or more carbon atoms (C_8 chemistry) to substances with a shorter chain length (C_{4-6} chemistry), has taken place. For all of the substance groups and all of the samples, the results from this study show that C_8 chemistry was dominant. PFOA was therefore the dominant perfluoroalkyl carboxylic acid (PFCAs), 8:2 FTOH the dominant fluorotelomer alcohol (FTOHs), 8:2 FTCA the dominant fluorotelomer acid (FTCAs), 8:2 FTAC the dominant fluorotelomer acrylate (FTACs) and 8:2 FTSA the dominant fluorotelomer sulfonic acid (FTSAs).

Migration of PFASs to saliva – Analyses of migration of PFASs from products to artificial saliva from the eight products showed that a small fraction of the total content of the materials migrated to the artificial saliva over a period of 3 hours.

For the majority of the investigated compounds, the concentrations in the artificial saliva were below the detection limit.

Thus, on average only about 1 % of the total content in the materials migrated to the artificial saliva. Migration rates were, however, significantly higher for the acids, which dominated the composition of PFASs. On average, 6 % of the material's content of PFCAs migrated to the simulant during the test period. There was a trend toward higher migration rates (in relation to the materials' content) for the short-chain PFCAs than the long-chain.

Release of PFASs by washing of the clothes - Preliminary methodological clarifications showed that it might be difficult to analyse some of the substances in the laundry water, as the surfactants in the detergent interfered with the detection of PFASs.

However, it was possible to obtain satisfactory results for the most important substances (the perfluoro carboxylic acids), and it was therefore decided to perform the washing tests still using the detergent because it was expected that the detergents would increase the leaching of substances from textiles. As was the case with migration, the proportion of the materials' content released to the laundry water, was significantly higher for the acids than for the other PFASs. The measured emission rates also corresponded very well with the observed migration rates. Of the total PFASs, only 1 % represented releases to the laundry water in a single wash. Of PFCAs, the release rates were significantly higher. The amount that was released to the laundry water represented an average of 12.2 % (standard error: 5.0 %) of the content of the materials.

In foreign studies, release rates for emissions to the laundry water 5-20 times higher have been found, but the analysed types of clothing is not totally comparable.

Emission of PFASs to air

Within the framework of the project it was not prioritised to investigate the release of the substances into the air, as this is investigated in a number of foreign studies. In a German study, it was found that 27 to 70 % of the materials' content of extractable 10:2 FTOH was emitted to air over a period of 5 days. Emission rates of 8:2 FTOH were somewhat lower, with emission rates of 7 to 16 % of the material's content. One difficulty in interpreting the results of this study and others is that no measurements have been conducted for an extended period of time. Since the FTOHs are volatile

substances, it must be expected that they would evaporate over time, and results must be affected depending on what period of time after production the samples are taken. When a significant proportion of FTOHs in products that may be many months old is emitted over 5 days, it must be expected that significantly higher amounts may have been emitted earlier in the life cycle.

Health assessment in relation to children

Exposure to PFASs – When exposed to PFASs, an almost complete absorption after oral ingestion or inhalation takes place, while on the other side, it is very difficult for the substances to penetrate intact skin. The absorbed PFASs bind to albumin in the blood and is distributed to most organs, but in most cases the target organ is the liver.

The residence time in the blood and distribution to other tissues and organs depends on the compound, the amount of exposure and the animal species. The residence time in the blood increases with the chain length; sulfonic acids have a longer residence time than carboxylic acids. The residence time is relatively short in rodents, slightly longer in monkeys and very long in humans.

The residence time in the blood is partly correlated with the ease of PFASs excretion. The acids are excreted in the urine via the kidneys. Excretion is faster and more efficient in female rats than in males which have more efficient re-absorption in the kidneys. The difference is hormone specific. Humans have the most efficient re-absorption (99 %) in the kidney and it is therefore difficult to separate the components, meaning that a risk assessment cannot be based on results from animal studies, as is usually done.

Health effects - both in animal studies with the individual substances and in population studies in which participants are exposed to a mixture of PFASs, the PFASs affect the fat metabolism with increased risk of incipient liver damage, increased cholesterol and low-density lipoproteins in the blood, and obesity as well as effects on insulin balance. A link between high concentrations of PFOS and PFOA in blood and thyroid disorders and endocrine disruption has also been shown. Effects on the endocrine systems are also seen in animal studies.

Other population studies suggest that PFASs can affect the immune system in children and induce hyperactivity (ADHD). There are further studies that suggest a link between PFOS and PFOA in the blood and chronic renal disease. Given the kidneys' central position in the body's detoxification of PFASs, this effect seems plausible.

Occupational studies, population studies and animal studies suggest that certain PFASs can induce hepatic, pancreatic, bladder, kidney, testicular, prostate and breast cancer – all mostly hormone-related cancers. The International Agency for Research on Cancer (IARC) recently classified PFOA as carcinogenic to humans (Group 2B).

Exposure of children

Regarding children's textiles, the present study shows that the most important PFASs substances that children may be exposed to from these products in Denmark at the present are PFOA and 8:2 FTOHs, which is a precursor to PFOA.

The textile products for children most often containing PFASs are snowsuits, ski wear and everyday wear, in the form of jackets, pants and waterproof clothing with matching mittens, hoods and hats. When children are wearing the clothes, dermal contact and oral exposure, when children suck on the fabrics, are the direct routes of exposure. In addition, the children are exposed to volatile PFASs through the indoor air.

Dermal exposure – Since impregnation with PFASs is found on the outside of children's clothes, it is estimated that dermal exposure to PFASs would be limited to the hands touching the outer body of the clothing, and possibly mittens used to dry the face.

This assessment is supported by worst-case calculations for three scenarios of dermal exposure/absorption for children of 1, 4 and 12 years, respectively. The calculations show a maximum average daily uptake of 0.03 to 0.09 ng/kg bw/day, which is about 100 times lower than the background intake with the diet.

Oral exposure - PFASs can be absorbed orally with saliva if the child suck, chew or lick on the clothes. This was verified by the extraction of PFASs from the textile materials with artificial saliva. The extracted PFASs were particularly water-soluble acids, i.e. PFOA. Despite the almost complete (> 90 %) uptake of PFOA in the gastrointestinal tract, worst-case calculations showed that the intake of PFASs through saliva is minimal, respectively from 0.03 to 0.08 ng/kg bw/day. At the same time, only a small part of the garment (sleeves, gloves) would regularly be exposed to saliva. On the other hand, the use of artificial saliva, which lacks some natural components, probably results in a smaller extraction of PFASs from the material.

Exposure via indoor air - When the clothes are used, stored indoors and dried, evaporation of volatile PFASs (e.g. fluorotelomers and sulfonamides) to indoor air may occur depending on the temperature. Textile dust is formed, wherein the less volatile PFASs (salts, carboxylates and sulfonates) are concentrated, and the more volatile PFASs may be condensed by a drop in temperature. Children will particularly be exposed to PFASs from outdoor wear in institutions and schools. In the absence of actual measurements of indoor air containing PFASs in institutions and schools where children stay indoors especially during the winter months, it is difficult to estimate children's exposure to PFASs in indoor air with certainty. Based on the measurement results from foreign shops selling everyday garments, two scenarios have been calculated, which give a maximum average daily recording of 29 ng/kg bw/day for a 4-year old and 12 ng/kg bw/day for a 12-year-old. The air exchange, however, was not taken into account and many of the parameters used are uncertain estimates.

These calculated estimations result in significant exposures, which are far greater than the background load of PFASs from food, etc., but the scenarios are certainly "worst case" and theoretical. Therefore, exposure calculations based on data on the evaporation of PFASs from the clothes have been carried out. The result is based on a "realistic worst case" where a maximum average daily uptake of 0.49 ng/kg bw/day for the 4 years old and 0.11 ng/kg bw/day for the 12 years old have been calculated. These calculations yield much lower results, demonstrating a very small contribution to children's exposure to PFASs. The fact that air circulation, which reduces the exposure, would also occur is not taken into account.

Several of the parameters used in exposure calculations for indoor air are estimated and particularly uncertain. Therefore it is recommended to carry out concrete indoor air measurements in rooms where children are staying indoors in winter and their outerwear is stored.

Overall uptake of PFASs released from clothes - If the "realistic worst case" contribution to the total absorption from dermal and oral exposure to PFASs in a snowsuit (0.03 + 0, 03 ng/kg bw/day) is added to the contribution from a 4-year-old's inhalation in institutions (0.49 ng/kg bw/day), the overall uptake can be estimated at 0.55 ng/kg bw/day. When compared with a DNEL (Derived No-effect Level) of 80-170 ng/kg bw/day, a total risk characterization ratio (RCR) of 0.003 to 0.008 can be estimated. The value is far below 1. If the RCR is greater than 1, there is a risk of health effects. In the calculation it is assumed that the child is wearing a snowsuit all the day. If, alternatively, the child part of the day use an infant sleeping bag (small children), use rainwear and in addition wear mittens, the RCR values will still be very low. Danish children's average daily intake

of PFOS and PFOA with the diet is estimated to be 1.8-7.2 ng/kg bw/day and 0.3-8.3 ng/kg bw/day, respectively (the two substances jointly form the largest part of the total intake of PFASs). The total intake in the indoor environment, which in this study only included the contribution from the clothes in institutions, can therefore potentially be of the same size as the intake of PFOA with the diet.

Environmental Risk Assessment

Environmental effects of PFASs - Regarding environmental effects, there are only sufficient data on PFOS and PFOA, while data for other PFASs and short-chain carboxylic and sulfonic acids are scarce or absent. Estimates of environmental toxicity (almost exclusively in the aquatic environment) are therefore made typically by extrapolation/"read across" from data for PFOS and PFOA.

The environmental toxicity of PFOS, PFOA and other PFASs is primarily due to the substances' persistence and potential for bioaccumulation, not least among top predators in marine food webs. There is a particular concern about the possible impact in the Arctic. Furthermore, for some substances, a potential for endocrine disrupting effects have been shown.

Environmental exposure via wastewater - On the basis of the calculated "worst case" discharges of PFASs with wastewater from washing clothes and PNEC (Predicted No Effect Concentration) values for PFOS, which is the most toxic of the described PFASs; a risk characterization ratio (RCR) of 0.001 is calculated. If the RCR is greater than 1, there is a risk for ecotoxicological effects. In the classical sense, it appears that there is no significant environmental risk to aquatic organisms associated with the discharge of PFASs to wastewater from washing of textiles and further on into the aquatic environment. However, there are indications in the literature regarding endocrine disrupting effects of some PFASs which are not sufficiently substantiated to be evaluated here.

As mentioned above, the main concern relates to the effects on top predators, particularly in the Arctic. Discharges of wastewater from laundry could contribute to the total emission of PFASs circulating in the environment. In German studies it is estimated that the laundry water from textile washing should only constitute 0.25 % of the sources of PFOA in sewage. The "worst-case" scenarios used in this study indicate that emissions from the laundry could potentially be a significantly larger source. Very different results in terms of discharges from washing of textiles have been found in this and previous studies, and a significant uncertainty is related to whether PFASs can be released from the PFAS-based polymers in relation to washing. It is possible that the total quantities released in the life cycle of the clothing are significantly greater than about 0.04 % of the total fluorine content in the textiles which are available as unbound, extractable PFASs.

Environmental exposure via air - PFASs emitted to air can travel long distances. It will primarily be the volatile PFASs, such as FTOHs, EtFOSE and MeFOSE that are distributed via the air. Of these, it would be FTOHs in particular which could be released from the clothes. The calculations show that even in the worst case, the release of FTOHs from children's clothing in use is unlikely to be a major source of FTOHs.

An unknown factor, however, is the extent to which the volatile PFASs may be released by biodegradation of the impregnated surfaces during the use of the textiles. New, foreign studies, based on various scenarios, show that emissions of volatile substances FOSA/FOSE and FTOHs from degradation of PFAS-based polymers may be of the same magnitude as the release of these substances from all other sources combined. There are no specific estimates for textiles, but the model calculations clearly indicate that textiles could be a significant source of emissions of volatile PFASs.

The potential for emissions of PFASs by the decomposition of impregnating agents in landfills is potentially much greater than the amount of extractable PFASs, and outdoor clothing may be a major source of the detected emission of PFASs from landfills in other countries. In Denmark, clothes (not

recycled) are incinerated at waste incineration plants where PFASs are destroyed and releases from the disposal phase would therefore be negligible. As mentioned above, clothing exported for recycling abroad is ultimately disposed of in uncontrolled dumps and landfills and can thereby contribute to environmental exposure via air and water.

Environmental exposure in soil - PFASs in sewage sludge applied to farmland result in concentrations in the soil estimated to be far below the level at which ecotoxicological effects would occur in the soil.

Data gaps

In relation to an assessment of the potential contribution to the environmental impact of PFASs, which could originate from the use of PFAS-based polymers in textiles, basic knowledge about the extent to which volatile PFASs are emitted immediately after the production of textiles and the extent to which PFASs can be formed by hydrolysis of the perfluorinated polymer side chains during use, washing and degradation of the textiles is lacking. Data on these matters could form the basis for a better assessment of the extent to which the use of PFASs in textiles contributes to the environmental load of these substances.

In addition, specific measurements of PFASs in indoor air in schools and institutions are lacking.

Overall conclusion

The study shows that children's direct exposure to PFASs from their winter clothes gives negligible exposures to PFASs and is not believed to cause health problems. Worst-case estimations of exposure to PFASs through the indoor air in kindergartens and schools do not indicate a health risk. However, there are a number of uncertainties related to the assumptions behind the estimations and it is also uncertain how many other sources could contribute. It is therefore recommended to carry out specific measurements of PFASs in the indoor climate in institutions during winter (incl. measurements of dust) to clarify how much indoor climate contributes to children's total exposure to the substances.

On the basis of the available knowledge, discharges from wastewater treatment plants are assessed not to cause effects in the aquatic environment, but there are uncertainties related to the substances' potential endocrine disrupting effects. The main issue in relation to the environment is the degree to which PFASs in clothes contribute to the total load on the global environment. There are great uncertainties about the extent of emissions from the entire life cycle of the textiles, including the long-term emissions from degradation of the PFAS-containing polymers. The available data indicate that the degradation can potentially contribute very significantly to the total global emissions of volatile PFASs.

1. Background

1.1 Objective of the study

Perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and related compounds were listed on the Danish Environmental Protection Agency's (DEPA's) list of undesirable substances (LOUS) in 2004. The reason for the listing was that the substances are persistent and toxic. The substances are included in a larger group of polyfluoroalkyl substances (PFASs), as introduced in section 1.2 of this report.

In order to further assess the need for additional regulation and improved management of PFASs, in 2012/2013, a mapping of these substances was undertaken (Lassen *et al.*, 2013). The survey was conducted as part of DEPA's series of surveys of all 40 substances/substance groups on LOUS. Based on the LOUS survey, DEPA developed three strategy papers for further action in relation to PFASs. One of the priority actions is to obtain more knowledge about the use of some of the types of PFASs, which are increasingly used as alternatives to the "old" PFASs (especially PFOA and PFOS), and obtain more knowledge about the effects of these substances on humans and the environment.

It is highly variable how thoroughly the substances are studied for harmful effects and the consequent risk to health and environment. Among the substances investigated most thoroughly, is observed carcinogenic, reprotoxic and acute toxic effects. Some of the substances are also suspected of being endocrine disruptors. In the environment, these substances are persistent and bioaccumulate in varying degrees.

PFAS-based surface treatment agents are used in clothing and other textiles to make the materials water and dirt resistant. Worldwide, the consumption for manufacture of textiles accounts for nearly half of the total consumption of PFASs (as detailed in the LOUS survey).

The agents used are discussed in Section 1.3. Today, mainly polyfluoroalkylated polymers with polyfluoroalkylated side chains are used. The polymers typically contain a perfluorinated portion that can be degraded to PFASs. The surface-treated materials may contain residues of starting materials, intermediates and degradation products, as detailed in section 1.3.

Previous studies have shown that PFASs can be extracted from clothing treated with PFAS-based coatings, and volatile PFASs can evaporate from the clothing (discussed further in Section 1.5). Studies have also shown that people in rooms containing a significant amount of PFAS-treated clothing are exposed to volatile PFASs such that this exposure may constitute a significant portion of their overall exposure to the substances (discussed further in Chapter 6).

This observation has given rise to concerns about the extent to which users of clothing and other textiles are exposed to the substances at a level that may pose a health risk. Since children's clothing are also treated with PFAS-based coatings, there is particular concern in relation to whether this pathway can contribute significantly to children's exposure to these substances and cause a health risk.

This study was undertaken as part of the Environmental Protection Agency's programme for the identification of chemicals in consumer products, which had a special focus on chemical substances in products for children in 2014.

The aims of the study are:

- To survey which consumer products of textiles for children contain PFASs (the results are discussed in Chapter 2).
- To analyse which PFASs are present in the materials and investigate to what extent the PFASs are released during use and washing of textiles (results described in Chapter 5).
- To assess whether the release of the substances constitutes an environmental risk and a health risks for children (described in sections 6 and 7).
- To describe relevant waste streams, waste volumes and the fate of the PFASs in waste treatment (described in Section 2.3).

1.2 Introduction to PFASs

Fluoroalkylated substances (PFASs) form a very large family of substances (mainly surfactants) with different uses and different characteristics in relation to the environment and health. The following description provides a very brief introduction to the substances; for a more thorough introduction, please consult the associated LOUS report (Lassen *et al.*, 2013).

PFAS-based agents are used, among other functions, for the surface treatment of clothing and other textiles to make the materials water and dirt resistant. The impregnation agents typically consist of a mixture of reactive PFASs, other reactive substances and a solvent. The impregnating agent is applied as a thin film on the surface of the fabrics, where polymerization and curing takes place. That may impact the material and the water and dirt resistant properties. By this process, polymers with polyfluoroalkylated side chains are formed, as further discussed in the next section (section 1.3). These polymers are referred to as "side-chain-fluorinated polymers" by some authors, but in this report the term “PFAS-based polymers” is used. These polymers should not be confused with the fluoropolymers that are discussed in section 1.4. Previously, non-polymeric PFASs were also used for the impregnation, but all available data indicate that this is no longer the case. Clothes and other textiles can, however, contain a variety of non-polymeric PFASs. They either consist of trace residues of raw materials, intermediates or metabolites from the production of impregnating agents and impregnated clothes, or they are formed by the decomposition of impregnation agents in the textiles.

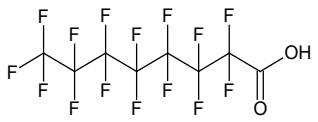
PFASs in textiles can, in addition to originating from PFAS-based impregnation agents, come from fluoropolymers where PFASs are used as processing aids in production, and may be present as residues in the finished textiles. The best known of these membranes is Gore-tex®. This application is discussed in a separate section (section 1.4).

The non-polymeric PFASs may be released during use of textiles and later in the life cycle of the textiles; this is the reason that the following includes a brief introduction to the substance group. As mentioned, reference is made to the LOUS report for more details.

For different purposes, a variety of PFASs are used. They differ in the functional groups (sulfonic acids, carboxylic acids, alcohols, acrylates, etc.), the length of the carbon chains and the number of fluorinated carbon atoms. PFASs can be divided into:

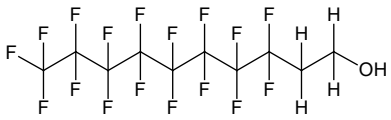
- Perfluoroalkylated substances having a chain of carbon atoms, wherein all hydrogen atoms are substituted by fluorine (= perfluorinated substances).
- Fluorotelomers and other polyfluoroalkylated substances, in which not all carbon atoms have fluorinated constituents, but the substances still contain a part in which the carbon chain is perfluorinated.

An example of a perfluorinated compound, which as mentioned is characterized by a chain where all carbon atoms are fully fluorinated, is illustrated by the structural formula of perfluorooctanoic acid (PFOA) below. Perfluoralkyl substances are of particular interest because the substances are persistent and toxic. The perfluorocarboxylic acids form the group of PFASs which, together with the fluorotelomer alcohols mentioned below, are found in the largest concentrations in textiles.



Perfluorooctanoic acid (PFOA)

An example of a fluorinated telomer (where only some of the carbon atoms in the chain are fluorinated) is 8:2 fluorotelomer alcohol (8:2 FTOH), as shown below. This substance contains a perfluorinated carbon chain of 8 carbon atoms, but also has a portion consisting of two carbon atoms which are not fluorinated. The perfluorinated part is highly stable, which means that the 8:2 FTOH could degrade in the environment to form the persistent perfluoroalkylated substances PFOA and perfluorononanoic (PFNA). Thus, the fluorotelomer acts as a precursor to the formation of the perfluoroalkylated substances.



8:2 fluorotelomer alcohol (8:2 FTOH)

In some contexts, the term "perfluorinated substances" is also used as a synonym for all polyfluoroalkyl substances, even if not all of the associated carbon atoms are fluorinated. In this report, the term “polyfluoroalkyl substances (PFASs)” is used for all the substances together.

The substances in the report are mainly indicated by abbreviations derived from substances' English names. A list of abbreviations, full substance names and CAS numbers is shown in Annex 1. Over time, many different abbreviations have been used for the same substances. In this report, chemical names and abbreviations are used in accordance with the terminology proposed in 2011 by an international group of scientists (Buck *et al.*, 2011). The same abbreviations are used in the LOUS survey of PFASs from 2013 (Lassen *et al.*, 2013). For the acids, the same abbreviations are used for the acid and the acid residue ion. In describing the substances, depending on the context, the names of the two forms are used. As an example, PFOS is indicated as perfluorooctane sulfonic acid or perfluorooctane sulfonate.

C_8 versus C_{4-6} technology

PFASs, used as the basis for the production of PFAS-based impregnation agents, have varying chain lengths. A distinction is often made between impregnating agents based on long-chain PFASs (with 8 perfluorinated carbon atoms (C_8) or more) and impregnating agents based on shorter-chain PFASs (with 6 perfluorinated carbon atoms (C_6) or less). PFOS, PFOA and 8:2 FTOH are examples of substances used in long-chain PFASs technology (C_8 technology). Traditionally, the impregnating agents were based on C_8 technology. In recent years, industry has moved away from the C_8 technology towards C_{4-6} technology, as the short-chain technology has so far has been considered to be less problematic (Buck *et al.*, 2012).

1.3 PFAS-based impregnation agents

The non-polymeric PFASs are not used intentionally in textiles; therefore, manufacturers, importers and retailers of textiles will consequently not have knowledge of the presence of these substances in the finished textile products. They would rather have knowledge of the types of impregnating agents used in the products. To get an overview of the PFAS-based impregnating agents that may be present in the products, a survey has been undertaken of the most common PFAS-based impregnating agents on the market. It also examined the knowledge available about the composition of the agents, as this could indicate which PFASs may occur in the finished products and, consequently, the potential for release by degradation of the PFAS-based polymers.

The following figure illustrates how the PFAS-based polymers are typically composed on the surface of the finished fabric. On a backbone of carbon atoms, which cross-links to form a two-dimensional polymer structure upon curing, both polyalkylfluorinated and non-fluorinated side chains are attached. The fluorinated side chains can be of varying lengths in the individual product. The chains are attached to the backbone by ester bonds.

The impregnating agents are applied as a thin film on the surface of the fabrics, usually in combination with other finishing agents, by a process in which the polymerization and curing occurs on the surface of the fabric (Knepper *et al.*, 2014). The side chains of the two-dimensional polymer structure formed during cure protrude as small hairs from the material, and aid in providing the dirt and water repellent effect.

By the finishing process, the polymers bind to the surface in an amount corresponding to 0.2 to 0.5 % by weight of fabric (fibres). This process results typical in a total fluorine concentration of 0.04 to 0.25 % of the fibres, equivalent to 400-2,500 mg/kg (Knepper *et al.*, 2014).

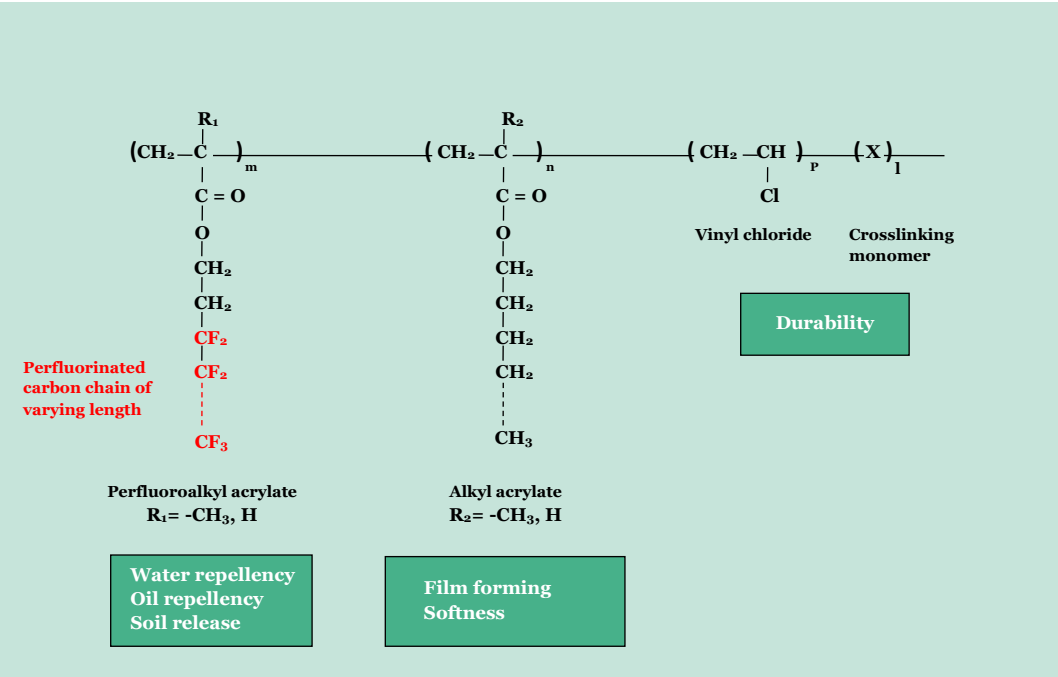


FIGURE 1
POLYMER STRUCTURE OF OIL AND WATER REPELLANTS FOR TEXTILES (BASED ON DAIKIN, 2009) THE PERFLUOROALKYL MOIETY OF THE POLYFLUOROALKYLATED SIDE CHAIN IS MARKED IN RED. THE PRODUCT IS MARKETED AS A FLUOROALKYL ACRYLATE COPOLYMER (BASERET PÅ DAIKIN 2009).

The composition of commercial products is confidential, and the exact composition of the individual impregnating agents is therefore not known. It is known, however, that the impregnating agents consist of very complex substances and polymers. This composition may, for example, be "bulky"-

substituted perfluoroalkyl sulfonamides or polyfluoroalkylated (telomer) acrylates, urethanes, phosphates or sulfonates. Impregnation agents also contain traces of more simple precursors such as perfluoroalkyl sulfonamides or fluorotelomer alcohols of different lengths.

Marketed surface treatment agents for textiles
A wide range of PFAS-based coatings for textiles are marketed.

The Knowledge Centre for Smart Textiles at Creative Business Centre in Denmark in 2012 compiled a list of coatings for textiles, including agents based on (co)polymers with perfluorinated side chains (VIT, 2013). Information on PFAS-based agents has been collected in Appendix 4, where the information from VIT (2013) is supplemented with information on other agents obtained from the Internet.

The following manufacturers and brands have been identified:

- DuPont/Huntsman: A number of brands in which Teflon® fabric protector or Oleophobol® are included in the name.
- BigSky Technologies LLC: GreenShield®
- BASF: Lurotex Protector RL ECO ®
- Pulcra Chemicals: Repellan KFC®
- Rudolf Group: Rucostar® EEE6; Bionic Finish®, Ruco-coat®, Ruco-protect®, Rucotec®
- Daikin: UnidyneTM
- Nano-Tex: Resist Spills™, Resists Spills and Releases Stains™
- Nicca: NK Guard S series
- 3M: Scotchgard™ Fabric Protector

These brand names have been used for an internet search for textiles that were treated with PFAS-based impregnation agents (detailed in section 2.2.2), and have been included in a questionnaire (discussed in section 2.2.1) whereby traders of textiles for children have been asked to whom they sell products treated with these brand names.

In the search for impregnating agents, information has been sought on ingredients, but as mentioned it has been difficult to get accurate information on the composition of the impregnating agents. For Oleophobol, for example, the only available information is that it is based on short-chain perfluorinated substances. Another example is Scotchgard by 3M, which was initially based on the PFOS-derived substances (Jensen *et al.*, 2008). According to the information in the MSDS, Scotchgard ™ Fabric Protector contains <3 % "fluorochemical urethane" and the CAS number is indicated as "Trade secret" (3M, 2014). As there is no obligation to register polymers under REACH, it is not possible to obtain information from registrations.

Product chains of polymers with polyfluoroalkylated side chains
As there is no knowledge about the composition of the impregnating agents, it has been attempted to obtain further information on possible residual PFASs in the finished products by looking at the product chains for these substances and the composition of the raw materials.

Figure 2 below illustrates a product chain for polymers with polyfluoroalkylated side chains. The polyfluoroalkylated intermediates in this product chain produced by Dupont under the trademark Capstone. These intermediates are used by Huntsman to produce the polymers. The final curing of the polymers (creating "cross-links") takes place at the manufacturer of the fabric. In this product chain, the treated clothing can be branded as protected with "Teflon Fabric Protector". The manufacturers of the fabric will likely only know that Phobol impregnating agents are used, but they will

not have knowledge of the exact composition of the impregnation agents, and furthermore will not have any knowledge of the possible content of non-polymeric PFASs in the finished products.

It should be noted that Teflon® Fabric Protector is a PFAS-based surface coating and thus essentially different from the material used for example in Teflon® pans. "Teflon" is part of the trade names for many different fluorine-based products manufactured by Dupont, and is used for example for the company's polytetrafluoroethylene (PTFE, used for non-stick pans and certain membranes, is further discussed in section 1.4).

FIGURE 2
EXAMPLE OF PRODUCT CHAIN OF POLYMERS WITH POLYFLORALKYLATED SIDE CHAINS FOR TEXTILES (DUPONT, 2013)

The above product chain is likely to be fairly representative of the product chain for imported products. There is no information to suggest that there will be significant differences between products produced in Europe and products produced in China or elsewhere in Asia. Based on information from the Chinese authorities reported in the LOUS survey (Lassen *et al.*, 2013), a very large portion of the polyfluorinated substances used for textiles in China is imported from foreign manufacturers.

However, it cannot be excluded that in certain places of the world, significantly different impregnation agents are used. It is possible that PFASs, which is not bound to the polymer, may be used for the manufacture of textiles. It must be expected that PFASs, which are not bound in the polymer, will be extracted more easily from the products. The results of previous studies of PFASs in textiles, as referenced in Section 1.5.2, however, do not provide any indications that some products stand out significantly from the rest.

Composition of raw materials - Raw materials and intermediates for production of the impregnating agents may be present as residues in the finished textile products. The products, which are used as raw materials in the production of polymers, are typically composed of PFASs of different chain lengths, as illustrated in Appendix 5. The table above shows products that are marketed by DuPont for the production of PFAS-based impregnating agents. The iodides are used (through different intermediate reactions) to form the individual side chains by reactions which form a polyfluoroalkyl acrylate (Appendix 5). In production, intermediates of different chain lengths are formed, and these may to some extent be present as residues in the final products. This is discussed further in Section 1.5.2. It is seen that Capstone ™ 62-I, which is primarily based on C_6 technology and marketed as such, contains 2-10 % perfluorooctyl ethyl iodide (C_8-fluoroalkyl) and up to 6 % perfluorodecyl ethyl iodide (C_{10}). This helps to explain that in studies of marketed textile products, PFASs with varying chain lengths are found in the individual products - as further discussed in section 5.4, which describes the results of the present study.

1.3.1 Studies of PFASs in impregnation agents for textiles

To reiterate, the composition of PFAS-based impregnating agents are confidential, but a number of studies have examined which PFASs can be found in the impregnating agents. These studies describe the composition of the agents before polymerization. The substances which do not react in the course of the polymerization and curing may be present in the finished textile products.

A previous study of impregnation fluids in the Swedish market has shown a high content of FTOH of between 0.2 and 9 g FTOH/L (200,000 to 9,000,000 µg/L) (Society for Nature Conservation, 2007).

In a Canadian study of seven waterproofing products for textile finishing, FTOHs were found in six of the products. FTOH accounted for up to 3.8 % by weight of the fluorine content of these products. In the last of the seven agents, PFOS precursors (N-alkyl-perfluorooctane sulfonamide) comprised 0.4 % of the fluorine content of the agent (Dinglasan-Panlilio *et al.*, 2006).

A later study of PFASs in consumer goods in Norway and Sweden from 2009 examined five impregnation products used by consumers for finishing of textiles (Herzke *et al.*, 2009). The five impregnating agents all contained PFASs. It is not clear which technologies the impregnating agents were based on, but it is possible that these agents for finishing are different from those used in the production of impregnated textile, as other techniques are used for the finishing. Three of the agents contained relatively high concentrations of FTOHs, as also seen in the other studies. The results for substances above the detection limit are shown in Table 1.

It is generally recognised that FTOHs are found in the highest concentrations in impregnation agents for textiles. The perfluoroalkyl carboxylic acids PFOA, PFNA and PFDoDA are also found in lower, but still significant concentrations.

Of importance for the discussion later in this report (section 6.5.3) is the high content of volatile FTOHs in these products as compared with the content of the non-volatile perfluoroalkyl carboxylic acids. The same difference is not found in studies of PFASs, which may indicate that significant quantities of FTOHs may be released at an early stage after the production of the textiles.

TABLE 1
PFASs IN IMPREGNATION AGENTS (HERZKE *ET AL.*, 2009).

Substance name	Acronym	Concentration of extracted PFASs from the 5 impregnation agents µg/L
6:2 Fluorotelomer alcohol	6:2 FTOH	535 – 13.250
8:2 Fluorotelomer alcohol	8:2 FTOH	54.780 – 330.800
10:2 Fluorotelomer alcohol	10:2 FTOH	17.800 – 120.721
Perfluorobutanoate	PFBA	75-142
Perfluorohexanoate	PFHxA	23-25
Perfluoroheptanoate	PFHpA	6-54
Perfluorooctanoate	PFOA	26-208
Perfluorononanoate	PFNA	593
Perfluorodecanoate	PFDA	168
Perfluorododecanoate	PFDoDA	1.200

For the following substances the concentration was below the detection limit in all agents: 6:2 FTUCA, 8:2 FTUCA, 6:2 FTSA, 8:2 FTSA, FOSA, PFPeS, PFHxS, PFHpS, PFOS, PFDS, PFPA, PFTrDA, 4:2 FTOH, MeFOSE, EtFOSE.

1.4 Fluoropolymers used in textiles and footwear

Fluoropolymers are to some extent used in clothing for the purpose of forming a membrane that prevents water penetration, and at the same time makes the material breathable. The fluoropolymers are plastics in which the fluorine atoms as directly attached to the backbone of carbon atoms. Certain types of fluoropolymers may contain residues of PFASs, which are used as processing aids in the production process. PTFE is a fluoropolymer which has traditionally been produced by polymerizing tetrafluoroethylene/-ethene with the sodium salt of PFOA (Na-PFOA) or of PFNA (Na-PFNA) as a processing aid. The product may therefore contain residues of PFOA and PFNA from the production process (Lassen *et al.*, 2013).

Gore-Tex® material contains a perforated membrane of PTFE. There are several equivalent materials, for example CHAMFAB®. In addition, some technical textile materials are made from with fibres of PTFE but these are not likely to be found in textiles for children. PTFE is also used for many other purposes as previously mentioned, including non-stick pans (also known as "Teflon pans" with reference to DuPont's trademark).

Traditionally, studies have been able to demonstrate small amounts of PFOA in PTFE. However, in recent years there has been a shift away from the use of Na-PFOA as a processing aid. Gore, which manufactures Gore-Tex®, completely eliminated the use of PFOA in the manufacture of the materials as of 2013 (Gore, 2014). Today, an alternative aid is used, but it has not been possible to find information about this. The product is referred to, however, as PFOA-free and not as PFAS-free, which could indicate the possible use of shorter-chain PFASs. An inquiry has been addressed to the international FluoroCouncil, which represents the main producers of fluorinated substances, but the Council has not been able to provide information on the alternatives used.

In analysis of clothes with Gore-Tex® membranes, discussed later in Section 1.5.2, various PFASs have been found. However, none of the studies exclusively reported the analyses on the membrane. An interpretation of the results is difficult, as clothes with Gore-Tex® membranes are also treated

with PFAS-based impregnating agents to make the surface water and dirt repellent (Gore, 2014). According to Gore (2014), PFOA-free impregnation agents have been used since 2011. The agents are based on perfluorinated molecules with short carbon chains (C_6 or less) as opposed to the longer-chain carbons (C_8 - C_{12} two), which were typically used in the past (Gore, 2014).

1.5 PFASs in finished textiles

1.5.1 Sources of PFASs in the textiles

PFASs in the finished textiles originate from different sources. There are basically four options:

1) The substances are traces of unreacted raw materials, intermediates or substances unintentionally formed during the production of the polymers or during finishing of the textiles;
2) The substances are side chains which split off through hydrolysis of the ester bond between the side chains and the polymer;
3) The substances are non-polymeric PFASs, intentionally used as part of the impregnating agent (no recent examples identified).
4) The substances are residues of processing aids from the production of fluoropolymers (PTFE).

It is not possible to analyse the content of PFAS-based polymers in the textiles. The non-polymeric PFASs extracted from the textiles, however, can be analysed.

The origin of the substances is of importance in relation to the interpretation of the measured migration rates and particularly in relation to the interpretation of the possible exposures and releases to the environment by subsequent waste treatment.

If the measured PFASs in the textiles are traces of intermediates, it should be expected that releases measured by migration tests represent a "first flush" and the migration rates would decrease after the first wash. If, on the other hand, the releases are representative of degradation products, the formation may occur throughout the life of the product (e.g. induced by UV light, wash and wear). If the side chains are degradation products, it is very likely that a substantial portion of the perfluorinated side chains by the degradation, for example, can be split off by the long-term degradation of the polymers. The measured content will therefore not fully express the potential for release of PFASs later on in the lifecycle of the textiles. The potential will be roughly estimated from the total fluoride content, under the assumption that total fluoride in the product is present in the perfluorinated side chains and potentially can be split off as PFASs, as the perfluorinated chain is more stable than the bond to the polymer. This assumption is discussed further in relation to the results of the washing tests in section 5.2.4.

1.5.2 Studies of PFASs in textiles

Several studies have demonstrated that a number of PFASs can be extracted from textiles in relatively low concentrations. The results are usually presented in $\mu g/m^2$ surface of the fabric. Strictly speaking, this measure cannot be indicated as the "concentration" of PFASs in the material (which should e.g. be expressed in mg/kg), but since it has been common practice in most of the literature, this measure is also used in the present study.

Over the years, a development in which PFASs are being used has taken place. Formerly, the precursors of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) were the substances most used, as they have optimum surfactant properties as concerns impregnation compared to other PFASs. Use of impregnating agents based on C_8-polyfluoroalkyl substances has either been

banned (agents based on PFOS and its precursors), or is being phased out voluntarily by the industry (agents based on PFOA and its precursors) in recent years. Findings that are a few years old may therefore not be representative for the situation today.

Results of three studies of the Norwegian environmental authorities (SFT, 2006), Norges Naturvern-forbund (2006) and Greenpeace (2012), are summarized in the substance groups in Table 2. Results and detection limits for individual substances are shown in Appendix 9. The table was developed during Phase 1 of the project (several studies have been published later and presented later in this section), and has been used to identify the PFASs most appropriate to analyse in the present study. In order to provide an easy overview, the table lists the total concentrations of the individual groups of PFASs (for example, total concentration of FTOHs). As later noted in the health assessment (Chapter 6), such totals do not have any relevance for health and environmental assessments, as the properties of the individual substance depend on the length of the perfluoralkyl chain (for example, the properties of 4:2 FTOH are different from the properties of 8:2 FTOH).

FTOHs are found in the highest concentrations in most studies, but the variation between samples is large. As demonstrated by the analyses of the individual substances in Appendix 9, it is 8:2 FTOH and 10:2 FTOH especially which are found in high concentrations in many products. 6:2 FTOH occur in fewer products (including many products where 8:2 FTOH and 10:2 FTOH not occur) and generally in lower concentrations, whereas 4:2 FTOH is below the detection limit in all products. FTOHs are found in all types of clothing.

Furthermore, concentrations above 30 µg/m² are found for the sum of fluorotelomer acrylates (FTACs) (a few examples), fluorotelomer sulfonic acids (FTSAs), perfluoralkane sulfonic acids (PFSAs) (a few examples), perfluoroalkyl carboxylic acids (PFCAs) (widespread), and perfluoralkane sulfonamides and -amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs) (a few examples).

In most studies, FTOHs and PFCAs account for by far the largest fraction of the total content of PFASs, but there are differences as concerns dominant chain length. Results of individual studies are briefly described below.

In a study of PFASs in 11 fabrics (mainly all-weather clothing) marketed in Norway in 2006, conducted by the Norwegian environmental authorities (SFT, 2006), extracts of 11 impregnated textiles from snowsuits, ski wear, sports/all-weather jackets for adults and children, and a tablecloth were analysed for 27 different PFASs. The survey found the following levels of unbound, non-polymeric PFASs in the textiles (number of measurements above the detection limit in parentheses): FTOHs: 0 -10,683 µg/m² (10), FTSAs/FTCAs: 0-6 µg/m² (7) , PFSAs: 0-31 µg/m² (9) PFCAs: 3-170 µg/m² (11), FASAs/FOSE: 0-23 µg/m² (8), PFOS: <0.02- 30 µg/m² (9), PFOA: 0.4- 34 µg/m² (11). Note small differences in the use of abbreviations, compared to the original source. Here, the abbreviations specified in Appendix 1 are used.

A more recent report by Greenpeace (2012) found similar levels as the above-mentioned studies in 13 pieces of outdoor clothing purchased in Germany, Austria and Switzerland. In this study, MTBE was used as an extraction solvent. The fluorotelomer-derived PFASs dominated in this study, and 6:2 FTOH was the most common raw material to replace 8:2 FTOH. Precursors to PFOA (8:2 FTA/FTOH) could still be found in some products. The highest measured concentration of PFOA was 5 µg/m² but the sum of all PFCAs reached 11 µg/m². The highest concentration of FTOHs was 352 µg/m² for 6:2 FTOH and 240 µg/m² for 8:2 FTOH. PFOS was not found in any of the samples. The study included clothing containing Gore-Tex® (which also has a dirt-repellent PFAS-based coating) and clothing with dirt and water repellent impregnation. The Greenpeace study - besides substances included in the Norwegian study - also analysed for three fluorotelomer acrylates (6:2 FTAC, 8:2 FTAC, 10:2 FTAC), which were found in a few items.

A small survey conducted by Greenpeace in Switzerland examined three rain jackets for children for a variety of chemicals, including perfluorinated compounds (Greenpeace, 2013). The concentrations found were at the same level as in the previous larger Greenpeace study (Greenpeace, 2012).

In a study from the Czech Republic of 40 samples of indoor and outdoor textiles, all extracts contained fluorinated substances (Becanova *et al.*, 2013). PFOS was demonstrated in nearly all samples. In one material from sportswear the concentration of PFOS was above the EU limit value of 1 µg/m². In two samples of furniture textiles the concentration of PFOA was 15-30 µg/m², and in 10 composite textile samples, PFHxA was found in concentrations of 2-16 µg/m².

A German study published in 2013 analysed the extracts of five samples of all-weather clothing, including rain pants and mittens for children (Schlummer *et al.*, 2013). In four of the samples the content of the sum of FTOHs varied between 20 and 40 µg/m², while the fifth sample had a very high content of nearly 300 µg/m². In all cases, 8:2 FTOH dominated, followed by 10:2 FTOH, as also seen in the studies shown in Appendix 9 and summarized in Table 2.

After the preparation of Phase 1, two new German studies (Knepper *et al.*, 2014 and Dreyer *et al.,* 2014) were published. They are addressed in more detail in the discussion of the results of the analysis programme of the present study and briefly described below.

In a study of Knepper *et al.*, (2014), FTOHs accounted for the vast majority of the total content of PFASs in 13 of 15 tested outdoor jackets. In 12 of the samples, 8:2 FTOH was the dominant FTOH followed by 10:2 FTOH. The concentration of 6:2 FTOH was negligible in these 12 samples, while one sample stood out, as 6:2 FTOH was the dominant PFAS. In this sample, the concentration of PFHxA was at the same level as the concentration of PFOA, while in most of the other samples it was much less. In most samples the dominant PFCAs were PFOA and PFDA. They examined jackets purchased during the period August 2011 to March 2012.

In another new German study of 16 outdoor jackets and gloves (Dreyer *et al.*, 2014), a somewhat different pattern was found. It is not stated when the clothes are purchased, but the analyses were conducted in October 2013 and the jackets were probably purchased immediately before. For the extraction of acids, methanol was used, while for the extraction of volatile PFASs, MTBE (methyl *tert*-butyl ether) was used. As in the other studies, PFCAs and FTOHs were the dominant groups of PFASs, but C_{4-6} chemistry was more prominent, while C_8 chemistry represented a minor part. In seven of the samples, the concentration of PFOA was above 1 µg/m². In four samples PFHxA was the dominant PFCA, and in 3 PFBA was found in the highest concentration. This pattern was even more pronounced for the FTOHs, where 6:2 FTOH was found in concentrations higher than 8:2 FTOH in 10 of the 15 samples, while 8:2 FTOH appeared in the highest concentrations in 5 samples. The total concentration of FTOHs was generally significantly higher than the levels found by Knepper *et al.* (2014), but similar high levels have previously been found in Norwegian studies (SFT, 2006). Dreyer *et al.* (2014) thus found a total concentration of FTOHs in 6 of 15 samples exceeding 500 µg/m² with the highest level being approximately 1,700 µg/m².

None of the studies have analysed the total content of fluorine (F), and it is therefore not possible to estimate how much of the total content of fluorinated substances is represented by the analysed PFASs.

TABLE 2
RESULTS OF ANALYSES OF PFASs IN TEXTILES.

Substance group	Substance	Concentration (µg/m²)	Number of products (above the detection limit)	Types of textile products	Source	Comments
Fluorotelomer alcohols (FTOHs)	Total FTOHs (sum 4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH)	<dg -10,683	10/11	Ski and sportswear for children and adults	SFT, 2006	Most fabrics have concentrations between 30 and 400 µg/m², only a single jacket contained 10.7 mg m². In 9 out of 11 products 8:2 FTOH was the substance found in the highest concentrations.
	Total FTOHs (sum 4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH)	27.1 - 1001	6/6	Rain jackets and anoraks	Norges Naturvern-forbund, 2006	8:2 FTOH was the main FTOH in all products.
	Total FTOHs (sum 4:2 FTOH, 6:2 FTOH, 8:2 FTOH, 10:2 FTOH)	<dg – 464.2	8/14	Outdoor clothing for children and adults	Greenpeace, 2012	6:2 FTOH has been the dominant FTOH in several products (9-11, 12), which indicates that several manufacturers have started using this substance instead of 8:2 FTOH
Fluorotelomer acrylates (FTACs)	Total FTACs (sum of 6:2 FTAC, 8:2 FTAC, 10:2 FTAC)	<dg – 78.3	11/14	Outdoor clothing for children and adults	Greenpeace, 2012	FTA are intermediates in polymer production
Fluorotelomer olefins (FTOs)	10:2 FTO	<dg	0/11	Ski and sportswear for children and adults	SFT, 2006	
	10:2 FTO	<dg – 1.11	3/4	Rain jackets and anoraks	Norges Naturvern-forbund, 2006	
Fluorotelomer sulfonic acids (FTSAs) and fluorotelomer -carboxylic acids (FTCAs)	Total (sum of 6:2 FTSA, 8:2FTSA, 6:2 FTCA, 8:2 FTCA)	<dg – 124,2	7/11	Ski and sportswear for children and adults	SFT, 2006	FTS can be degraded to perfluoroalkyl sulfonates
	Total (sum of 6:2 FTSA, 8:2 FTSA, 6:2 FTCA, 8:2 FTCA)	<dg – 3.87	4/6	Rain jackets and anoraks	Norges Naturvern-forbund, 2006	

Substance group	Substance	Concentration (µg/m²)	Number of products (above the detection limit)	Types of textile products	Source	Comments
Perfluoroalkane sulfonic acids (PFSAs)	Total (sum of PFBS, PFHxS, PFOS, PFDS)	<dg – 30.5	9/11	Ski and sports wear for children and adults	SFT, 2006	PFOS was not detected in samples from 2012, reflecting the ban from 2008.
	Total (sum of PFBS, PFHxS, PFOS, PFDS)	0.02 – 23.3	6/6	Rain jackets and anoraks	Norges Naturvern-forbund, 2006	
	Total (sum of PFBS, PFHpS, PFHxS, PFOS, PFDS)	<dg	0/14	Outdoor clothing for children and adults	Greenpeace, 2012	
	Total (sum of PFBS, PFHpS, PFHxS, PFOS, PFDS)	<dg	0/3	Rain jackets and anoraks	Greenpeace, 2013	
Perfluoroalkyl carboxylic acids (PFCAs)	Total (sum of PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA)	2.97 - 170	11/11	Ski and sports wear for children and adults	SFT, 2006	Used for the production of fluoropolymers such ad Teflon® fabric protector. Is an indication that there is used a fluorine-based impregnation or membrane. Also occur as degradation products of telomeres. Perfluoroalkyl carboxylic acids are partially water soluble and can be expected to be released to the environment during cleaning. PFOA was detected in all samples from both 2006 and 2012.
	Total (sum of PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA)	1.89 - 428	6/6	Rain jackets and anoraks	Norges Naturvernforbund 2006	
	Total (sum of PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA)	0.66 – 10.96	14/14	Outdoor clothing for children and adults	Greenpeace, 2012	

Substance group	Substance	Concentration (µg/m²)	Number of products (above the detection limit)	Types of textile products	Source	Comments
	Total (sum of PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTeDA)	<dg – 5.48	2/3	Rain jackets for children	Greenpeace, 2013	
Perfluoralkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)	Total (sum of FOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE)	<dg – 22.8	8/11	Ski and sports wear for children and adults	SFT, 2006	FOSA and MeFOSE were measured most frequently and in the highest concentrations. In the samples from 2012 (Greenpeace), none of these substances could be quantified, possibly due to the relatively high detection limits in the study.
	Total (sum of FOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE)	1.43 - 107	6/6	Rain jackets and anoraks	Norges Naturvernforbund 2006	
	Total (sum of FOSA, MeFOSA, EtFOSA, MeFOSE, EtFOSE)	<dg	0/14	Outdoor clothing for children and adults	Greenpeace, 2012	

1.5.3 Studies of degradation of PFAS-based polymers in textiles

As mentioned, PFASs may be released from the finished fabrics by hydrolysis of the ester bond between the polyfluoroalkylated side chains and the backbone of the polymer. Data are available showing the extent to which this occurs in the course of the life of the textiles and subsequent waste treatment, but a recent study illustrates the potential for formation of PFASs by degradation of the impregnation agents. In addition, some scenarios for the release from the degradation have recently been published (Wang *et al.*, 2014b), as discussed in more detail in section 7.6.2.

Eschauzier and Knepper (2013) examined the content of PFASs in two impregnating agents: Capstone (DuPont), which is used for textiles, and Cartafluor CFI (Clariant), which is used for paper. Only the results for Capstone will be discussed here. The analysed Capstone was of a new type and based on the C_{4-6} technology.

In the study the polymers were oxidized so that the bond between the PFAS-based side-chains and carbon backbone of the polymer was broken. Thus, it was possible to demonstrate the quantities of PFASs that could potentially be released from the impregnating agents by decomposition.

There was a marked increase in the concentration of a number PFASs in the extraction solvent (methanol) after oxidation. The concentration of the three perfluoroalkyl carboxylic acids PFBA (C 4), PFPA (C5), and PFHxA (6) increased from about 1 µmol/mL without oxidation to 5,000-10,000 µmol/mL after oxidation. Similar effects, but only resulting in concentrations of 1 to 100 µmol/mL, were observed for a number of other PFASs. The results show that the potential for release of PFASs through decomposition of the impregnating agents are much higher than the quantities that can be immediately extracted with methanol. This is discussed further in relation to the handling of waste in section 2.3.

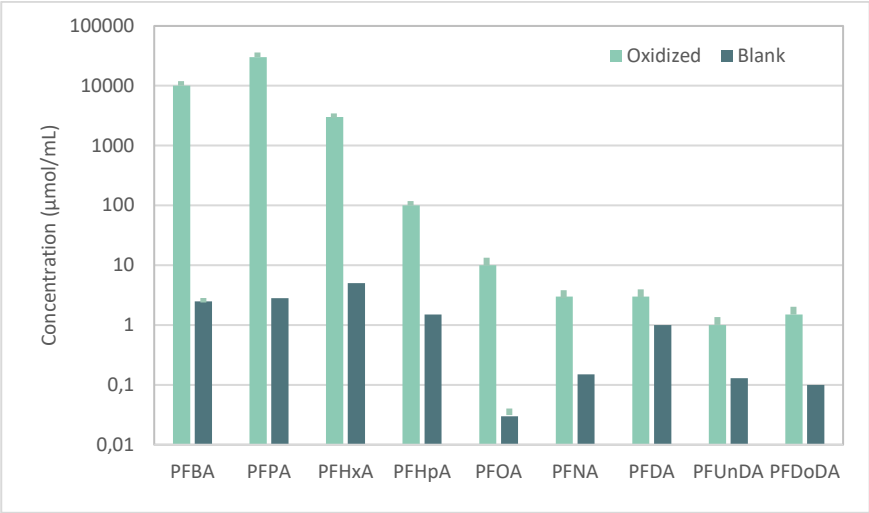


FIGURE 3
PERFLUORINATED SUBSTANCES EXTRACTED FROM CAPSTONE BEFORE (BLANK) AND AFTER OXIDATION (AFTER ESCHAUZIER OG KNEPPER, 2013). NOTE LOGARITHMIC SCALE.

1.6 Current regulation

PFOS and its derivatives are included in the list of restricted substances under the Stockholm Convention on Persistent Organic Pollutants with some specific exemptions and acceptable applications. None of these exemptions and acceptable applications includes the use of textiles.

The same substances are restricted via the POP Regulation (Regulation (EC) no. 850/2004 on Persistent Organic Pollutants (POPs) with a few specific exemptions in the EU. PFOS and derivatives may not be used for textiles in concentrations above 10 mg/kg or in quantities of more than 1 µg/m² of the surface of the treated material.

PFOS furthermore has a harmonized classification under the CLP Regulation (Regulation (EC) no. 1272/2008 on classification, labelling and packaging of substances and mixtures, etc.) (discussed further in the health assessment). For the other PFASs, there is no restriction in the EU; these substances do not have a harmonized classification under CLP.

PFOA and its salt APFO are included in the Candidate List[1] under REACH because of their CMR properties. Four other long-chain perfluoroalkyl carboxylic acids are included in the Candidate List due to their PBT properties: PFUnDA, PFDoDA, PFTrDA, PFTeDA, collectively referred to as C_{11}-C_{14} PFCAs. None of the substances are yet on the list of substances subject to authorization (Annex XIV of REACH).

A harmonized classification of PFOA and APFO has been proposed, but the substances still do not have a harmonized classification under CLP regulation.

For two other long-chain perfluoroalkyl carboxylic acids, a proposal for harmonized classification as toxic to reproduction is in the pipeline.

In Norway, production, import, export and sale of PFOA (+ salts and esters) in consumer goods has been prohibited from 1 June 2014. Since the ban has proven difficult to implement, a transitional arrangement for products produced or imported before 1 June 2014 was established. These may be sold until 1 January 2018 (Environment Directorate, 2014a, 2014b). According to the ban, no part of a textile, carpet or other consumer product may contain more than 0.1 % (by weight) PFOA or more than 1 µg PFOA/m^2 (Norway, 2013).

[1] The Candidate List is a list of substances that may have serious effects on human health or the environment and includes candidate substances for possible inclusion in the Authorisation List (Annex XIV).

2. Survey

2.1 Methodology

2.1.1 Internet search for textile products for children, which may contain PFASs

As mentioned in Chapter 1 and shown in Appendix 4, a number of PFAS-based impregnation agents for textiles are on the market, including:

- Scotchguard®
- Teflon® fabric protector
- NanoTex®
- GreenShield®
- Lurotex®
- Unidyne®
- Crypton Green®

In addition to the PFAS-based impregnating agents, PFASs may also be present in the membranes of the plastic type of polytetrafluoroethylene (PTFE) used for waterproof clothing. The best known of these membranes are:

- Gore-tex® (også Gore Tex, Gore-Tex)

PTFE may contain residues of PFOA and PFNA from the production process, although alternative processing aids are generally used at present (whose chemical composition is not known).

An internet search was conducted using search profiles that combine the above trademarks with textile products for children (snowsuits, infant sleeping bags, etc.) as this might indicate that the products are treated with PFAS-based impregnating agents (see further description in section 1.3). The search showed that it was only possible to find children's products which specified that they contained Teflon® fabric protector or Gore-Tex®. Therefore, further searching concentrated on these brands.

Subsequently, the following internet searches were performed:

- Site-specific search on Google on a range of Internet stores that sell children's clothing and other products for children (i.e. the search included on all pages on the individual stores' websites). On the websites of eight stores it was specifically stated that some products contained "Teflon", and a full search was made of these sites (i.e. all pages which include the search terms were read). It was noted how many products were listed with Teflon® or Gore-Tex® (for more details see section 2.2). For product types known to contain impregnating agents, the total numbers of products on the website were also counted (for more details see section 2.2).

- Search for clothes on the websites of outdoor stores' by combining the search terms "Teflon" or "Gore-Tex" with boys* and girls* as these stores were expected to sell clothes with impregnation.

The latter search was unsuccessful. The websites of the outdoor stores also had very few textile products for adults indicated as Teflon®-treated. The reason is likely that treatment with Teflon®

fabric protector or other PFAS-based impregnating agents is not included in the marketing and the description of the products.

2.1.2 Information from market actors

Danish children's shops and children's clothing manufacturers were identified by searching the company database Kompass and through an Internet search. In all, 55 companies were contacted by email and/or phone and sent a questionnaire to answer. The questionnaire is shown in Appendix 3.

2.2 PFASs in textile products for children on the Danish market

2.2.1 Questionnaire

Of the 55 companies contacted, about half responded to the inquiry, while only 17 returned a completed questionnaire. Several companies answered that their participation would not be relevant, either because they were not dealing with products treated with perfluorinated substances, or because they had no products for children in their range.

Among the companies that returned completed questionnaires were the expected major players in the market. Most companies answered that their children's products were not treated with perfluorinated substances.

Six of the seventeen companies that returned a completed questionnaire informed the authors that they sell products for children that are treated with fluorine-based technology. Two of these companies answered that their products were Teflon® treated and a single company stated that the garment is treated with a water repellent C_6-based agent from a Japanese manufacturer of the product series NK guard S (see Appendix 4). The other four indicated that they do not know what PFAS-based impregnating agents the products are treated with. Among the reported impregnated product types are:

- Snowsuits
- Outdoor and rainwear
- Skiwear
- Gloves
- Boots
- Bags
- Backpacks.

None of the companies surveyed indicated that they used PFAS-based impregnation agents for baby and childcare products, such as recliners, wear belts or children's car seats, but companies specializing in childcare products generally had no knowledge of whether there were PFAS-based impregnating agents in their products. The same result was obtained with respect to childcare articles from the responses from a similar, ongoing study on chemicals in car seats and other textile products for children.

Four companies responded that they previously had used fluorine-based technology to achieve water and/or dirt-repellent properties, but that now they had switched to a PFAS-free alternative, Bionic Finish® Eco. Bionic Finish® Eco is based on an alternative polymer technology (dendrimer technology) and, according to the manufacturer, does not contain PFASs or other fluorine compounds (Rudolf Group, 2014a, 2014b). Products treated with this technology are water-resistant, but not oil and dirt repellent to the same extent as products treated with PFAS-based agents.

Four of the companies surveyed, in part the same as reported having products treated with PFAS-based impregnating agents, referring to the Oeko-Tex criteria as the key restriction on the use of

PFASs in their products (Oeko-Tex criteria restricts certain PFASs in the textiles). Three companies informed the authors that they have a policy that clothes must not contain C_8-based impregnating agents (but allows the use of impregnating agents based on short-chain PFAS) technology.

The listed clothing brands with PFAS-based preservative represent products from the upper and the mid-price segment, which is in line with the fact that treatment with PFAS-based impregnating agents is a relatively expensive surface treatment method.

A number of companies informed the authors that they have nursing pads or nursing cushions that are stain and water resistant without PFAS technology. An internet search indicates that nursing pads and nursing cushions indicated to be water-resistant are typically made with a water-resistant plastic material on the surface (referred to as polyester, acrylic coated, film coating, coated cotton satin, etc.). It cannot be ruled out that some nursing pad or nursing cushions made with PFAS technology are marketed, but it is likely that a large number of products should be examined in order to find products where the technology is used.

Share of outdoor wear with water and dirt repellent impregnation
The majority of respondents (63 %) responded that a water repellent finish would be present in 50-75 % or > 75 % of all children's clothing for outdoor use, but at the same time 10 out of 17 companies answered that they do not have products with PFAS-based impregnation agents. Of the products from the six companies that responded that they market clothes impregnated with PFASs, probably only some of the products contain PFASs (not specified in the answers).

2.2.2 Internet search

An Internet search of products with "water and dirt repellent properties" for children identified the following product types:

- Snowsuits (all-in-one infant suit)
- Rainwear
- Skiwear
- Softshell jackets
- Gloves and mittens
- Sportswear
- Boots and shoes.

These are essentially the same products listed in the responses to the questionnaire.

Subsequently, an internet search was carried out covering eight internet shops offering children's clothing where one or more products are indicated to contain the trademarks Gore-Tex® or Teflon® (typically the brand name Teflon® is used without "fabric protector" on the websites). Only one of the eight stores had answered the questionnaire. A search with the keywords "Teflon" and "Gore-Tex" (with various spellings and with and without ®) was performed on the websites of the stores using a Google site-specific search (i.e. all pages on the website of the store). Some internet shops do not indicate whether their products are treated with Teflon®; among these are two companies which indicated that they have products with Teflon® in the questionnaire. For the eight companies that declare that the products are Teflon-treated, it is believed that the majority of the Teflon-treated products would indicate this. It is therefore assumed that the proportion declared to be Teflon®-treated roughly reflects the proportion of products that actually are Teflon®-treated.

As shown in Table 3, according to the search on the eight online shops, primarily outerwear such as snowsuits, skiwear, gloves and jackets are sold with Teflon® treatment. The table shows the total number of products (defined with photos on the store's website) with and without indication of Teflon® treatment. It was not examined whether there is product overlap between the stores, but in

this case, it is assumed that any overlap would be the same for products with and without Teflon® fabric protector.

The table also shows the number of identified products with Gore-Tex®. Only footwear with Gore-Tex® was identified. The percentage of the footwear declared to contain Gore-Tex® was not been examined, as footwear with Gore-Tex® is beyond the scope of the study.

TABLE 3
NUMBER OF HITS REGARDING CHILDREN'S PRODUCTS TREATED WITH "TEFLON" AND "GORE-TEX" FROM SEARCHES OF EIGHT INTERNET SHOPS.

Product type	Number of products with Teflon®	Number of products with Gore-Tex®	Total number of products *1	Percentage with Teflon®
Snow-suits Bantings?	31	0	624	5 %
Gloves/mittens	11	0	191	6 %
Caps	1	0	617	0 %
Jackets	13	0	1,564	1 %
Rain suits	0	0	321	0 %
Ski pants	4	0	90	4 %
School bags/backpacks	0	0	238	0 %
Rain cover for baby carriages	3	0	66	5 %
Infant sleeping bags	2	0	59	3 %
Footwear	0	61		
Total	65	61	3,770	

* The products are counted on the basis of photo documentation on the pages so that all images are considered to represent a product; for example, same style but in a different colour).

The identified products treated with Teflon or Gore-Tex are much the same as indicated in the replies to the questionnaire. However, none of the eight Internet shops sell rain suits or bags (specified in the questionnaire) indicated as Teflon®-treated (but they can be sold without it being specified).

Other children's equipment claimed to be Teflon®-treated
Apart from the products mentioned above, a comprehensive internet search identified the following products for children (one of each type except for nursing bags, of which there were several), where it is indicated that the textiles are treated with Teflon®:

- Awning for pram
- Stroller
- Harness
- Nursing bag (but not the nursing mat itself).

2.2.3 Summary of the survey of children's clothing with PFAS-based impregnating agents

It is difficult to estimate the proportion of outdoor wear for children which are treated with PFAS-based impregnating agents based on the responses received and the internet search, as products

may well be treated with PFAS-based impregnation agents without it being indicated on the product.

Outdoor wear
It is estimated that the total percentage of snowsuits, gloves and skiwear treated with PFAS-based impregnating agents is likely to be around 10-30 %, while the percentage for rainwear would be somewhat lower. The percentage is expected to be at the high end for distributors specializing in brands that use PFAS-treated clothing, while for others which deliberately avoid PFAS-based impregnating agents, it would be zero.

Given the size of snowsuits and the large number sold, the snowsuits are presumed to represent the largest tonnage. Most children aged 1-7 years have a snowsuit which is changed regularly. Trade statistics from Statistics Denmark do not provide specific information about import of snowsuits or other children's clothing; therefore it has not been possible to estimate the total tonnage. Over the past seven years, on average 61,000 children were born each year in Denmark (Statistics Denmark, 2013). Assuming all children aged 1-7 years receive a new snowsuit each year and a proportion of these are probably reused, the consumption would be around 200,000-300,000 suits a year. If 10-30 % of these contain PFAS-based impregnating agents, about 20,000 to 90,000 suits a year would contain PFASs. In section 2.2.3 on waste streams, the quantities of PFASs that could potentially be disposed of as regards treated children's clothing and other textiles for children are estimated.

Bags
School bags and backpacks with PFAS-based impregnating agents are occasionally identified, but are not common.

Infant sleeping bags
A few examples of infant sleeping bags treated with PFAS-based impregnating agents are identified, but this usage does not seem to be widespread.

Other products
For the other products mentioned, the percentage of clothing treated with PFAS-based impregnating agents is estimated to be less than 10 %.

2.2.4 Textiles with fluoropolymers for children on the Danish market

Based on interviews with selected market actors and internet searches it appears that, apart from footwear, a very small number of products containing Gore-Tex® for children are marketed. A few high-end snowsuits, skiwear and jackets use Gore-Tex®, but they represent only a small market share. One of the major supermarket chains informed the authors that they do not have a single product containing Gore-Tex® for children. In an internet search shown in Table 3, only footwear containing Gore-Tex® was identified. As previously mentioned, the footwear is not the focus of this survey, and Gore-Tex® membranes are currently made without the use of PFOA (but may contain other PFASs).

2.3 Mapping of waste streams

There are no specific surveys of the disposal of children's clothing in Denmark.

Quantities disposed of
Overall figures for the disposal of clothing in Denmark are taken from the report "Toward a Nordic textile strategy" (Palm *et al.*, 2014). In Denmark, around 89,000 tonnes of new clothes are sold every year. Around 41,000 t/year of used clothes are collected by different organizations. Of the collected 41,000 t/year, or about 23,000 t/year is exported and 12,000 t/year is reused/recycled (a small portion of this is recycled – this is estimated at 1,000 t/year) and 6,000 t/year is incinerated.

With a total consumption of clothing, incl. reuse of 100,000 t/year (89,000 + 11,000), around 59,000 t/year (100,000 to 41,000 + 6,000) is estimated to be sent directly to incineration without prior collection.

The final disposal of the 89,000 t/year new clothes must, therefore, be considered to be:

- Incineration: about 65,000 t/year
- Export: about 23,000 t/year
- Recycling (for waste cotton used e.g. in the auto repair shops): ca. 1,000 t/year

It has not been investigated as to what extent the textiles used as cotton waste in garages after use is disposed of as hazardous waste (because it is oil saturated) or disposed of for municipal solid waste incineration.

No specific estimates of the amount of children's clothing are identified. There is a widespread reuse of children's clothing among families and friends (about 60 % is handed down to family and friends, Tojo *et al.*, 2012), which are be covered by the available surveys, but in this context this may be regarded simply as an extension of the first-time use of the clothing.

Other products with surface treated textiles for children are to some extent reused in Denmark, whereas exports of these products must be considered to be negligible. It is estimated that the products will ultimately end up in waste incinerators.

Exposure related to the disposal routes
Waste incineration - Virtually all surface-treated clothing and other articles with surface-treated fabric that is not exported for reuse outside Denmark (especially developing countries), ultimately end up in waste incinerators via the collection of domestic waste.

Recycling - There is some recycling of textiles for production of cotton waste being used, *inter alia* in auto repair shops, instead of cloth. For this purpose, pure cotton textiles are used, and the surface-treated fabrics are not suitable for this purpose.

Reuse in Denmark - Reuse in Denmark can be considered a life extension of the articles, and there are no particular exposure routes for reuse.

Export for reuse - Clothing exported for reuse abroad is assumed to be disposed of to landfill and less controlled waste dumps. The problems associated with the disposal of reused clothing are not different from the problems associated with disposal of new clothing exported to these countries and is discussed separately below.

Quantities of PFASs
There are no inventories of the total consumption of PFASs in articles in Denmark, but based on the information in the LOUS survey (Lassen *et al.*, 2013), the total content of PFASs in products sold in Denmark is estimated to be around 14-40 tonnes per year. Clothing represents about 50 % of the consumption of fluorotelomer and fluorotelomer-based polymers worldwide, and this is probably also the case in Denmark. As fluorotelomer and fluorotelomer-based polymers account for the majority of the consumption of PFASs, the total consumption of PFASs in textiles is probably around 10-20 t/year. Note that the quantities indicate the total weight of both PFAS-based polymer (the major part of the total weight) and non-polymeric PFASs in the products.

Of this, children's clothing probably only accounts for a small part. In recent years, PFASs-based coatings largely have been replaced by some of the largest manufacturers of children's clothing. It is

estimated that today, only 10-30 % of outdoor wear for children are coated with PFAS-based coatings. Leisure and outdoor clothing for adults still appear to a larger extent to be treated with PFAS-based coatings (assuming the situation in Denmark is similar to the situation in Germany described by Knepper *et al.*, 2013) as the major brands of outdoor clothing are just now investigating the possibility of replacing PFAS-based impregnation agents.

The children's clothing currently disposed of may be assumed to contain more PFAS-based impregnation agents than the clothing currently sold.

The trade statistics include specific data on the quantities of clothing for children (the statistics are broken down by sex), and surface treated textiles are typically reported together with non-treated textiles in subcategories covering all outerwear or rainwear.

It is estimated that, on the present basis, the total quantity of PFASs which in the future will be disposed of with children's clothing (based on the present consumption) would be around 1-10 tonnes/year (note that the amount includes both PFAS-based polymers and non-polymeric PFASs).

Fate by waste incineration- The main issue in relation to the disposal of textiles with PFAS-based coatings in Denmark is to what extent the substances are degraded by waste incineration.

The LOUS survey (Lassen *et al.*, 2013) provides a review of the knowledge in the field. The present results indicate that there is an effective destruction of PFASs at temperatures around 1100 °C, while there is more uncertainty about the destruction efficiency at temperatures down to 850 °C, which can occur in waste incinerators for municipal waste (although Danish plants today usually operate with higher temperatures). PFASs that are not destroyed by incineration are likely to be captured by the flue gas treatment system and end up in the flue gas cleaning products, similarly to dioxins and furans. There are no actual measurements of the cleaning efficiency for the substances. Since the PFASs are relatively large compounds, it is reasonable to assume that they are adhered to the activated carbon and other particles in the exhaust gas and captured in the filters of the incinerators. The same assumption is made in a new publication that discusses uncertainties in a new estimate of global emissions of PFASs (Wang *et al.*, 2014b).

It is consequently believed that the combustion of textiles with PFASs in incinerators is not a significant source of emissions of PFASs to the environment.

Fate by landfilling - Clothing exported for reuse abroad must be considered to be ultimately disposed of in uncontrolled landfills and dumps or spread in an uncontrolled manner in nature.

The non-polymer-bound PFASs (similar to the substances which could be extracted from the textiles) must be considered to be released from uncontrolled landfills to the air (FTOHs and other volatile PFASs) relatively quickly or can be discharged with penetrating water (the other non-polymeric PFASs). Ahrens *et al.* (2011) have shown that the air over two investigated landfills in Canada contained three to thirty-fold higher concentrations of PFASs (primary FTOHs) than the reference locations, and calculated a total emission of PFASs into the air from the two landfills of respectively 0.1 and 1 kg/year). It must be anticipated that a larger proportion of the PFASs in the textiles are emitted to air from uncontrolled landfills without land cover (which is common in developing countries) than from land covered, sanitary landfills in industrialized countries.

Buser and Morf (2009) review existing knowledge on PFASs in the leachate from landfills as part of a Swiss substance flow analysis for selected PFASs. The dominant PFASs are perfluoroalkyl carboxylic acids (especially PFOA) and perfluoralkane sulfonic acids (especially PFOS). On the basis of the review, the authors estimate that emissions of PFOA may correspond to 10-30 % of quantities disposed of in the landfills. Under controlled conditions, as is the case in Switzerland (and many EU

countries) percolate is not discharged directly into the environment. Under uncontrolled conditions, which would be the typical situation in developing countries, the percolate will typically be discharged untreated to surface water or groundwater.

The main issue, however, is to what extent the surface coatings of the textiles will be degraded with subsequent releases of PFASs in the long term. The potential for release of PFASs from textiles is, as discussed in Section 1.5.3, thousands of times greater than the amounts that can be immediately extracted (and immediately washed out with leachate). No studies have been identified that model the long-term emissions from landfills.

A new inventory of the sources of the total global emissions of PFASs (Wang *et al.*, 2014a) does not include emissions from landfills. The authors mention in a parallel publication that discusses the gaps in knowledge that PFAS-based polymers deposited could possibly be biodegraded, but that it is not possible to estimate the scale due to uncertainties about half-lives of the degradation that can range from thousands of years to decades (Wang *et al.*, 2014b).

Regardless of the time perspective, the fact remains that textiles account for almost half of the global consumption of PFASs (as discussed in Lassen *et al.*, 2013). In the long term, textiles may potentially be a major source of emissions to the environment in countries outside waste and thereby to the global environment (furthermore, uncontrolled waste dumps have a limited lifetime before they become effectively a very polluted part of nature).

3. Initial hazard screening and development of exposure scenarios

3.1 Initial health hazard screening

During phase 1 of the study, an initial health hazard screening was drawn up. The purpose of this screening was primarily to identify the substances that would be most relevant to analyse for and identify the main exposure scenarios for choosing the migration tests. The screening was also taken as a basis for the further health assessment. To avoid unnecessary repetition, the majority of the screening has been moved to the health assessment in Chapter 6, where it is further developed. Therefore, only a brief summary describing the basis for the choice of substances for the chemical analysis and the choice of migration tests is provided below. Reference is made to the health assessment in Chapter 6 for references to the cited literature.

Summary of screening of health effects

Of the many hundreds of polyfluoroalkyl substances (PFASs) used in society, sufficient data are only available for perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) and to some extent perfluorohexane sulfonic acid (PFHxS) to carry out fairly comprehensive health assessments. This statement applies to both epidemiological studies where these three substances normally are found in the highest concentrations in blood of the surveyed populations, and to animal studies, in which these substances are the most studied. There have been more studies of precursors in recent years, i.e. substances that can be degraded/metabolised to PFOS (EtFOSE, MeFOSA etc.) or PFOA (8:2 fluorotelomer). There also tend to be significantly more studies of the analogues with shorter perfluoroalkyl chains used as substitutes for C_8 fluorinated substances.

PFASs are shown to be effectively absorbed after oral ingestion or inhalation in animal studies, whereas the substances were absorbed through the skin to a lesser extent. This generally applies to all the investigated PFASs. In the blood, the substances are bound to the protein albumin. Although PFASs occur in most tissues, they accumulate primarily in the liver. In rodents, PFASs are excreted very quickly (hours/days) in the kidney with the urine, whereas virtually no excretion is observed in humans, due to a re-absorption of the PFASs in the kidneys. Extrapolation of the results of animal tests to humans is therefore unreliable.

In population studies, a correlation between exposure to PFOS, PFHxS, PFOA or PFNA and hormone disorders, behavioural problems in children, reduced fertility, kidney disease and effects on the immune system and foetal development exposure scenarios have been demonstrated.

The toxicity of PFASs increases with the length of perfluoroalkyl chain, but the number of studies of the long-chain PFASs (with perfluorinated chain lengths longer than 8 carbon atoms) is limited.

Although sufficient data are available to conduct a comprehensive health assessment for only a few of the substances, it was decided to analyse all the substances that are likely to occur in the textiles.

In the health assessment in Chapter 6, it is assessed to what extent it is possible to aggregate exposures to more substances and prepare an assessments for a broader group using "read-across" or "worst-case" assessments.

Preliminary consideration regarding exposure scenarios
Previous studies of exposure of children to PFASs are discussed in the health assessment in Section 6.4. An estimate of the average exposure of the European population to selected PFASs is shown in Table 4. For the non-volatile PFASs, diet is the main source. House dust is another significant source. For the more volatile PFASs, indoor air is an important source.

TABLE 4
EXPOSURE TO SELECTED PFASs FROM DIFFERENT SOURCES (FROMME *ET AL.*, 2009)

Exposure source	Daily intake (pg/kg bw)							
	PFOS		PFOA		ΣFTOHs		ΣFOSE/FOSA	
	Average	Max	Average	Max	Average	Max	Average	Max
Indoor air	0.9	0.9	4.7	4.7	38	105	460	2,050
Outdoor air	1.3	12	0.1	1.0	3.0	3.2	1.1	12
House dust	16	1,028	32	4,217	103	1,017	983	2,033
Food	2,817	11,483	1,500	4,483	-	-	217	6,866
Drinking water	22	87	23	130	-	-	-	-
Total intake	**2,857**	**12,611**	**1,560**	**8,836**	**144**	**1,125**	**1,661**	**10,962**

The survey includes only some of the main PFASs, and does not take into account the specific exposure conditions for small children in the indoor environment. The survey has not included direct exposure to PFASs in consumer products that can originate from PFAS-treated clothing, as limited data regarding such a contribution are available. The data in the table are consequently insufficient for estimating the total exposures, but the data are used later in the health assessment to compare with the calculated intakes of PFASs from impregnated clothing.

The textile products for children which most often contain PFASs are snowsuits, ski suits and all-weather jackets/trousers and rainwear with corresponding mittens, caps and hats. When the children wear the clothes, the direct exposure routes will occur from dermal contact with the substances in textiles and exposure via saliva by children who suck on the textiles. By skin contact, some limited absorption after release of the substances from contact with the clothes may take place; this also includes the use of mittens to wipe the face. Activity and sweat production is likely to increase the absorption of the substances. A less common, but direct exposure route, occurs if the child sucks/licks the clothes, including the mittens.

When the clothes are used, stored indoors or dried, the volatile PFASs (dependent on the temperature) may possibly evaporate (e.g. fluorotelomers and sulfonamides) to indoor air. In addition, textile dust, wherein the less volatile PFASs (salts, carboxylates and sulfonates) are concentrated, and the more volatile substances are condensed by temperature decreases (Ahrens *et al.*, 2012), also forms. Children's clothing will therefore contribute to the overall general PFAS exposure indoors with indoor air and house dust.

Selection of migration tests - Based on the above assessment, it was prioritized to undertake tests of the migration of PFASs to saliva since PFASs are easily absorbed when taken orally. From other studies, some knowledge of the emission of volatile PFASs from the clothes to the environment exists. As the available results suggest that the majority of the volatile PFASs in the clothes are

released during the life of the clothing, it was considered that further studies of releases to the air would not contribute significantly. It was therefore decided to base the estimations of the possible release to air on the results of other studies, as discussed further in Section 6.4.3. .

3.2 Initial environmental hazard screening

In part 1 of the project an environmental risk screening was drawn up. The purpose of this screening was primarily to identify the substances it would be most appropriate to analyse for and to establish the main exposure scenarios. The screening also formed the basis for further environmental assessment of the project. To avoid unnecessary repetition, the majority of the screening was moved to the environmental assessment in Chapter 7, where it has been further developed. Consequently, a brief summary describing the basis for the choice of substances for the chemical analyses and the exposure scenarios is provided below. References to the cited literature are found in the environmental assessment in Chapter 7.

Environmental fate and effects

A literature-based review of properties, fate and effects of PFASs in the environment has been undertaken as part of the LOUS survey (Lassen *et al.*, 2013). The initial screening has been based on this review.

PFOS and PFOA are the best-described PFASs in the literature. Both substances are considered to be very persistent in nature. They are, in practice, neither biodegradable aerobically nor anaerobically, and they do not degrade abiotically by hydrolysis and only marginally by photolysis. PFOA is more water-soluble than PFOS and undergo long-range transport to an even greater extent than PFOS via water systems, including ocean currents.

The substances EtFOSE and MeFOSE that are precursors for PFOS may undergo primary degradation, both biologically and by hydrolysis, and are thereby transformed into PFOS. Because they can decompose and ultimately be converted into PFOS, the half-life in the atmosphere of EtFOSE is relatively short (half-life of 16 hours).

Long chain (C_{11}-C_{14}), perfluorinated carboxylic acids (PFCAs) have fewer associated studies, but for four of the substances, it is concluded that they meet the PBT criteria for persistence and bioaccumulation, and are thus regarded as vPvB substances.[2]

Perfluorinated carboxylic and sulfonic acids with shorter chain lengths, especially C_4-based substances such as PFBS, have proven not to be bioaccumulative, as they are rapidly eliminated from the organisms in which they are taken up. Still, however, they are persistent in the environment.

Fluorotelomer alcohols (FTOHs) such as 6:2 FTOH exhibit a higher degree of primary reactivity than the corresponding carboxylic acids, but are converted to either the corresponding carboxylic acid or, under certain circumstances, to a carboxylic acid with a slightly longer chain length in the microbial environment. It is shown that 6:2 FTOH can be degraded to PFHpA (heptanoate) or PFHxA (hexanoate), but these substances undergo further degradation afterward.

The best-characterized of the substances, such as PFOS and PFOA, are not very toxic to organisms in the environment based on standard toxicity tests, but there are some indications that they have certain endocrine disrupting properties. There are virtually no data on the ecotoxicity of the short-chain PFASs.

[2] vPvB = Very persistent and very bioaccumulative. Is along with PBT (persistent, bioaccumulative and toxic) properties which under REACH make substances considered of particular concern.

Environmental exposure
Releases of PFASs to the air take place very slowly through migration to and evaporation from the surface of the impregnated fabric. Some substances have such low vapour pressure that the release is minimal. The release will also increase proportionally with the ambient temperature. In addition, PFASs could be released from the textiles during use when exposed to rain or, to a lesser extent, by wear and tear. It is considered that these types of release primarily result in exposure of the soil environment to PFASs. Finally, PFAS-impregnated clothing could be released to the sewage system through washing of the textiles.

It is estimated that environmental exposure by waste disposal of the impregnated textile is low, as the textiles are expected ultimately to be disposed of to waste incineration plants. Here, efficient combustion at high temperature and the cleaning of flue gas together ensure that only negligible amounts of PFASs are emitted to the atmosphere. It cannot be excluded that low residue concentrations could be detected in slag and flue gas cleaning products. This applies to textiles disposed of in Denmark; see the description of the particular conditions for textiles exported, in section 2.3.

Environmental hazard assessment
The environmental hazard of PFOS, PFOA and other PFASs is based primarily on the substances' persistence and potential for bioaccumulation, not least among top predators in marine food webs. Furthermore, for some substances a potential for endocrine disrupting effects has been demonstrated, although the likelihood is that this will only be expressed at higher concentrations than are normally found in the environment. It is unlikely that short-term effects (acute toxicity) could occur at concentrations that are realistic to expect in the environment.

Selection of tests and substances for analysis
The initial environmental risk screening gave no reason to limit the number of PFASs included in the analysis programme. It is projected that, as a consequence of the presence of PFASs in impregnated textiles for children, the most significant environmental load would occur in connection with the washing of textiles and by evaporation to the atmosphere.

When washing the textiles, the laundry water is subsequently discharged to the wastewater system from which residues in sludge (especially) could end up in soil and residues in the treated wastewater could end up in streams and/or coastal marine waters. It was therefore chosen to examine the extent to which the substances are potentially released to wastewater by washing.

As mentioned above, it was decided not to investigate the release to the atmosphere from impregnated textiles within the framework of the project, as this has been investigated in several previous investigations. The assessment of environmental exposure by evaporation will therefore be based on these studies (further detailed in section 6.4.3 and 7.6.2).

4. Selection of substances and products for analysis

4.1 Substances

In addition to an initial screening for total fluorine, which was used as a basis for selecting products to undergo the more expensive analyses for all PFASs, a number of PFASs were selected for chemical analysis of the content of the products. The substances were primarily selected on the basis of the substances found in previous studies, and substances which are expected to occur in the treated fabrics. Although some of these substances have more serious health and environmental effects than others, a decision was made that it would be inappropriate to limit the number of substances included in the analyses.

The analytical methods used are summarized in the table below and described in detail in the next chapter.

TABLE 5
PFASs SELECTED FOR CHEMICAL ANALYSIS. CAS NUMBERS AND CHEMICAL NAMES ARE SHOWN IN APPENDIX 1.

Substance group	Substance	Reference standard	Group/analysis method
Perfluoroalkane sulfonic acids (PFSAs)	PFBS PFPeS PFHxS PFHpS PFOS PFDS	Ja	Acid/LC-MS-MS
Perfluoroalkane sulfonamides and amidoethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)	FOSA MeFOSA EtFOSA MeFOSE EtFOSE	Ja	Neutral substances/GC-MS PFOSA was analysed with LC-MS-MS. MeFOSA and EtFOSA was analysed with both GC-MS and LC-MS-MS
Perfluoroalkyl carboxylic acids (PFCAs)	PFBA PFPA PFHxA PFHpA PFOA PFNA PFDA PFUnDA PFDoDA PFTrDA	Ja	Acid/LC-MS-MS PFBA could not be analysed

Substance group	Substance	Reference standard	Group/analysis method
Fluorotelomer alcohols (FTOHs)	4:2 FTOH 6:2 FTOH 8:2 FTOH 10:2 FTOH	Ja	Neutral substances/GC-MS
Fluorotelomer carboxylic acids (FTCAs)	4:2 FTCA 6:2 FTCA 8:2 FTCA 10:2 FTCA	No	Neutral substances/GC-MS Possible to use FTOH for quantification
Fluorotelomer methacrylates (FTMACs)	8:2 FTMAC 10:2 FTMAC	Yes	Neutral substances/GC-MS
Fluorotelomer acrylates (FTACs)	8:2 FTAC 10:2 FTAC	Yes	Neutral substances/GC-MS
Fluoretelomerolefins (FTOs)	10:2 FTO	No	Could not be analysed
Fluorotelomer sulfonic acids (FTSAs) and fluorotelomer carboxylic acids (FTCAs)	4:2 FTSA 6:2 FTSA 8:2 FTSA	Yes	Acid/LC-MS-MS
	6:2 FTCA 8:2 FTCA 10:2 FTCA	Yes	Acid/LC-MS-MS

4.2 Products

Twenty-two products were purchased and initially analysed for the content of total fluorine. Added to this were further eight samples from car seats, purchased as part of another study of chemicals in car seats, which was also carried out by DEPA. The distribution of samples by product groups is shown in the following Table 6.

It should be emphasized that the purpose of the analysis program was not to examine the frequency of PFASs in the various types of products in the Danish market. The number of purchased products is too small to obtain a statistically significant result, since the frequency of products with PFASs for the majority of products is relatively small. If, for example, 20 % of the snowsuits contain PFASs, there would still be a fairly large uncertainty associated with the observed rate, even if snowsuits accounted for all 30 analyses. Products from manufacturers not known to use PFAS-based impregnating agents were not purchased.

The primary objective is to examine the concentration of the different PFASs in the textiles of products treated with PFAS-containing impregnating agents. Likewise, the aim is to find products that are suitable for the migration and wash tests. The study provides significant new information as regards migration and washing tests in relation to previous investigations of PFASs in textiles.

As can be seen from the survey of the market, it is mainly in those cases where Teflon® Fabric Protector technology is used that the products are claimed to be treated with PFAS-containing impregnating agents. The reason may be that Teflon® fabric protector technology is more widely used, but may also be due to the fact that the trademark Teflon® is known from many different applications.

The study therefore aimed at identifying products with other PFAS-based impregnating agents. Four of the six companies that informed the authors they market PFAS-impregnated products did not indicate which trademarks were used. The selected products therefore include a number of products treated with Teflon® fabric protector, a number of products with impregnating agents from the four companies that replied that they do not know the fabric protector brand name, and a number of products from companies that did not respond to the survey, but where a claim is made that the products are water and dirt resistant.

It is assumed that companies that did not reply to the questionnaire either are not aware of the problem, or do not want to answer that they use PFAS-based impregnating agents. There is therefore a good chance that products which are claimed to be water and dirt resistant actually contains PFASs.

It is assumed that products that are treated with Teflon® fabric protector would contain the same substances, whether it is a snowsuit, jacket or gloves, but for the interpretation of the results, this assumption was tested. Therefore, a number of different products treated with Teflon® fabric protector were selected.

It cannot be ruled out that there might be a nursing pad or nursing cushion treated with PFAS technology, but it is likely that a large number of products would have to be tested to find a single such product. The analysis programme therefore focused on outdoor clothing and infant sleeping bags where there is a reasonable chance of finding PFASs.

None of the companies which indicated that they use PFAS technology, but do not know the brand name, sells PFAS-impregnated infant sleeping bags. But a large number of products on the market claim to be stain and water resistant, and given the potential for exposure, it was of particular interest to find out whether these products contain identifiable PFASs.

TABLE 6
PRODUCTS PURCHASED DIVIDED BY PRODUCT GROUPS

Product group	Number of products		
	Teflon® fabric protector	Products with impregnating agents from the four companies that replied that they use PFAS technology, but do not know the brand name	Companies that have not responded to the survey, but where it is claimed that the material is water and dirt resistant
Snowsuit	1	2	2
Jackets	1	2	2
Gloves	1	1	1
Infant sleeping bags	1		3
Rain suit	1	2	2
Auto chairs for children			8
Total	5	7	18

5. Analysis and test programme

The analysis programme was initiated with the analysis of total fluorine in 22 products representing relevant types of children's clothing and infant sleeping bags, as well as in eight samples taken from car seats purchased as part of a survey of chemicals in car seats, also performed for DEPA. The purpose of the total fluorine analyses was to ensure that the products selected for the more expensive analyses of PFASs had a proven content of fluorine (so there was a high probability of finding PFASs in the products). Furthermore, the total fluorine analyses formed the basis for a discussion of the potential for release of PFASs from the fabric during its entire life cycle.

Among the products with fluorine contents above the detection limit, 15 products were selected for further analysis of the content of PFASs. Of these, eight products were further selected for wash tests and analysis of migration of PFASs to saliva.

5.1 Analysis methods and tests

5.1.1 Analysis of total-fluorine in materials

A known subsample of textiles was combusted in a flask containing oxygen. The combustion gas containing fluorine in the form of hydrogen fluoride was recovered in a wash bottle with deionized water. The collection liquid was analysed for fluoride by ion selective electrode. The content of fluorine was determined against the calibration curve. The analysis was carried out as a double determination.

The detection limit of the method is 20 mg/kg. The method was validated by analysis of a reference material, BCR 73, which has a certified content of 12.07 % fluorine. In the two analyses, the detected concentrations were 11.8 % fluorine and 11.7 % fluorine, respectively, resulting in a recovery of 97 %.

5.1.2 Migration tests

The migration fluid (artificial saliva) was prepared using de-ionized water, calcium chloride, magnesium chloride, potassium carbonate, potassium chloride, potassium phosphate, sodium chloride and hydrochloric acid to adjust the pH to 6.8 according to the JRC Report 20001 EUR 19826 EN. The artificial saliva was heated to 37 °C to mimic body temperature.

A subsample of known weight and area were sampled from the textiles. The subsamples were placed in a known volume of the heated artificial saliva in a temperature-controlled oven at 37 °C with static contact with the simulant for three hours.

After three hours of migration to the artificial saliva at 37 °C, the textiles were removed from the migration fluid and the migration fluid was stored refrigerated until shipment in a cooler bag for subsequent analysis occurred.

All the bottles for migration tests were plastic bottles made of polyethylene (PE).

5.1.3 Washing test

For the washing test a simulated washing situation was applied, as tests in a real washing machine would require that many of each item of clothing should be washed together in order to obtain sufficiently high concentrations in the laundry water.

Wash water was made from tap water and detergent "Neutral" for coloured textiles. The laundry water was heated to 40 °C, to mimic the typical wash temperature for outerwear.

A subsample of known weight and area were removed from each product. The partial sample was placed in the heated laundry water, which was shaken, and the sample was placed in a temperature controlled oven at 40 °C for half an hour with continuous agitation, to simulate a washing situation.

After the wash test at 40 °C, the textiles were removed from the laundry water. The laundry water was stored refrigerated until shipment in cooler bag for subsequent analysis occurred.

All the bottles used were made of polyethylene (PE).

5.1.4 Analysis of PFASs in textiles, migration fluid and laundry water

Extraction and analysis of neutral substances (GC-MS)

For the extraction of fabrics (10 cm^2), 2 x 50 mL of ethyl acetate was used. Before extraction, the samples had ^{13}C or deuterium-labelled substances added. The samples were extracted with ultrasound for 30 minutes, and the solvent was evaporated to 1 mL volume with nitrogen. The extract was analysed by gas chromatography coupled to mass spectrometry (GC-MS). Positive chemical ionization (PCI) was used. For each substance, two specific ions were selected for identification. In addition to the two specific ions and their ratio, the chromatographic retention time was also used as an identification criterion.

The concentration of substances in the samples was calculated with linear regression through the execution of a series of calibration standards containing both labelled and unlabelled substances. ^{13}C or deuterium-labelled substances were used to calculate the recovery rates of the corresponding unlabelled substances in the samples.

For analysis of migration samples (synthetic saliva), a 25 mL sample had ^{13}C or deuterium-labelled substances added and was then purified on solid phase columns (SPE, C18). SPE columns were eluted with 10 mL of ethyl acetate, which was evaporated to 1 mL and analysed by the same GC-MS method as used for the content analysis. The same methods were used for concentration and analysis of laundry water (200 mL).

Extraction and analysis of acids and sulfonamides (LC-MS-MS)

For the extraction of fabrics (10 cm^2), 50 + 30 mL of methanol was used. Before extraction, the samples had ^{13}C or deuterium-labelled substances added. The samples were extracted with ultrasound for 30 minutes, and the solvent was evaporated to 1 mL volume with nitrogen.

The extract was analysed by liquid chromatography coupled with dual mass spectrometry (LC-MS-MS). The substances were ionized with negative electrospray ionization (ESI). For each substance, two specific ion-pairs (parent ion/daughter ion) were selected for identification. In addition to the two specific ion pairs and their ratio, the chromatographic retention time was also used as an identification criterion. The concentration of the substances in the samples was calculated with linear regression through the execution of a series of calibration standards containing both labelled and unlabelled substances. ^{13}C or deuterium-labelled substances were used to calculate the recovery rate of corresponding unlabelled substances in the samples.

For analysis of migration samples (synthetic saliva), a 25 mL sample had ^{13}C or deuterium-labelled substances added and was then purified on solid phase columns (SPE, C18). SPE columns were eluted with 10 mL of methanol which was evaporated to 1 mL and analysed by the same LC-MS-MS method used for the textiles samples. The same analysis method was used for concentrating the laundry water (200 mL).

Detection limits and recovery rates
The detection limits are calculated as three times the signal/noise ratio of the peak in the chromatogram of the standard with the lowest concentration, and corrected for recovery of the specific matrix. The volume of the samples analysed in each method were also taken into account. The detection limits for each substance and the various matrices are shown in Appendix 6.

Recovery rates are calculated for spiked samples, which consist of blind-textile (i.e. fabric without PFASs, pre-tested for the presence of PFASs), artificial saliva and laundry water, respectively. Recovery rates for the three matrices are shown in Appendix 7. PFPeS, PFPA, 4:2 FTOH, 6:2 FTOH, 4:2 FTAC, 6:2 FTAC and 4:2 FTSA could not be found in spiked laundry water.

For the PFCAs with the highest concentrations (from PFHxA to PFTrDA), between 90 to 110% of the added amounts was recovered in all three matrices with one exception, for PFHpA in the laundry water. For the analysed FTOHs the recovery from textile and artificial saliva was in the range of 90 to 121 %. The recovery in the laundry water was somewhat lower; for 8:2 FTOH 57 % and 10:2 FTOH 72 %, while 4:2 FTOH and 6:2 FTOH as mentioned not could be recovered from the laundry water. PFCAs 8:2 FTOH and 10:2 FTOH constituted the majority of PFASs in the three media, and the recovery rates therefore give a clear indication that the results broadly reflect the actual content of non-polymer PFASs in the matrices.

Quality assurance
Laboratory blanks consisting of reagents with the labelled standards added were extracted and analysed each time samples were analysed. In addition, the liquids used for migration and washing tests were also analysed to check for any blank values. In addition, recovery samples consisting of spiked textile, artificial saliva and laundry water were prepared.

5.2 Analysis results

5.2.1 Total-fluorine in materials

Results of total fluorine analyses in children's clothing and infant sleeping bags
Total fluorine in concentrations above the detection limit in the outer material was found in 19 of the 22 examined pieces of children's clothing and infant sleeping bags (Table 7). From each of the products two samples were taken, which were analysed independently (double determination). The samples are indicated as Sample 1 and Sample 2 in the table. The concentrations are indicated as mg/kg outer fabric and µg/m² outer shell. Samples were deliberately not taken of the parts of the outer shell, which had labels, buttons, special reinforcements and the like.

The study shows that there is great variation in the content of total fluorine. This result may be partly due to differences in the amount of impregnating agent used in the production of the various textiles and differences in the concentration of fluoride in the impregnating agents. There is no general trend toward higher concentrations in certain product categories.

The contents of the 19 products with concentrations above the detection limit varied from 8,000 to 365.000 µg/m² with an average of about 72,700 µg/m².

The highest values were found in the group of products for which it was specifically claimed that the products are treated with Teflon® fabric protector. In these products the fluoride concentration was above the detection limit in all products and the average of the total fluorine concentration was about 163,000 µg/m². The highest values were found in an infant sleeping bag and in a rain suit.

In the other two groups ("PFASs technology" and "water and dirt resistant"), the averages in products where the concentration was above the detection limit were 55,000 µg/m² (water and dirt resistant) and 29,000 µg/m² (PFASs technology), respectively. Overall, the average in these groups was 43,000 µg/m².

In the group of products where the only information available is that the products are stain and water resistant, the concentration of total fluorine was above the detection limit in 8 of 10 products (80%). Thus, the results indicate that there is widespread use of PFAS-based impregnating agents in products claimed to be stain and water resistant. This information also applies to the companies in the survey which indicate that they are not aware whether their products contain PFASs. As mentioned in the survey (Chapter 2), a majority of manufacturers of children's clothing do not currently use PFAS-based impregnating agents. Impregnated products from these manufacturers are typically claimed only to be water-resistant or waterproof, meaning that the former information on stain repellency should be revoked.

It is indicated in a recent German study (Knepper *et al.*, 2014) that PFAS-based polymers are applied in quantities corresponding to 0.2 to 0.5 % of the fabric, and that this results in a typical fluoride concentration of 0.04 to 0.25 % (equivalent to 400-2,500 mg/kg). The majority of the examined textiles in this study had fluoride concentrations that were slightly lower than 400 mg/kg. The results, however, are generally assessed to be in correspondence with the information from the German study, and there is no basis to conclude that PFAS-treated children's clothing contains lesser amounts of PFAS-containing impregnation agents than outdoor wear for adults.

TABLE 7
RESULTS OF TOTAL FLOURINE (F) ANALYSES IN CHILDREN'S CLOTHING

No	Product type	Total-F content, mg/kg		Total-F content, µg/m²		Weight of material g/cm²	Category
		Sample 1	Sample 2	Sample 1	Sample 2		
F1	Snowsuit	350	350	59.975	59.975	0,017	PFAS technology
F2	Jacket	< 20	< 20	< ca. 3000	< ca. 3.000	not measured	Water and dirt repellent
F3	Softshell suit	330	330	127.797	127.797	0,039	Water and dirt repellent
F4	Gloves	310	300	58.842	56.944	0,019	PFAS technology
F5	Infant sleeping bag	160	170	20.603	21.891	0,013	Water and dirt repellent
F6	Infant sleeping bag	270	290	40.487	43.486	0,015	Water and dirt repellent
F7	Infant sleeping bag	3.000	3.100	358.895	370.858	0,012	Teflon®
F8	Snowsuit	250	270	44.620	48.190	0,018	Teflon®
F9	Rain jacket	290	280	32.697	31.570	0,011	Water and dirt repellent
F10	Rain suit	1.200	1.200	115.496	115.496	0,010	Teflon®
F11	Snowsuit	71	69	7.616	7.401	0,011	PFAS technology
F12	Jacket	120	120	12.335	12.335	0,010	PFAS technology
F13	Softshell suit	< 20	< 20	< ca. 3.000	< ca. 3.000	0,028	PFAS technology
F14	Snowsuit	39	38	6.960	6.781	0,018	Water and dirt repellent
F15	Snowsuit	380	340	157.313	140.753	0,041	Water and dirt repellent
F16	Rain suit	< 20	< 20	< ca. 3.000	< ca. 3.000	0,018	Water and dirt repellent
F17	Rain suit	83	79	19.321	18.390	0,023	PFAS technology
		100	100	16.472	16.472	0,016	PFAS technology
F18	Rain suit	300	300	26.702	26.702	0,009	PFAS technology
F19	Ski jacket	650	540	104.849	87.105	0,016	Teflon®
F20	Mittens	910	850	199.845	186.668	0,022	Teflon®
F21	Infant sleeping bag	170	160	11.047	10.398	0,006	Water and dirt repellent
F22	Mittens	440	450	48.107	49.200	0,011	Water and dirt repellent

Results of total fluorine analyses in children car seats
Textiles from eight car seats were analysed for total fluorine. In all of the seats, the concentration of total fluorine in the textiles was below the detection limit of 20 mg/kg. It is projected that, on this basis, none of car seats were treated with PFAS-based impregnating agents, and they were consequently not analysed for PFASs in the textiles.

5.2.2 PFASs in materials

Based on the results of total fluorine test, 15 of the 22 products were selected for analysis of the content of PFASs. These products were primarily selected from the products which had the highest concentrations of total fluorine. The products were also selected on the basis that all types of products should be represented. A set of gloves and a set of mittens were therefore selected, although the concentration of total fluorine was not among the top 15. The results are shown in Table 8, which indicates the average of the duplicate determinations. All analytical results are shown in Appendix 8.

The total concentration of the investigated PFASs ranged from 18 to 407 µg/m².

The samples of the three textiles with Teflon® brand fabric protector contained (as in the case of total fluorine) high concentrations of PFASs, varying from 120 to 410 µg/m². These three samples were among the four samples with the highest concentrations. In all three samples, 8:2 FTOH accounted for more than 65 % of the concentration of PFASs.

As shown in Figure 4, a weak correlation between the content of total fluorine and the total content of analysed PFASs was seen. The calculated correlation coefficient, R2, is 0.23, which expresses a certain relationship, but due to the low value, the correlation may very well be coincidental.

The content of total fluorine was on average 1,600 times higher than the total content of non-polymer PFASs (calculated from the regression line). The ratio is quite well in accordance with the results discussed in Section 1.5.3, where a study has shown that the content of each perfluoroalkyl carboxylic acid increased up to 10,000 times by the oxidation of the compound between the PFAS-based side chains and the carbon atom backbone of the polymer (Eschauzier and Knepper, 2013).

Samples of fabrics treated with Teflon® fabric protector are indicated by the red marker in Figure 4. These samples do not stand out from the other samples in terms of the ratio between total fluorine and PFASs, and the results suggest that these additives are of the same type as the other, but that more impregnating agent is simply used in the production of textiles.

In Figure 4, the content of extractable PFASs is indicated as the total quantity of substances (in mg/m²). If instead it is selected to calculate how much of the fluorine content the extractable PFAS represents, it is necessary to convert the concentration of extractable PFASs to a concentration of fluoride. Assuming that fluoride on average accounts for 70 % of all PFASs (as is the case with 8:2 FTOH, present in the highest concentrations), it can be projected that the fluorine in the non-polymer PFASs represents about 0.04 % of the fabric's total content of fluorine. If all of the fluoride in the polymers can eventually be released as PFASs by the decomposition of the textiles and the PFAS-containing impregnating agents, there would be a potential for release of PFASs of about 2,500 times higher than the total amount of extractable PFASs.

Eschauzier and Knepper (2013) estimate that fluoride accounts for about 12 % of the weight of PFAS-based (side-chain fluorinated) polymer, and mentions that corresponding values are specified by the manufacturer DuPont. Applying this value, the average fluoride content of 72,700 µg/m² means that the surface is treated with 0.06 mg PFAS-based impregnation agent per cm². The average weight of the textiles is determined at 17.4 µg/m², and the weight of the PFAS-based polymer can thus be estimated at 0.3 % of the weight of the fabric. Knepper *et al.* (2014) indicates that the

weight of the PFASs based polymer typically would be 0.2-0.5 % of the weight of the fabric. The observed value of 0.3% supports the hypothesis that the measured fluorine derives from PFAS-based polymers and not from other fluorine sources.

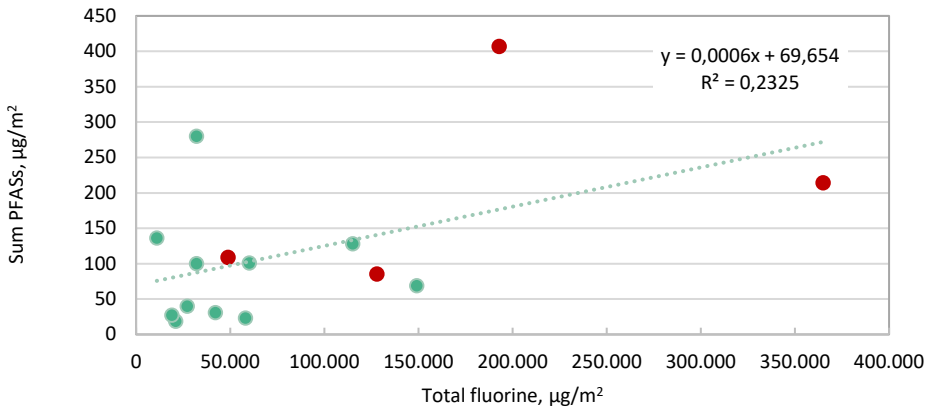


FIGURE 4
RELATIONSHIP BETWEEN TOTAL FLUORINE CONTENT AND THE SUM OF ANALYSED PFASs. SAMPLES OF TEXTILES WITH TEFLON® FABRIC PROTECTOR ARE MARKED IN RED.

The results of the analyses show that fluorotelomer alcohols (FTOHs) were present in the highest concentrations of all products followed by perfluoroalkyl carboxylic acids (PFCAs). Together these two groups accounted for more than 94 % of the total in all products.

The group of PFCAs are of particular interest because they occur at high concentrations in the tested textiles, are persistent and accumulate in the environment. Aside from this, it is these substances that FTOHs decompose into. They are discussed in more detail later in this section.

Fluorotelomer alcohols, FTOHs
FTOHs accounted for 46 % to 99 % of the total content of the analysed PFASs in each of the samples.

Two of the samples (F1 and F2) differed in that FTOHs represented less than 50 % of the total content in these samples, while FTOHs in the other samples accounted for 81 % to 99 %. The two samples with low FTOHs contents were also distinctive in that PFOA represented a relatively large share of the total content of PFASs of 31 % and 15 %, while the share of PFOA in the other samples ranged from 0.02 % to 3.6 %. The composition of F1 is illustrated in Figure 5, together with the composition of F7, which was representative of a typical composition of most of the other samples.

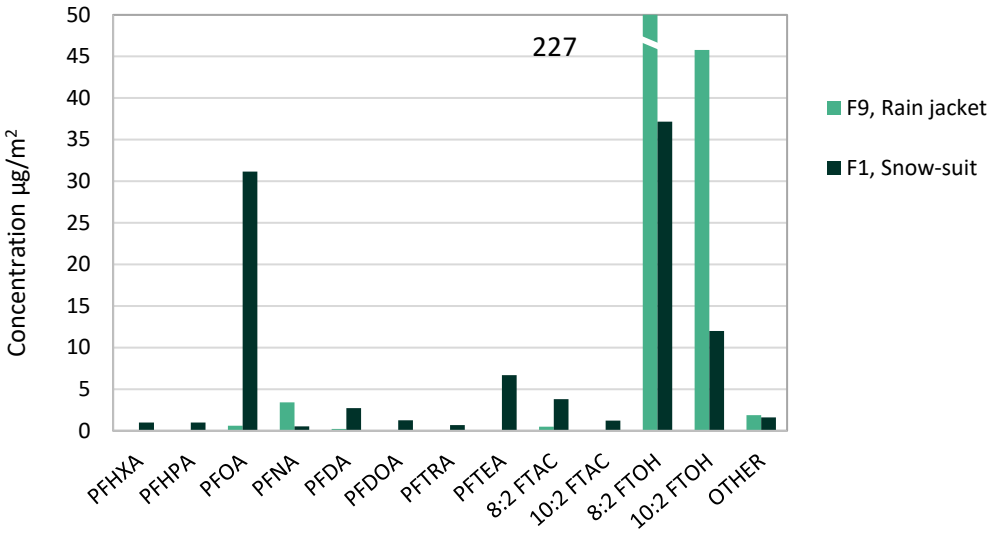


FIGURE 5
TWO EXAMPLES OF COMPOSITION OF PFASs. TOTAL CONCENTRATION OF F1 WAS 101 µg/m² WHILST THE F9 WAS 280 µg/m²

8:2 FTOH was present in the highest concentrations in all samples. The concentrations ranged from 7 to 265 µg/m². This FTOH amounted from between 25 % to 80 % of the total content of measured PFASs in the textiles.

Perfluoralkyl carboxylates (PFCAs)

PFOA was the predominant substance of the PFCAs identified. In two of the samples, PFOA was found in concentrations of 13 and 31 µg/m², respectively, which was significantly higher than the concentrations in the other samples. In five of the samples PFOA were found in concentrations above 1 µg/m² (1.2; 1.7; 3.0; 13.2 and 31.2 µg/m²). Since the PFOA content of the five samples exceeds the Norwegian limit value of 1 µg/m² for PFOA in textiles, the products may not be produced in or imported into Norway.

In several of the products, all 10 PFCAs were found in concentrations above the detection limit. The composition of PFCAs in the two samples with the highest concentrations differed from the other samples, as PFCAs accounted for a significant proportion of the total quantity; this is shown in the figure below (F1 and F3). The figure shows also the composition of the sample F15, which together with F1 and F3 had the highest PFCAs concentrations. As can be seen, the three samples had significant concentrations of PFCAs with longer chains than PFOA. Whether or not these substances occur as traces of intermediate products from the production of the impregnating agents, or are formed by oxidation of the bond to the polymer, it is estimated that the side chains of the polymers must therefore contain similar chains of varying length. Therefore, by degradation of the polymers, PFASs of varying lengths could be released, a result also found in the study summarized in Figure 3 in Section 2.2.7.

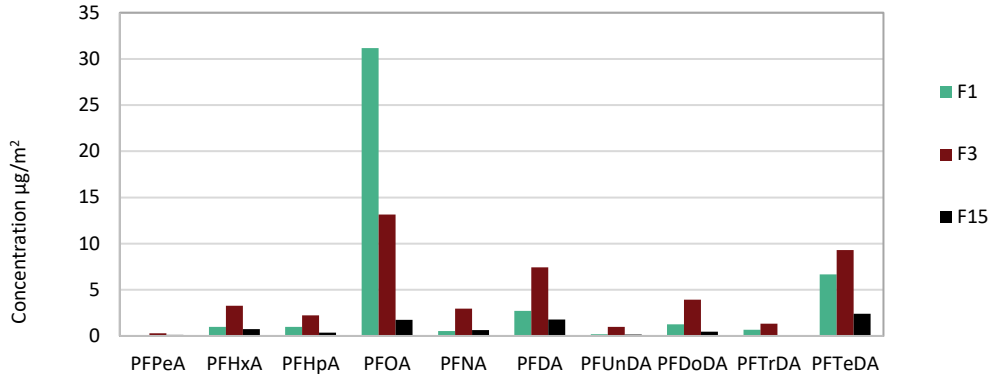


FIGURE 6
COMPOSITION OF PERFLUOROALKYL CARBOXYLIC ACIDS (PFCAs) IN THE THREE SAMPLES WITH THE HIGHEST LEVELS.

The German study of Knepper *et al.* (2014) found a clear correlation (R2 = 0.86) between the concentration of PFOA and PFNA. However, there was a single work jacket which differed significantly from the others. The authors concluded on this basis that, most likely, the same type of raw material mixture was used by all manufacturers of the impregnating agents.

A similar correlation was not found in this study. With a correlation coefficient (R2) of 0.06, it was demonstrated that the concentrations of PFOA and PFNA are not correlated. A better correlation was found between PFOA and the long-chain PFTeDA (R2 = 0.65). There was an even better correlation between the presence of several of the long-chain perfluoroalkyls: PFTrDA and PFTeDA (R2 = 0.94) and PFDA and PFTeDA (R2 = 0.87). As correlations could only be demonstrated between some of the substances, the conclusion of Knepper *et al.* (2014) may well be an over-interpretation. The results rather indicate that the starting materials are produced by production processes that result in somehow uniform compositions of substances, but the impregnation agents are not necessarily being produced from the same raw materials.

PFOS and other perfluoroalkane sulfonic acids (PFSAs)
The concentrations of PFSAs were relatively low, and the total concentration in all samples was lower than 0.22 µg/m². The concentration of PFOS in all samples was below 0.21 µg/m². The highest value, measured in an infant sleeping bag, was 0.001 mg/kg. The values are far below the limit value as a trace contaminant which is set at 10 mg/kg or 1 µg/m² in the POP Regulation (Regulation (EC) no. 850/2004).

Other PFASs
The analysis programme included analyses of fluorotelomer (FTCAs), fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs). These are not included in most of the previous studies. The total content of these substances accounted for 0.3 % to 5.5 % of the total content of PFASs. Among these substances, 8:2 FTAC occurred in the highest concentrations in all samples, varying from 0.17 to 4.81 µg/m².

The concentrations of perfluoroalkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs) were relatively low in all samples and accounted for less than 0.1 % together in all samples. The total concentrations of fluorotelomer sulfonates (FTSAs) were also relatively low in all samples except in one sample (one infant sleeping bag), where the concentration was 3.67 µg/m². The composition of PFASs in this sample did not differ, apart from this, from the composition of the other samples.

TABLE 8
CONCENTRATION OF PFASs IN TEXTILES FOR CHILDREN (AVERAGE OF TWO ANALYSES)

	Concentration, µg/m² textile (average of two analyses)														
No	**F1**	**F3**	**F4**	**F5**	**F6**	**F7**	**F9**	**F10**	**F11**	**F15**	**F17**	**F18**	**F20**	**F21**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Gloves**	**Infant sleeping bag**	**Infant sleeping bag**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Snowsuit**	**Rain suit**	**Rain suit**	**Mittens**	**Infant sleeping bag**	**Mittens**
Category	PFAS tech-nology	Water and dirt repel-lent	PFAS tech-nology	Water and dirt repel-lent	Water and dirt repel-lent	Teflon®	Water and dirt repel-lent	Teflon®	PFAS tech-nology	Water and dirt repel-lent	PFAS tech-nology	PFAS tech-nology	Teflon®	Water and dirt repel-lent	Water and dirt repel-lent
Total-F µg/m² (average)	60,000	128,000	58,000	21,000	42,000	365,000	32,000	1,200	70	360	100	300	880	165	445
Perfluoroalkane sulfonic acids (PFSAs)															
PFBS	0.01	0.00	<dg	0.00	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFPeS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFHxS	0.00	<dg	0.02	0.01	<dg	<dg	<dg	<dg	<dg	<dg	0.00	<dg	0.00	0.01	0.00
PFHpS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	0.15	<dg	<dg	<dg	<dg	<dg
PFOS	0.06	0.01	0.03	0.21	0.00	<dg	0.05	<dg	<dg	0.06	<dg	0.00	0.01	0.00	0.02
PFDS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	0.01	<dg	<dg	<dg	<dg
Sum	0.07	0.01	0.05	0.22	0.00	<dg	0.05	<dg	<dg	0.21	0.01	0.00	0.01	0.01	0.02
Perfluoralkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)															
FOSA	<dg	<dg	<dg	<dg	<dg	<dg	0.01	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
MeFOSA	0.01	0.01	0.01	0.02	0.01	<dg	<dg	<dg	<dg	0.01	<dg	<dg	<dg	<dg	<dg
EtFOSA	0.01	0.01	0.00	0.01	0.00	<dg	<dg	<dg	<dg	0.05	<dg	<dg	0.04	<dg	<dg
MeFOSE	0.02	0.04	<dg	<dg	0.00	<dg	<dg	<dg	<dg	<dg	<dg	<dg	0.05	<dg	<dg
EtFOSE	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	0.04	<dg	<dg
Sum	**0.04**	**0.05**	**0.01**	**0.02**	**0.01**	<dg	**0.00**	<dg	<dg	**0.05**	<dg	<dg	**0.13**	<dg	<dg
Perfluoroalkyl carboxylic acids (PFCAs)															
PFPA	0.08	0.28	0.01	0.07	0.02	0.00	<dg	0.02	<dg	0.12	0.02	0.01	0.01	0.01	0.04
PFHxA	0.99	3.26	0.20	0.25	0.02	0.14	0.05	0.22	0.00	0.73	0.01	0.14	0.07	0.00	0.54
PFHpA	0.99	2.22	0.02	0.16	0.02	0.05	0.03	0.04	0.00	0.37	0.01	0.04	0.02	0.01	0.20

	Concentration, µg/m² textile (average of two analyses)														
No	**F1**	**F3**	**F4**	**F5**	**F6**	**F7**	**F9**	**F10**	**F11**	**F15**	**F17**	**F18**	**F20**	**F21**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Gloves**	**Infant sleeping bag**	**Infant sleeping bag**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Snowsuit**	**Rain suit**	**Rain suit**	**Mittens**	**Infant sleeping bag**	**Mittens**
PFOA	31.18	13.15	0.11	0.66	0.12	1.15	0.60	0.64	0.05	1.74	0.04	0.35	0.09	0.03	3.03
PFNA	0.55	2.97	0.02	0.13	0.06	0.05	3.41	0.04	0.09	0.63	0.00	0.03	0.04	0.03	0.21
PFDA	2.72	7.44	0.03	0.33	0.06	0.66	0.24	0.24	0.02	1.78	0.01	0.18	0.04	0.02	1.53
PFUnDA	0.20	0.98	0.00	0.06	0.01	0.04	1.05	0.02	0.03	0.17	0.00	0.01	0.01	0.01	0.08
PFDoDA	1.25	3.94	0.00	0.13	0.01	0.46	0.02	0.13	0.00	0.46	0.00	0.07	0.00	0.00	0.57
PFTrDA	0.69	1.34	<dg	0.19	0.00	<dg	0.02	0.03	0.01	0.07	0.00	0.01	0.00	0.00	0.07
PFTeDA	6.67	9.31	0.02	1.04	0.57	0.12	0.07	0.17	0.02	2.41	0.01	0.13	0.01	0.02	1.36
Sum	**45.30**	**44.88**	**0.41**	**3.00**	**0.90**	**2.67**	**5.48**	**1.54**	**0.23**	**8.48**	**0.10**	**0.97**	**0.28**	**0.13**	**7.62**
Fluorotelomer carboxylic acids (FTCAs)															
6:2 FTCA	<dg	0.05	<dg	<dg	<dg	0.04	<dg	<dg	<dg	<dg	<dg	<dg	0.02	<dg	0.02
8:2 FTCA	0.22	0.15	<dg	0.05	0.09	1.18	0.32	0.07	0.11	0.19	0.03	0.04	0.09	0.05	0.09
10:2 FTCA	0.28	0.20	<dg	<dg	0.05	1.04	0.14	0.04	0.03	0.04	<dg	0.02	0.03	0.02	0.05
Sum	**0.50**	**0.39**	<dg	**0.05**	**0.14**	**2.26**	**0.46**	**0.11**	**0.14**	**0.23**	**0.03**	**0.05**	**0.14**	**0.08**	**0.17**
Fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs)															
8:2 FTMAC	<dg	<dg	<dg	<dg	<dg	0.37	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
10:2 FTMAC	<dg	<dg	<dg	<dg	<dg	0.29	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
4:2 FTAC	<dg	<dg	<dg	<dg	<dg	0.65	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTAC	0.08	<dg	<dg	<dg	<dg	0.53	<dg	0.12	<dg	<dg	<dg	<dg	<dg	<dg	<dg
8:2 FTAC	3.80	0.85	0.06	0.17	0.18	0.78	0.49	4.81	0.77	0.62	0.48	0.24	2.72	0.72	1.10
10:2 FTAC	1.22	0.26	0.01	0.07	0.03	0.27	0.08	1.54	0.14	0.13	0.16	0.13	0.72	0.20	0.40
Sum	**5.06**	**1.11**	**0.06**	**0.24**	**0.21**	**1.97**	**0.57**	**6.46**	**0.91**	**0.75**	**0.64**	**0.37**	**3.44**	**0.92**	**1.49**
Fluorotelomer alcohols (FTOHs)															
4:2 FTOH	0.24	0.07	0.15	0.33	<dg	0.33	<dg	0.30	0.07	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTOH	0.41	0.61	7.53	6.19	3.62	0.83	0.25	0.48	0.40	0.02	0.44	0.44	83.05	2.68	1.77
8:2 FTOH	37.18	32.85	11.47	6.52	20.60	169.99	227.20	97.15	81.24	44.82	20.39	30.79	264.99	109.44	80.44

	Concentration, µg/m² textile (average of two analyses)														
No	**F1**	**F3**	**F4**	**F5**	**F6**	**F7**	**F9**	**F10**	**F11**	**F15**	**F17**	**F18**	**F20**	**F21**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Gloves**	**Infant sleeping bag**	**Infant sleeping bag**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Snowsuit**	**Rain suit**	**Rain suit**	**Mittens**	**Infant sleeping bag**	**Mittens**
10:2 FTOH	12.02	5.30	3.25	1.83	5.04	32.57	45.78	21.86	17.06	14.26	5.55	7.03	54.54	22.60	17.05
Sum	**49.73**	**38.80**	**22.40**	**14.87**	**29.27**	**203.55**	**273.10**	**119.80**	**98.57**	**59.09**	**26.39**	**38.26**	**402.58**	**134.73**	**99.25**
Fluorotelomer sulfonates (FTSAs)															
4:2 FTSA	<dg	<dg	<dg	<dg	<dg	0.08	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTSA	<dg	<dg	<dg	<dg	<dg	0.42	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
8:2 FTSA	<dg	<dg	<dg	<dg	<dg	3.17	0.07	0.02	0.03	0.02	0.01	0.02	0.02	0.02	0.03
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**3.67**	**0.07**	**0.02**	**0.03**	**0.02**	**0.01**	**0.02**	**0.02**	**0.02**	**0.03**
Sum PFASs	**100.69**	**85.24**	**22.93**	**18.40**	**30.52**	**214.11**	**279.74**	**127.93**	**99.88**	**68.83**	**27.18**	**39.67**	**406.59**	**135.88**	**108.58**

<dg. Below the detection limit. See separate list with detection limits for the individual substances in Appendix 6.

Comparison with previous studies

German study from 2014 - The total concentrations of PFASs found in this study is within the same range as the concentrations measured in a recently published German study of PFASs in 15 outdoor jackets (Knepper *et al.*, 2014). The examined outdoor jackets in the German study were purchased during the period of August 2011 to March 2012. *N*-hexane was used for the extraction of PFASs from the textiles. In the study the total content of PFASs varied in the 14 samples, in which PFASs were found in the range of 5 to 719 µg/m², as compared to the total content of PFASs found in the present study, ranging from 18 to 407 µg/m². In the study of Knepper *et al.* (2014), two samples differed significantly from the other samples with high total PFASs concentrations of 458 and 719 µg/m², respectively, while the total concentrations in the other samples were less than 76 µg/m² on average. One of the samples with a high concentration, a work jacket (719 µg/m² of FTOHs) also differed, as the vast majority of the PFASs were made up of PFCAs, while FTOHs represented only a small percentage of the total concentration. Samples with such a composition are not found in the present study. The results could indicate that this jacket was treated with impregnation agents significantly different than the agents used for the other jackets. Three of the jackets are indicated to have PTFE membranes, but the composition of PFASs in these jackets were not significantly different from the others, possibly because they were also treated with a PFAS-based coating (this is not specified). There were no analyses carried out on the membranes alone.

FTOHs accounted for the vast majority of the total content of PFASs in 13 of 15 outdoor jackets analysed by Knepper et al. (2014). In 12 of the samples, 8:2 FTOH was the dominant FTOH followed by 10:2 FTOH. The concentration of 6:2 FTOH was negligible in these 12 samples, while one sample differed as 6:2 FTOH was the dominant PFAS identified. In this sample, the concentration of PFHxA was at the same level as the concentration of PFOA, while in most of the other samples it was much less. In most samples the dominant PFCAs were PFOA and PFDA. The same pattern was found in the present study. Fluorotelomer carboxylic acids (FTCAs), fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs) were not included in the German study, so it is not possible to compare the values of these substances.

Greenpeace study from 2012 - A Greenpeace study on outdoor wear purchased the summer of 2012 in Germany, Austria and Switzerland found FTOHs to a lesser extent in the products. This finding can be explained by the fact that the method used had a very high detection limit for FTOHs (Greenpeace, 2012). The FTOH concentration was above the detection limit in only 7 out of 14 samples. In these 7 samples, 6:2 FTOH occurred in the highest concentrations in five of the samples and 8:2 was dominant in two samples. PFOA was the dominant PFCAs in 12 of the samples, while PFHxA was found in the highest concentrations in 2 samples.

Another German study from 2014 - In another new German study of 16 outdoor jackets and gloves (Dreyer *et al.*, 2014) a somewhat different pattern was found. It is not stated when the clothes are purchased, but the analyses were conducted in October 2013 and the jackets were probably purchased immediately beforehand. For the extraction of acids methanol was used, while for the extraction of volatile PFASs MTBE (methyl tert-butyl ether) was used. As in the other studies, PFCAs and FTOHs were the dominant groups of PFASs, but C_{4-6} chemistry was more prominent, while C_8 chemistry represented a minor part of the picture. The total concentrations of PFCAs were on the same level as found in the present study, but in only 8 out of 15 samples (above the detection limit) was PFOA the dominant PFCA. In seven of the samples, the concentration of PFOA exceeded 1 µg/m². In four samples PFHxA was the dominant PFCA, and in three samples, PFBA was found in the highest concentration. This pattern was even more pronounced for FTOHs, where 6:2 FTOH appeared in concentrations higher than 8:2 FTOH in 10 of the 15 samples, while 8:2 FTOH appeared in the highest concentrations in 5 samples. The total concentrations of FTOHs were generally significantly higher than the levels found in this study and those found by Knepper *et al.* (2014), but similar high levels were previously found in Norwegian studies (SFT, 2006). Where the highest

concentration of FTOHs found in the present study are 403 µg/m², Dreyer *et al.* (2014) found a total concentration of FTOHs exceeding 500 µg/m² in 6 of 15 samples, with the highest level of approximately 1.700 µg/m².

C_8 chemistry versus C_{4-6} chemistry

The results of the present study do not indicate any shift from C_8 chemistry toward short-chain chemistry. PFOA was the dominant perfluoralkyl carboxylic acid (PFCAs), 8:2 FTCA the dominant fluorotelomer acid (FTCAs), 8:2 FTAC the dominant fluorotelomer acrylate (FTACs), 8:2 the dominant fluorotelomer alcohol (FTOHs) and 8:2 FTSA the dominant fluorotelomer sulfonic acid. In total, C_8 substances accounted for 41 to 82 % of the total content of analysed PFASs. The remainder consisted mainly of 6:2 FTOH of 10:2 FTOH. The aforementioned German study of outdoor jackets for adults (Knepper *et al.*, 2014) showed, in agreement with the present study, that C_8 chemistry was dominant, and the authors conclude that there is no evidence that C_8 chemistry is in the process of being replaced. The Greenpeace survey of outdoor wear (both children and adults) purchased in the summer of 2012 showed a more complex pattern, with PFOA as the dominant PFCAs, while 6:2 FTOH was the dominant FTOH (Greenpeace, 2012). The study, however, had problems with the analyses of FTOHs. In contrast to the results of this study and the study of Knepper *et al.* (2014), another German study (Dreyer *et al.*, 2014), found the C_{4-6} chemistry to be dominant in about half of the examined pieces of clothing (more than half as concerns the FTOHs; it is not specified whether the clothes were for children or adults). The samples in that study were apparently purchased in October 2013, about one and a half years after the samples analysed by Knepper *et al.* (2014). The differences may be due to a change that occurred during this period. However, this does not explain the differences in relation to the results of the present study.

The results could indicate that some of upmarket brands in outdoor wear for adults have switched from C_8 chemistry C_{4-6}-chemistry, but that this movement apparently has not occurred for children's clothes. On the other hand, there has been a move away from fluorine chemistry by those producers of children's clothing most aware of the problem.

5.2.3 Migration of PFASs to saliva

Based on the analyses of PFASs, eight of the 15 products were selected for migration and wash tests (results of the washing tests are presented in section 5.2.4). As all products had a relatively high content of FTOH compared to the other PFASs, it was the content of the other PFASs which was decisive for the selection. The products selected were consequently the products with the highest concentrations of other PFASs, thereby increasing the possibility of the concentrations in the migration liquid and laundry water being above the detection limit.

The measured rates of migration from textiles to artificial saliva over a period of three hours are shown in Table 9. The composition of PFASs in the artificial saliva was significantly different from the composition in the materials. The composition in the artificial saliva was dominated by the acids: PFCAs, PTCAs and FTSAs. In a few samples, FTOHs accounted for a significant portion, but in general, the content of FTOHs in the artificial saliva was low compared with the contents of FTOHs in the materials. As an average for all PFASs, the volume in artificial saliva corresponded to about 1.0 % (standard error: 0.4 %) of the content of the materials. For PFCAs, the release rates were significantly higher. The amount that migrated to the artificial saliva corresponded to 6.1 % (standard error: 1.9 %) on average of the content of the materials. For PFOA, the proportion was even higher, although the difference is not significant, as the amount of artificial saliva corresponded to 8.2 % (standard error: 2.0 %) of the content of the materials.

On average, PFCAs accounted for 55 % of the total content of PFASs in the artificial saliva. Of this PFOA was the predominant substance, and this substance alone constituted about 30 % of the total content.

The highest release rate found was 2.87 µg/m² fabric. Of this, PFOA accounted for 1.46 µg/m², the other PFCAs accounted for 1.22 µg/m², and 8:2 FTOH for 0.15 µg/m², while all other PFASs accounted for 0.05 µg/m².

TABLE 9
RELEASE OF PFASs TO ARTIFICIAL SALIVA OVER A PERIOD OF THREE HOURS

	Migrated quantity, µg/m² textile *							
No	**F1**	**F3**	**F7**	**F9**	**F10**	**F15**	**F20**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Mittens**	**Mittens**
Perfluoroalkane sulfonic acids (PFSAs)								
PFBS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFPeS	<dg	<dg	<dg	<dg	0,005	<dg	<dg	<dg
PFHxS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFHpS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFOS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFDS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**0,00**	**<dg**	**<dg**	**<dg**
Perfluoralkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)								
FOSA	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
MeFOSA	<dg	<dg	<dg	<dg	0.00	<dg	<dg	<dg
EtFOSA	<dg	<dg	<dg	<dg	0.02	<dg	<dg	<dg
MeFOSE	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
EtFOSE	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**0.02**	**<dg**	**<dg**	**<dg**
Perfluoroalkyl carboxylic acids (PFCAs)								
PFPA	0.02	0.02	0.02	<dg	<dg	0.02	<dg	<dg
PFHxA	0.10	0.43	0.07	0.01	0.03	0.10	0.01	0.02
PFHpA	0.12	0.30	0.06	0.01	0.01	0.06	0.00	0.01
PFOA	1.47	1.46	0.22	0.04	0.07	0.14	0.00	0.05
PFNA	0.01	0.19	0.03	0.07	0.00	0.02	0.00	0.00
PFDA	0.02	0.27	0.06	0.00	0.00	0.03	0.00	0.01
PFUnDA	<dg	0.01	0.01	0.01	<dg	<dg	<dg	<dg
PFDoDA	<dg	<dg	0.01	<dg	<dg	<dg	<dg	<dg
PFTrDA	0.00	<dg	0.00	<dg	<dg	<dg	<dg	<dg
PFTeDA	0.00	<dg	0.01	<dg	<dg	<dg	<dg	<dg
Sum	**1.75**	**2.68**	**0.49**	**0.14**	**0.12**	**0.37**	**0.01**	**0.08**
Fluorotelomer carboxylic acids (FTCAs)								
6:2 FTCA	0.00	0.01	0.00	0.00	0.00	<dg	0.01	0.01
8:2 FTCA	0.02	0.01	0.23	0.04	0.01	0.01	0.01	0.02
10:2 FTCA	<dg	0.00	0.08	0.00	<dg	<dg	<dg	0.00
Sum	**0.02**	**0.02**	**0.31**	**0.04**	**0.01**	**0.01**	**0.02**	**0.02**

	Migrated quantity, µg/m² textile *							
No	**F1**	**F3**	**F7**	**F9**	**F10**	**F15**	**F20**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Mittens**	**Mittens**
Fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs)								
8:2 FTMAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
10:2 FTMAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
4:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
8:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
10:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**
Fluorotelomer alcohols (FTOHs)								
4:2 FTOH	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTOH	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
8:2 FTOH	0.10	0.15	0.11	0.13	0.02	<dg	0.08	0.26
10:2 FTOH	0.00	0.02	0.00	<dg	0.00	0.01	0.02	0.02
Sum	**0.10**	**0.17**	**0.11**	**0.13**	**0.03**	**0.01**	**0.10**	**0.28**
Fluortelomersulfonater (FTSAs)								
4:2 FTSA	<dg	<dg	0.05	<dg	<dg	<dg	<dg	<dg
6:2 FTSA	0.00	0.00	0.36	0.00	0.00	0.00	0.00	0.00
8:2 FTSA	<dg	<dg	1.29	0.01	<dg	0.01	0.00	0.01
Sum	**0.00**	**0.00**	**1.70**	**0.01**	**0.00**	**0.01**	**0.00**	**0.01**
Sum PFASs	**1.88**	**2.87**	**2.60**	**0.32**	**0.18**	**0.40**	**0.13**	**0.39**
Content of PFASs in product	100.69	85.24	214.11	279.74	127.93	68.83	406.59	100.69
% of content of PFASs in product	1.9 %	3.4 %	1.2 %	0.1 %	0.1 %	0.6 %	0.03 %	1.9 %
Content of PFCAs in product	45.30	44.88	2.67	5.48	1.54	8.48	0.28	45.30
% of content of PFCAs in product	4 %	6 %	18 %	2 %	8 %	4 %	5 %	4 %

* All figures rounded to two decimals.

Figure 7 shows the composition of PFCAs in the artificial saliva from the three material samples (F1, F3 and F15), which had the highest content of PFCAs (note that migration from F7 is higher than migration from F15). A comparison with the composition of PFCAs in textile samples (shown in Figure 6) demonstrates a shift in the artificial saliva toward the shorter-chain PFCAs. The four PFCAs with the longest chains were all below or close to the detection limit in artificial saliva, while the analyses of the content (Figure 6) showed that the concentration of these substances in the materials generally were higher than the concentration of short-chain PFCAs (shorter than PFOA).

Comparison with previous studies

There are no examples in the literature on analysis of migration of PFASs to saliva from textiles. It is therefore not possible to discuss the results in relation to previous studies.

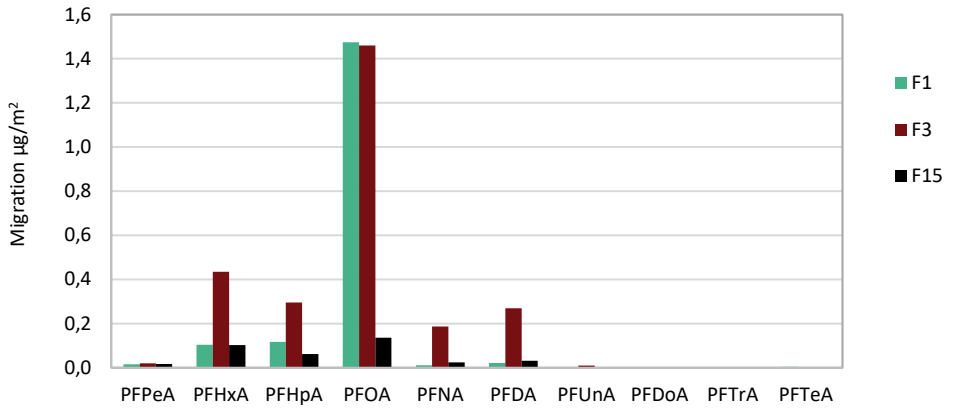


FIGURE 7
COMPOSITION OF PERFLUOROALKYL CARBOXYLIC ACIDS (PFCAs) IN ARTIFICIAL SALIVA FROM THE THREE SAMPLES WITH THE HIGHEST CONTENT OF PFCAs.

5.2.4 PFASs in laundry water

The 8 products that were selected for the migration test were also tested for release of PFASs to the laundry water by washing of the textiles. The results are shown in Table 10. The results are very similar to the results of migration to the artificial saliva. The composition of PFASs in the laundry water was significantly different than the composition of the PFASs in the materials, and as seen for the migration to the artificial saliva, the composition was dominated by the acids. In the laundry water FTOHs accounted for a slightly higher proportion compared to the artificial saliva; however, content of FTOHs in the laundry water was still relatively low in view of its high concentrations in the materials. As an average for all PFASs, the amount released to laundry water corresponded to approximately 1.0 % (standard error: 0.9 %) of the content of the materials. For PFCAs, the release rate was significantly higher. The amount that was released to the laundry water on average corresponded to 12.2 % (standard error: 5.0 %) of the content of the materials. For PFOA, the proportion was even higher, although the difference is not significant; the amount in the laundry water corresponded to 13.7 % (standard error: 5.5 %) of the content of the materials. As is clear from the values of the standard error (which expresses the uncertainty of the mean value), there was significantly greater variation in the detected discharge rates in the washing tests as compared with the migration tests.

Overall, there are no significant differences between the release to saliva and to laundry water, and there is nothing to suggest that the presence of detergent has affected the release pattern significantly.

TABLE 10
RELEASE OF PFASs TO LAUNDRY WATER

	Amount released to laundry water, µg/m² textiles *							
No	**F1**	**F3**	**F7**	**F9**	**F10**	**F15**	**F20**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Mittens**	**Mittens**
Perfluoroalkane sulfonic acids (PFSAs)								
PFBS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFPeS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFHxS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFHpS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFOS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
PFDS	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**	**<dg**
Perfluoralkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)								
FOSA	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
MeFOSA	<dg	<dg	<dg	<dg	0.00	<dg	<dg	<dg
EtFOSA	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
MeFOSE	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
EtFOSE	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
Sum	**<dg**	**<dg**	**<dg**	**<dg**	**0.00**	**<dg**	**<dg**	**<dg**
Perfluoroalkyl carboxylic acids (PFCAs)								
PFPA	0.03	0.18	0.04	0.02	0.00	0.01	0.01	<dg
PFHxA	0.07	0.31	0.07	0.01	0.04	0.10	0.00	0.03
PFHpA	0.17	0.34	0.08	0.01	0.08	0.12	0.00	0.02
PFOA	2.08	0.84	0.46	0.03	0.24	0.15	0.00	0.14
PFNA	0.05	0.21	0.08	0.06	0.01	0.05	0.00	0.01
PFDA	0.12	0.19	0.24	0.00	0.06	0.06	<dg	0.04
PFUnDA	0.01	0.03	0.02	0.01	0.00	0.01	<dg	0.00
PFDoDA	0.02	0.03	0.07	0.00	0.01	0.01	<dg	0.01
PFTrDA	<dg	<dg	0.01	<dg	<dg	<dg	<dg	<dg
PFTeDA	0.01	0.01	0.02	0.00	0.00	0.01	<dg	0.00
Sum	**2.56**	**2.13**	**1.09**	**0.13**	**0.44**	**0.52**	**0.02**	**0.25**
Fluorotelomer carboxylic acids (FTCAs)								
6:2 FTCA	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
8:2 FTCA	0.03	0.02	0.42	0.02	0.01	0.01	0.00	0.01
10:2 FTCA	<dg	<dg	0.44	<dg	<dg	<dg	<dg	<dg
Sum	**0.03**	**0.02**	**0.87**	**0.02**	**0.01**	**0.01**	**0.00**	**0.02**
Fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs)								
8:2 FTMAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
10:2 FTMAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
4:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg

	Amount released to laundry water, µg/m² textiles *							
No	**F1**	**F3**	**F7**	**F9**	**F10**	**F15**	**F20**	**F22**
Product name	**Snowsuit**	**Softshell-suit**	**Infant sleeping bag**	**Rain jacket**	**Rain suit**	**Snowsuit**	**Mittens**	**Mittens**
6:2 FTAC	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
8:2 FTAC	0.00	0.01	0.00	<dg	0.00	<dg	0.00	0.00
10:2 FTAC	0.00	0.00	0.00	<dg	0.00	<dg	<dg	<dg
Sum	**0.00**	**0.01**	**0.00**	**<dg**	**0.01**	**<dg**	**0.00**	**0.00**
Fluorotelomer alcohols (FTOHs)								
4:2 FTOH	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg
6:2 FTOH	0.01	0.03	<dg	<dg	0.00	<dg	0.01	0.03
8:2 FTOH	0.01	0.71	1.01	0.33	0.04	0.27	0.06	0.21
10:2 FTOH	0.10	0.13	0.10	0.06	0.01	0.11	0.02	0.05
Sum	**0.12**	**0.87**	**1.11**	**0.39**	**0.06**	**0.38**	**0.09**	**0.29**
Fluortelomersulfonater (FTSAs)								
4:2 FTSA	<dg	<dg	0.03	<dg	<dg	<dg	<dg	<dg
6:2 FTSA	<dg	<dg	0.39	0.00	<dg	<dg	<dg	0.00
8:2 FTSA	0.01	0.00	12.21	0.02	0.01	0.01	0.00	0.01
Sum	**0.01**	**0.00**	**12.64**	**0.02**	**0.01**	**0.01**	**0.00**	**0.01**
Sum PFASs	**2.73**	**3.03**	**15.71**	**0.56**	**0.52**	**0.91**	**0.12**	**0.56**
Content of PFASs in product	100.69	85.24	214.11	279.74	127.93	68.83	406.59	100.69
% of content of PFASs in product	2.7 %	3.6 %	7.3 %	0.2 %	0.4 %	1.3 %	0.0 %	0.5 %
Content of PFCAs in product	45.30	44.88	2.67	5.48	1.54	8.48	0.28	45.30
% of content of PFCAs in product	6 %	5 %	41 %	2 %	28 %	6 %	7 %	3 %

* All figures rounded to two decimals.

Figure 8 shows the composition of PFCAs in the laundry water from the three material samples (F1, F3 and F15) with the highest content of PFCAs. A comparison with the composition of PFCAs in the material samples, as shown in Figure 6, shows a shift in the laundry water towards the shorter-chain PFCAs. This is the same pattern seen for the composition in the artificial saliva in Figure 7. The four PFCAs with the longest chains were all below or close to detection limit in the laundry water and in artificial saliva, while the content analysis (Figure 6) showed concentrations of these substances in materials as being generally higher than that of the concentration of short-chain PFCAs (shorter one PFOA).

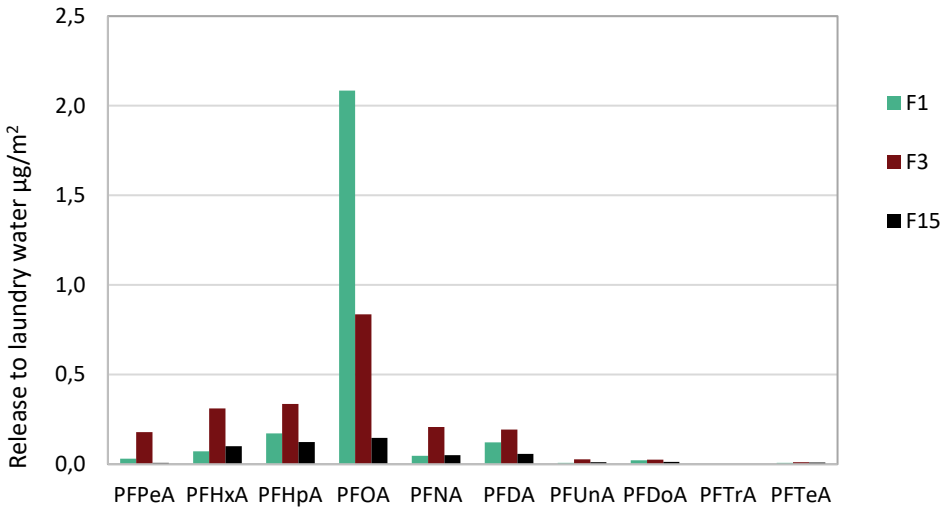


FIGURE 8
COMPOSITION OF PERFLUOROALKYL CARBOXYLIC ACIDS (PFCAs) IN LAUNDRY WATER FROM THE THREE SAMPLES WITH THE HIGHEST CONTENT OF PFCAs.

Comparison with previous studies
The results of this study are very different from previous studies, as significantly higher release rates were found in previous studies.

German study from 2014 - Knepper *et al.* (2014) investigated the release of 11 PFCAs to the laundry water from four jackets in a washing machine. The study did not include measurements of FTOHs. The jackets were washed without the use of detergent, because the content of surfactants in the detergent interfered with the analyses. In the test, four jackets were washed simultaneously. Two of the jackets contained Gore-tex® membranes. It is reported that some tests were made with the use of detergents, but this resulted in recovery rates of less than 5 % of the added PFASs. The following PFCAs were not present in concentrations above the detection limit: PFBA, PFPeA, PFHpA PFDoDA and PFTrDa. The results for the other PFCAs are shown in Table 11.

In this German study, the total amount released from two washes corresponded to more than twice the amount of PFHxA and PFOA extracted with solvent. For PFNA and PFDA, the total release was nearly the same as extracted with solvent, and for PFUnDA and PFTrDA, somewhat smaller. There was an inverse correlation between the release rates (expressed as the percentage of the content in the textiles) and the length of the carbon chains. The same result is found in the present study. The high rates found in the German study might be due to mechanical wear, but since few measurements were performed, the results should be replicated in other studies before certain conclusions can be drawn.

Repeated washing in the German study resulted in release rates of nearly half that of the first wash. The results may indicate that during the entire useful life of the clothing, the quantities released to laundry water may be many times greater than the extractable content of the clothes.

TABLE 11
RELEASE OF PERFLUOROALKYL CARBOXYLIC ACIDS (PFCAs) TO LAUNDRY WATER (KNEPPER *ET AL.*, 2014). RESULTS OF SIMULTANEOUS WASHING OF FOUR JACKETS OF WHICH TWO INCLUDED A GORE-TEX® MEMBRANE

	Release in percent of extractable content			
	Experiment 1A *	Experiment 1B *	Experiment 2A **	Total of 1A and 2A
PFHxA	155	151	60	216
PFOA	197	178	39	235
PFNA	96	67	35	131
PFDA	99	85	21	119
PFUnDA	13	11	22	35
PFTrDA	0	0	77	77

* 1st wash for unspecified period of time in a washing machine at 30°C without the use of detergent.
* 2nd wash - repeat of 1. wash.

German study from 2013 - In another German study, Bringewatt *et al.* (2013) found that total PFASs in the laundry water from washing clothes with PFAS-based impregnating agents in a washing machine ranged from <0003-10 g/L. It is not specified whether detergents were used and not indicated whether the clothes have Gore-tex® membranes. The results are presented in figures which are difficult to interpret. The concentrations of the PFASs in the materials are also not specified. The highest value (PFHpA) occurred in a single sample resulting in a discharge per wash at 188 µg/kg textile. The highest estimated release of PFOA was about 100 µg/kg textile, and from several of the samples the release of 8:2 FTOH was in the range of 20-80 µg/kg textile. The concentrations of PFAAs (PFCAs and PFSAs) decreased with the number of washes, but this was not the case with 8:2 FTOH, which apparently could be formed by degradation of the bonds between the side chains and the backbone. This supposition was confirmed by the fact that the emissions of FTOHs to air were similar before and after washing. The highest values found in this investigation for PFCAs and FTOHs, respectively, were 12.2 µg PFOA per kg textile emitted from sample F1 and 8.4 µg 8:2 FTOH per kg textile emitted from sample F7 (note that the unit here is g/ kg textile, whereas the tables show µg/m²).

Discussion related to the results of the present study - Another explanation may be difficulties with the analyses. Knepper *et al.* (2014) indicated that the tests show that the recovery rates was less than 5 % when using detergents. In the present study, however, recovery rates (Section 5.1.4) of about 100 % were found in the laundry water with detergent; hence, low recovery rates cannot explain the differences.

One difference between the experimental conditions is that half of the jackets in the German study (Knepper *et al.*, 2014), contained Gore-Tex® membranes, while none of the samples in this study contained Gore-Tex®. Since all the jackets in the German study were washed, it is not clear whether there was a particularly high release of PFASs when washing jackets containing Gore-Tex®. To what extent this may explain the differences can only be determined by conducting experiments whereby fabrics are washed with and without Gore-Tex® membranes separately.

5.3 Overall discussion of the analysis results in relation to the environmental and health assessment

Human exposure via saliva

The results of the migration test showed that acids especially were released to saliva. The highest measured rate for the sum of all PFASs was 2.87 µg/m² fabric. Of this amount PFOA accounted for 1.46 µg/m², the other PFCAs accounted for 1.22 µg/m², 8:2 FTOH 0.15 µg/m², while all other PFASs accounted for 0.05 µg/m². Overall, for all samples, PFOA was found in the highest concentrations in the artificial saliva; the health assessment will therefore be based on the exposure to this substance.

For a "worst case" estimate it will initially be assumed that the effects of the other PFASs are similar to the effects of PFOA in order to count for the total releases of all PFASs.

Environmental exposure via laundry water

As in the case of migration to artificial saliva, it was largely acids which were released to laundry water. The results demonstrate widely varying release rates between this and previous German studies, but the available information indicates that the release of PFASs to the laundry water during the products' entire useful life may be several times larger than the extractable content. PFOA is the dominant PFAS in the laundry water, and the environmental assessment will therefore be based on emissions of this substance. For a "worst case" estimate it will initially be assumed that the effects of the other PFASs are similar to the effects of PFOA, in order to count for the total emissions of all PFASs.

6. Health assessment

6.1 Introduction

The health assessment includes an initial introduction to the substances and a review of the health effects of the various PFASs. The review includes both a description of the present knowledge about the effects of PFASs from population surveys, and a description of the results of experimental studies. There is limited knowledge about many of the substances; the review will therefore also serve as a basis for generalisations made in the health risk assessment, which is based on the derived no-effect level (DNEL) for one of the substances, PFOA.

After the review of the effects, knowledge on the exposure of children to PFASs is briefly reviewed. The review provides a basis for later comparison between the exposure of children to PFASs released from children's clothing and exposure to PFASs from other sources.

In the health risk assessment, "reasonable worst case scenarios" for exposure of children are established and the quantities children are exposed to and the potential daily intake via the different routes of exposure are calculated. The exposure assessment is performed for relevant exposure routes based on the principles of the REACH guidance, "Guidance on Information Requirements and Chemical Safety Assessment. Chapter R.15 - Consumer exposure estimation" (ECHA, 2012a).

Based on the review, a derived no-effect level (Derived No-Effect Level, DNEL) is established. In the absence of data for most of the substances, and based on the fact that PFOA and its precursor, 8:2 FTOHs, constitute a large part of PFASs in the textiles, a DNEL for PFOA based on human studies and blood data is used.

The risk assessment is carried out by comparing the calculated exposure (internal dose) with this DNEL value and calculating a Risk Characterisation Ratio (RCR).

6.2 Grouping of PFASs

The substances described are mainly designated by use of abbreviations in the following. A list of abbreviations, full substance names and CAS numbers are shown in Appendix 1.

It is very common in the context of health and environmental assessments to group the polyfluoroalkylated substances (PFASs) into the following groups (Buck *et al.*, 2011):

1. PFSAs: Perfluoroalkane sulfonic acids and their salts, perfluoroalkyl sulfonates. The substances are less volatile and more water-soluble than their precursors. A well-known substance in this group is PFOS.

2. Precursors to PFSAs are often sulfonamides of various kinds (FASAs, phase, etc.), but may also be the starting substance POSF. All of these substances can degrade to PFSAs through several stages over time.

3. PFCAs: Perfluoroalkyl carboxylic acids and their salts, perfluoro carboxylates. These substances are less volatile and more water-soluble than their precursors. A well-known substance in this group is PFOA.

4. Precursors to PFCAs are often fluorotelomer alcohols (FTOHs) of different lengths, but can also be fluorotelomer carboxylates (FTCAs), fluorotelomer sulfonates (FTSAs) and fluorotelomer phosphates (FTP) and starting materials such as fluorotelomer iodides and fluorotelomer alkenes. All these telomers can degrade to PFCAs through several steps over time.

Groups 1 and 3 (PFSAs and PFCAs) are collectively termed perfluoroalkyl acids (PFAAs), while groups 2 and 4 together can be described as precursors. It appears from the results of the analysis programme in Chapter 5 that it is particularly PFCAs and their precursors, the fluorotelomers, that occur in the fabrics.

In the health assessment, differences in the properties of the fluorinated substances are more related to the length of perfluoroalkyl chain (C_nF_{2n+1}- or $F(CF_2)_n$-) than to the functional group. Substances with a perfluoroalkyl chain of seven to eight carbon atoms (C_{7-8}, for example PFHxS, PFOA, PFOS and PFNA) have the lowest surface tension, the biggest negative impact on cell membranes, the largest bioaccumulation, and a lengthy half-life time in the organism. Furthermore, they are the best studied toxicologically, and seems to be more harmful than the shorter-chained substances. If the perfluoroalkyl chain is shorter than six carbon atoms, substances are still persistent, but the bioaccumulation in organisms in nature and in humans is much less. Perfluoroalkyl carboxylic acids (PFCAs) with longer carbon chains are more reminiscent of fatty acids, may influence fat metabolism, and are often more harmful.

In many studies, the measured concentrations of FTOHs or PFCAs are aggregated into an overall figure (which is also indicated in the tables with the results shown in Table 8). The figure can serve to provide an overview and help compare the contents of PFASs in different products, but these totals have no relevance in health and environmental assessments, as the characteristics of the individual substance depends on the length of the perfluoroalkyl chain and substance degradation to perfluoroalkyl acids (PFAAs).

6.3 Health effects

Health effects of PFASs are described in a recent and comprehensive review article by Stahl *et al.* (2011), which, among other sources, has formed the basis for the description in the following sections.

6.3.1 Harmonised classification

Only PFOS and its derivatives have a harmonized classification in accordance with the CLP Regulation. The classification is shown in the table below.

TABLE 12
HARMONISED CLASSIFICATION ACCORDING TO THE CLP REGULATION (EC) NR 1272/2008

Index No.	International chemical identification *1	CAS No.	Classification	
			Hazard Class and Category Code(s)	Hazard statement Code(s)
607-624-00-8	Perfluorooctane sulfonic acid [1]; potassium perfluorooctane sulfonate [2]; diethanolamine perfluorooctane sulfonate [3]; ammonium perfluorooctane sulfonate [4]; lithium perfluorooctane sulfonate [5]	1763-23-1 [1] 2795-39-3 [2] 70225-14-8 [3] 29081-56-9 [4] 29457-72-5 [5]	Carc. 2 Repr. 1B STOT RE 1 Acute Tox. 4 * Acute Tox. 4 * Lact. Aquatic Chronic 2	H351 H360D*** H372** H332 H302 H362 H411

*1 All substances are C_8 PFASs. Reference is made to the CLP Regulation (Regulation (EC) 1272/2008) for further explanation for *, ** and ***, which concern the translation from Directive 67/548/EEC to the CLP-Regulation for the Hazard Classes indicated.

6.3.2 Population studies

As populations are exposed to various mixtures of PFASs, it is difficult to determine the possible health effects of individual PFASs from population studies, but population studies have shown positive correlations between levels of certain perfluoroalkyl acids (PFAAs) in the blood and various disease outcomes and disease-related parameters. These relationships are shown particularly for PFOS and PFOA, which usually occur in the highest concentrations in the blood.

Effects on lipid metabolism, and more cholesterol in the blood

Exposure to most PFASs affects fat metabolism in experimental animals. In population studies similar effects have been demonstrated in humans.

According to a review paper of Steenland *et al.* (2010a), a positive association between PFOA and cholesterol and other lipids in blood was demonstrated in several occupational studies and in studies of particular exposed populations.

Similarly, in another study, a clear relationship between PFOS and increased total cholesterol and low-density lipoproteins (LDL) and high-density lipoproteins (HDL) (Gallo *et al.*, 2012) was demonstrated. This study also found a positive association between PFOS and PFOA in blood and blood levels of alanine transferase (ALT), which is a sign of incipient liver damage.

In a study based on data from the large 2003-2004 National Health and Nutrition Examination Survey (NHANES) in the United States, a positive relationship was also found between total cholesterol and the concentration of PFOS (mean 25 ng/mL), PFOA (mean 4.6 ng/mL) and in particular PFNA (mean 1.3 ng/mL) in the blood serum (Nelson *et al.*, 2010).

A Canadian study found a significant association between a very high blood concentration of PFHxS (mean 2.18 mg/L) and total cholesterol, LDL, total cholesterol/HDL ratio and non-HDL cholesterol, as well as increased risk of high blood cholesterol number (Fisher *et al.*, 2013).

A Danish study found a positive correlation between plasma PFOA and PFOS and total cholesterol in 753 individuals of a Danish middle-aged population drawn from a larger cohort (Eriksen *et al.*, 2013).

In a Norwegian mother-child cohort study from 2003-2004, a clear correlation between blood concentrations of seven PFASs and HDL cholesterol in the blood was demonstrated. This was particularly true for PFOS, with an average concentration of 13 ng/mL (Starling *et al.*, 2014).

Overweight
Many studies have demonstrated that PFOS, PFOA and other PFASs can be transmitted from pregnant women to the foetus (including Fei *et al.,* 2007; Gützkow *et al.,* 2012; Vedsted *et al.,* 2013). It is also shown that early exposure to PFASs may result in overweight in later life. A study of 665 pregnant Danish women found a positive correlation between their PFOA concentrations in blood serum and overweight/obesity among their daughters when they reached 20 years of age (Halldorsson *et al.*, 2012).

Breast milk
Transfer of PFASs to breast milk is limited and concentration in breast milk is only a few percent of the concentration of the same substances in maternal blood (Fromme *et al.*, 2010). A published study suggests there may be a correlation between concentrations of PFOS and PFOA in the mother's blood and a shortening of the lactation period for women who have other children (Fei *et al.*, 2010).

Effect on birth weight
An analysis of data retrieved from the Danish birth cohort, consisting of more than 90,000 pregnant Danish women in the period from March 1997 to November 2002, found that the higher the maternal blood plasma concentration of PFOA, the less the birth weight. This correlation was not found for PFOS (Fei *et al.*, 2007). In a follow up study, a link between maternal PFOA levels in early pregnancy and both lower birth length and shorter abdominal circumference was found. For each ng/mL increase of PFOA, the birth length decreased by 0.069 cm and the abdominal circumference decreased by 0.059 cm (Fei *et al.*, 2008).

Studies from foreign cohorts both confirm and disprove this link between PFASs levels in maternal blood and birth weight and birth length (Maisonet *et al.*, 2012; Savitz *et al.*, 2012ab).

Effect on women's fertility
An analysis of data from 1,400 pregnant women taken from the Danish birth cohort revealed a correlation between the time it took a woman to become pregnant and the concentrations of PFOA and PFOS in her blood. This finding suggests that PFASs can reduce women's fertility (Fei *et al.*, 2009; 2012). This result was not confirmed in a recent small study of 222 Danish couples (Vestergaard *et al.*, 2012). In a Norwegian cohort study, Whitworth *et al.* (2012) found the same correlation as Fei *et al.* but only for women who had given birth before (parous).

There is evidence of a significant correlation between serum PFOA and PFNA concentrations and endometriosis, which among other factors reduces fertility (Louis *et al.*, 2012).

In a study from the USA, women with high blood levels of PFASs had an earlier onset of menopause than women with the lowest concentrations, taking some other factors into account (Taylor *et al.*, 2014).

Effect on male fertility - decreased sperm count
A study of 105 young Danish men showed that the ones with the highest concentrations of PFASs in their blood had reduced sperm quality with fewer normal sperm (Joensen *et al.*, 2009). Similar results were obtained in studies of Americans and in Greenland (Raymer *et al.*, 2011; Toft *et al.*, 2012).

In an area in the USA (Mid-Ohio Valley), where drinking water was contaminated with PFOA from a factory making fluoropolymers, it was demonstrated that children with the highest PFOS/PFOA concentrations in their blood had a 3-6 month delay in the onset of puberty (Lopez-Espinosa *et al.*, 2011).

Effect on insulin level
A study of gene expression profiles from Norwegian menopausal women showed a clear effect of PFOA on fatty acid metabolism and the function of insulin (Rylander *et al.*, 2011).

Effect on the thyroid gland
Data from NHANES' 1999-2006 studies involving almost 4,000 people found a correlation between high serum concentrations of PFOS (> 36.8 ng/mL) and PFOA (> 5.7 ng/mL) and thyroid disease in the general American population (Melzer *et al.*, 2010).

Data from NHANES' 2007-2008 studies have been used to assess the effect of PFOS, PFOA, PFNA, PFDA, PFHxS, and 2-(N-methyl-perfluorooctane sulfonamido) acetic acid (CAS 2355-31-9) on six indicators of gland function; the concentration of triiodothyronine increased with increased PFOA in blood serum, and thyroxine concentration increased with the concentration of PFHxS (Jain, 2013).

Effects on the immune system
An investigation of children aged 5 and 7 years from the Faroe Islands in the Atlantic showed that commonly prevalent exposures to PFOS, PFOA, PFHxS, PFNA and PFDA measured in blood serum were associated with lower antibody responses to childhood immunizations (vaccinations) and an increased risk of antibody concentrations below the level needed to provide long-term protection against diphtheria and tetanus (Grandjean *et al.*, 2012).

A study from Taiwan showed significantly higher PFASs serum concentrations in children with asthma compared to children without asthma (Dong *et al.*, 2013). In a new study from the United States, a link between PFOA and asthma among 12-19 year-olds was found (Humblet *et al.*, 2014). This correlation was not found for the other analysed PFAAs (PFOS, PFNA and PFHxS).

Hyperactive children
Data from NHANES' 1999-2004 survey and the C_8 Health Project in the USA showed a positive association between the concentration of PFASs in the blood and hyperactivity in children (ADHD) (Hoffman *et al.*, 2010). A later study identified a link between ADHD and PFHxS in the blood (Stein and Savitz, 2011). In addition, a link between high concentrations of PFOS, PFNA, PFDA, PFHxS and FOSA (but not PFOA) in children's blood and significantly increased impulsiveness was found (Gump *et al.*, 2011). In this study, PFHxS was the second most abundant PFAS in the blood with a mean blood concentration of about 6 ng/mL. The mean concentration of PFOS was about 10 ng/mL, while the mean concentration of PFOA was about 3 ng/mL.

Uric acid
Three major studies have shown a positive correlation between PFOA and uric acid in the blood. Uric acid is a natural product of purine metabolism. In a study of an adult population with a high environmental exposure to PFOA, for both PFOA and PFOS, a significant positive correlation with

uric acid levels was found. The correlation was particularly evident for PFOA (Steenland *et al.*, 2010b).

Chronic kidney disease
Chronic kidney disease is a growing health problem in the USA and in other countries (Coresh *et al.*, 2011). As it is well known that the kidney is a major target organ for PFASs, the relationship between serum concentrations of PFOS and PFOA and chronic kidney disease was examined in a study of the 1999-2008 NHANES data from 4,500 people. For the exposed, a significant positive association between PFOS and PFOA and chronic kidney disease, which was independent of normal confounders, was found (Shankar *et al.*, 2011).

Increased risk of cancer?
Studies from the work environment suggest a potential cancer risk when working with PFASs substances. Workers engaged in the production of PFOSF who are exposed to this starting material for the production of PFOS had a significantly increased morbidity from liver and bladder cancer (Alexander *et al.*, 2003).

A study of workers at a fluoropolymer plant in the USA showed that they had an increased risk of kidney cancer with a statistically significant exposure-effect relationship (Steenland *et al.*, 2012). The most vulnerable residents near the plant experienced double the risk of kidney cancer (Viera *et al.*, 2013) and triple the risk of testicular cancer (Barry *et al.*, 2013).

In a Danish cohort study of about 57,000 healthy subjects aged 50-65 years selected in 1993-1997 and followed until July 2006, there was no significant increase in cancer incidence from exposure to PFOS and PFOA, but for PFOS, a small increased risk of prostate cancer was found for the 3 highest quartiles relative to the lowest (Eriksen *et al.*, 2009). As in the case of exposure to low background levels in the population, it is difficult to detect effects.

In Greenland, where breast cancer has been increasing dramatically in recent years, a case-control study has been made in which blood serum concentrations of various environmental contaminants (incl. PFOS PFHxS, FOSA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA and PFTrDA) were measured and compared between breast cancer patients and a control group (Bonefeld-Jørgensen *et al.*, 2011). Breast cancer patients had significantly higher concentrations of PFOS with a median of 45 ng/mL compared to 21.9 ng/mL in the controls. Similarly, the median of the sum of PFCAs was higher in the breast cancer patients (8.0 ng/mL) than in the controls (5.2 ng/mL).

A new study from Sweden showed that 200 older men with prostate cancer had higher concentrations of 6 different PFASs in their blood than the 186 controls; the difference was not statistically significant (Hardell *et al.*, 2014).

In June 2014, an IARC expert group evaluated the carcinogenicity of PFOA (Benbrahim-Tallaa *et al.*, 2014). On the basis of limited evidence in humans that PFOA causes cancer of the kidneys and testes, as well as limited evidence from studies in experimental animals, the working group classified PFOA as possibly carcinogenic to humans (Group 2B). The group also assessed the lack of knowledge about the mechanisms of the effect of the substance.

6.3.3 Experimental studies
As shown in LOUS report (Lassen *et al.*, 2013)[3], the vast majority of experimental studies concern PFOS and PFOA, but there gradually more data is becoming available that concern FTOHs and the short-chain and long-chain perfluoroalkyl acids (PFAAs).

[3] If the specific references are not provided, please see the LOUS survey (Lassen et al., 2013).

Uptake, transformation, distribution, half-lives and elimination

It is known from animal studies that examined polyfluoroalkylated substances (PFASs) are absorbed almost completely in the gastrointestinal tract after oral exposure and in the lungs by inhalation, while the substances are absorbed through the skin to a limited extent. In an *in vitro* study, 2 % of the applied ammonium PFOA (APFO) penetrated through rat skin during 48 h (Fasano *et al.*, 2005). The uptake through human skin was 0.05 % (34 times slower). For 8:2 FTOH, the skin absorption was <1 % after 6 hours of exposure in rats (Fasano *et al.*, 2006).

Perfluoroalkyl acids (PFAAs) are considered inert and non-metabolised in the body as the strong C-F bond prevents normal metabolism. Any PFAA precursor will ultimately be transformed into PFAAs. This includes the fluorotelomers. Thus 8:2 FTOH is converted in several steps, including via the 2H,2H-perfluorodecanoic acid to PFOA and to a lesser extent, to PFNA (Martin *et al.*, 2005).

PFAAs have a low affinity for fatty substances and are almost completely bound to serum albumin in the blood. Binding affinity and the binding site of albumin depends on the animal species and the specific PFAS substance (Bischel *et al.*, 2011).

PFAAs are mostly associated with the cell membrane surfaces. Due to their surface-active properties, PFAAs can change the structure, properties, and function of the membrane (Jones *et al.*, 2003). The cell membrane becomes "fluid", the calcium channels are enhanced and the permeability becomes greater. As result, the calcium concentration inside the cells becomes larger. PFOS exerts most effects on the membrane in *vitro*, but PFHxS is also active (Liao *et al.*, 2009).

Although perfluoroalkane sulfonic acids (PFSAs) and perfluoroalkyl carboxylic acids (PFCAs) appear to be closely related in structure, they have different biological effects *in vitro* and *in vivo*. PFOS is generally more toxic and accumulative than PFOA. The toxicity generally increases with the length of the perfluoroalkyl chain, and with increasing chain length, more of the substance is accumulated in the liver, which is the primary target organ (Kudo *et al.*, 2001).

The primary excretion of PFAAs in experimental animals occurs through the kidneys into urine. Transport via the membrane-transport proteins and re-absorption in the kidneys appear to be the fundamental mechanisms that the PFCAs with relatively long chain length have longer half-lives in rats (Kudo *et al.*, 2001). In these studies, females have more efficient hormone-related clearance than males due to less re-absorption. This explains why PFOA is more toxic in male rats than in female rats (Han *et al.*, 2011).

The excretion of the non-polar fluorotelomer alcohols is different from the above. For 8:2 FTOH, the majority (> 70 %) in experiments with rats was excreted in faeces either unchanged or conjugated (Fasano *et al.*, 2006). Less than 4 % was excreted in the urine, about a quarter of this as PFOA. A week after exposure, 4-7 % of the dose was still present in animal tissues with the highest concentrations in fat, liver, thyroid and adrenal glands.

The half-life of PFASs in the blood depends on the substance, the animal species and the sex. From the blood the substances are distributed to various organs or excreted through the urine. In general, the half-life in blood is longer for PFSAs than for PFCAs, and it is also increased with longer chain length and shorter in females. In rats and mice, the half-life is typically hours or a few days, in the monkeys a little longer and in humans several years for e.g. PFOA, PFOS and PFHxS (Olsen *et al.*, 2007). The half-life is also dose-dependent, with longer half-lives for the lower concentrations, which are typically what people are most exposed to (Seals *et al.*, 2011). The reason for the special long half-life in the blood in humans is that humans have the highest degree of re-absorption (> 99 %) in the kidney (Harada *et al.*, 2005).

Toxicological mechanisms
In rodent studies the liver is - as mentioned above - the target organ for PFASs. In the liver the PFAAs "imitate" analogous non-fluorinated fatty acids, so the metabolism of lipids is affected. Like the fatty acids, PFAAs are bound to the so-called liver fatty acid-binding protein (L-FABP) (Luebker *et al.*, 2002). Most PFAAs activate the so-called "peroxisome proliferator-activated alpha" receptor (PPARα). This activation increases with the chain length of PFAAs until C_9, and the activity increased to a greater degree with PFCAs than with PFSAs. The activity increases in the order: PFBS <PFOS <PFHxS <PFBA <PFPeA <PFHxA <PFOA (Wolf *et al.*, 2012).

A typical effect, especially of PFAAs with long chains (C_7 -C_{10}), is the inhibition of the cells' internal communication, necessary for normal growth and function. This inhibition can lead to various diseases, *inter alia* cancer (Upham *et al.*, 2009).

The metabolism of many PFAAs, especially PFOA and PFOS, can generate reactive oxygen substances (ROS) and thus induce oxidative DNA damage (Eriksen *et al.*, 2010).

PFASs are endocrine disrupters and affect the function of the thyroid hormones by binding to thyroxine (T_4) plasma transport protein and transthyretin (TTR), and can thus reduce T_4 concentration in the blood. The binding is, however, far weaker than the binding within the thyroxine itself. In an *in vitro* test, decreased binding in the order: PFHxS > PFOS/PFOA > PFHxA > PFBS was observed, while PFBA and fluorotelomer alcohols (FTOHs) had no effect (Weiss *et al.*, 2009).

In experiments with rats, PFASs have a specific impact on the function of neurons in the hippocampus region of the brain. Among the sulfonates, PFOS had the greatest impact. For perfluoroalkyl carboxylic acids the effect grew by chain length and PFTeDA had the strongest effect (Slotkin *et al.*, 2008).

PFOS and substances (precursors) that may degrade to PFOS
PFOS includes both perfluorooctane sulfonic acid and its salts (sulfonates). PFOS and more complex substances that can degrade to PFOS with a perfluoroalkyl chain of 8 carbons have traditionally been the substances which have given rise to the greatest concern. Despite a comprehensive ban on the production and of most previous applications, PFOS is often still present in higher concentrations in the environment, animals and humans than other PFASs. Substances such as EtFOSE, MeFOSE, EtFOSA and MeFOSA, precursors to PFOS, may to some extent be accumulated in organisms, but they decompose rapidly to PFOS, and have the same effects as PFOS. The same is true of sulfonamides with shorter or longer perfluoroalkyl chains.

As mentioned, PFOS as an acid and four salts (CLP Index no. 607-624-00-8) have a harmonized classification as carcinogenic, toxic to reproduction and acute toxicity in category 4.

PFOS is the best-studied substance. The acute toxicity in adult rats is moderate by oral administration with an LD_{50} of 250 mg/kg bw; however, the acute toxicity of PFOS is 25 times higher in newborn mice (Lau *et al.*, 2004). Repeated exposure of rodents and monkeys induces decreased body weight and increased liver weight and liver damage. In short-term studies, the NOAEL for liver effects in rats was 5 mg/kg bw/day, while the NOAEL for thyroid hormone effects in monkeys was only 0.15 mg/kg bw/day (Seacat *et al.*, 2002; 2003).

In a two-generation rat study the NOAEL for PFOS was determined to be 0.1 mg/kg bw/day (Luebker *et al.*, 2005).

PFOS and its derivatives are not mutagenic in various test systems, but may increase the genotoxicity of other chemicals, such as the drug cyclophosphamide (iron bridge *et al.*, 2007).

PFOS and its precursor EtFOSE induce cancer in the liver, and thyroid and mammary glands in animal studies (Seacat *et al.*, 2003; Thomford *et al.*, 2002).

EFSA in 2008 established a tolerable daily intake of PFOS of 150 ng/kg bw/day (EFSA 2008).

Other perfluoroalkyl sulfonates and precursors
Among the other PFSAs, perfluorohexane sulfonate (PFHxS) and perfluorobutane sulfonate (PFBS) are the best studied.

PFHxS - PFHxS (6) has a perfluoroalkyl chain of 6 carbon atoms, and is thus two links shorter than PFOS. PFHxS is more toxic than PFBS. In a two-generation study in the rat, lipid metabolism was affected by daily oral exposure to 0.3 mg/kg bw for 42 days. Liver damage was found in the parent generation after exposure to 3 mg/kg bw/day (Butenhoff *et al.*, 2009). In this study, the NOAEL was 1 mg/kg bw/day. There was no damage observed in offspring at the lowest applied exposure at 10 mg/kg bw/day, which is therefore the NOAEL for offspring. In another recent study, as a consequence of exposure of mouse embryos for a single dose of 9.2 mg K-PFHxS/kg bw on postnatal day 10, the mice displayed behavioural disturbances as adults (Viberg *et al.*, 2013).

PFBS - PFBS is much less toxic than PFOS but large doses can affect the liver, kidneys and blood parameters at concentrations about 50 times higher than that of PFOS (Lau *et al.*, 2007). The NOAEL for PFBS as a potassium salt in a two-generation rat test was determined to be 100 mg/kg bw/day for the parents, which is 1000 times higher than the NOAEL (0.1 mg/kg bw/day) for PFOS (Liederbach *et al.*, 2009).

PFBSF - Perfluorobutane sulfonyl fluoride (PBSF), which is the starting material for the production of PFBS, is an acid fluoride and as such is very reactive. It is self-classified as a skin and eye irritant and acutely toxic (see C & L inventory on ECHA's website).

***N*-Methyl perfluorooctane sulfonamido ethyl acrylate**- *N*-Methyl perfluorooctane sulfonamido ethyl acrylate (C_4-acrylate) is a derivative of PFBS used as an industrial intermediate. The substance has a low acute toxicity, but is irritating to eyes and causes skin allergies. It has a short half-life in rats. Metabolites have not been studied (Kjølholt *et al.*, 2015).

Perfluoroalkyl carboxylic acids (PFCAs) and precursors
The most important of the PFCAs is PFOA (C_8), which includes both perfluorooctanoic acid and its salts. In animal studies, the target organ for PFOA is the liver, where peroxisome proliferation is induced (Kennedy *et al.*, 2004).

The acute toxicity is moderate with an oral LD_{50} of between 430 and 1,800 mg/kg bw/day. It is thus less acutely toxic than PFOS. The liver is the target organ and weight loss, liver enlargement and changed lipid metabolism are observed (Kennedy *et al.*, 2004).

In a 90-day oral study with rats, the LOAEL for liver changes was determined to be 0.3-1 mg PFOA/kg bw/day (Loveless *et al.*, 2006), while the NOAEL was 6 mg PFOA/kg bw/day for male rats and 30 mg PFOA/kg bw/day for female rats (Butenhoff *et al.*, 2012).

Many animal studies, particularly on mice, have shown that the pregnant female's exposure to PFOA can damage the foetuses (Lau *et al.*, 2003; 2007).

For pregnant mice fed PFOA by gavage, there was slower foetal development, reduced birth weight by exposure to 1 mg/kg bw/day and higher mortality of pups and delayed eye opening by exposure to at least 5 mg PFOA/kg bw/day (Lau *et al.*, 2006; Wolf *et al.*, 2007).

In another study, maternal exposure to 1-3 mg PFOA/kg bw/day resulted in a delay of e.g. breast development in offspring with subsequent disruption of milk production when the offspring were full-grown (White *et al.*, 2011).

PFOA is also known to be carcinogenic in the liver, pancreas, testes, and mammary gland (Biegel *et al.*, 2001).

Norway has suggested classifying PFOA (acid and ammonium salt) as acute toxic category 3, eye-irritant category 2, carcinogen category 2 and toxic for reproduction. ECHA's Risk Assessment Committee has concluded that PFOA's classification should be: Repr. 1B and STOT RE 1 (ECHA, 2011).

EFSA in 2008 established a tolerable daily intake of PFOA 1,500 ng/kg bw/day, which is 10 times higher than the equivalent of PFOS (EFSA, 2008).

Other perfluoroalkyl carboxylic acids

PFBA - In rodents, perfluorobutanoic acid, PFBA (C_4) is toxic to the liver and affects the thyroid gland. It induces peroxisome proliferation in rat livers but less so than for exposure to PFOA. In a 90-day study with rats, the NOAEL value for PFBA was 30 mg/kg bw/day in females and 5 mg/kg bw/day in males (Butenhoff *et al.*, 2012).

PFHxA - Perfluorohexanoic acid, PFHxA (C_6) is also hepatotoxic. In a 90-day gavage study in rats a NOAEL value for PFHxA of 20 mg/kg bw/day was identified based on effects on the liver and blood parameters (Loveless *et al.*, 2009). In another study, in which PFHxA was administered in drinking water, a NOAEL of 50 mg/kg bw/day in males and 200 mg/kg bw in females was determined (Chengelis *et al.*, 2009). This is higher than the NOAEL for PFOA.

PFNA - For perfluorononanoic acid, PFNA (C_7), the liver is also the target organ in rodents (Mertens *et al.*, 2010). Exposure leads to weight gain, disruption in the metabolism of glucose and induction of peroxisome proliferation (Fang *et al.*, 2012). The substance is also immunotoxic and teratogenic (Fang *et al.*, 2008). Several effects of PFNA are confirmed in population studies. For young people, a correlation has been demonstrated between increasing PFNA concentrations in the blood and a decrease in blood insulin and increased sugar content and HDL cholesterol levels in the blood (Lin *et al.*, 2009). Furthermore, there was a correlation between blood levels and the disease ADHD (Hoffman *et al.*, 2010). In the EU, Sweden has proposed PFNA to be classified as toxic to reproduction.

PFDA - The target organ for perfluorodecanoic acid, PFDA (C_{10}) is also the liver, where it induces peroxisome proliferation and acyl-CoA oxidase (ACOX) enzyme activity in rodents (Brewster and Birnbaum, 1989). It is considerably more toxic to the liver than PFOA (Olson and Anderson, 1983; Kawashima *et al.*, 1995). Besides, PFDA affects the thyroid gland (Harris *et al.*, 1989) and the function of the testes in rats (Book Staff *et al.*, 1990). In the EU, PFDA has been suggested to be classified as toxic to reproduction by Sweden.

PFUnDA - Perfluoroundecanoic acid, PFUnDA (C_{11}), has been poorly studied. In an *in vitro* study, low concentrations of PFUnDA activated PPARα in mice (Wolf *et al.*, 2012). A weak association between PFUnDA exposure and thyroid hormone disorders in humans have been demonstrated (Bloom *et al.*, 2010).

PFDoDA - In rodents, perfluorododecanoic acid, PFDoDA (C_{12}), causes reduced body weight at 5 mg/kg bw/day, and the substance is toxic to the liver with increased cholesterol levels in the blood at 10 mg/kg bw/day (Shi *et al.*, 2007). The same levels of PFDoDA have a specific degrading effect

on the testes, and 0.2 mg/kg bw/day lowers testosterone levels (Shi *et al.*, 2010). The substance is self-classified in the EU as a skin and eye irritant and acutely toxic.

PFTrDA - There are no animal tests of perfluorotridecanoic acid, PFTrDA (C_{13}), but some studies suggest a possible disruptive effect on thyroid hormones in humans (Ji *et al.*, 2012; Kim *et al.*, 2011).

PFTeDA - Perfluorotetradecanoic acid, PFTeDA (C_{14}) has not been studied much, but as mentioned above, PFTeDA with PFOS had the strongest effect on neurons in the brain among the investigated perfluoroalkyl carboxylic acids (Liao *et al.*, 2009).

8:2 Fluorotelomers

All 8:2 fluorotelomers including alcohols, acrylates and phosphates can decompose/metabolize to PFOA and PFNA, and can potentially have the same effects as these. Conversion is, however, not complete and more reactive unsaturated 8:2 fluorotelomer carboxylic acids and 8:2 fluorotelomer aldehydes are formed (Martin *et al.*, 2005).

The most important and best studied substance is 8:2 fluorotelomer alcohol (8:2 FTOH) which, in rodents, induces increased liver weight and liver enzymes. The NOAEL value is determined to be 5 mg/kg bw/day (Kudo *et al.*, 2005).

In a sub-chronic study in which 8:2 FTOH was administered orally to rats, the NOAEL was determined at 5 mg/kg bw/day, and 25 mg/kg bw/day induced liver necrosis (Ladics *et al.*, 2008).

A mixture of FTOHs with 27 % 8:2 FTOH has, in addition, been shown to be toxic to reproduction. NOAEL for reduced litter size was 25 mg/kg bw/day (Mylchreest *et al.*, 2005).

8:2 FTOH was shown to reduce testosterone levels and cause hormonal effects in studies in human cell cultures *in vitro* (Maras *et al.*, 2006; Vanparys *et al.*, 2006; Rosenmai *et al.*, 2013).

Norway has proposed 8:2 FTOH classified as toxic to reproduction.

Other fluorotelomers

The main substances in this group are 6:2 fluorotelomers and 10:2 fluorotelomers. They have increasing application because of the ongoing phase-out of 8:2 fluorotelomers. All 6:2 fluorotelomers can, among other things, be degraded/metabolized to PFHpA and PFHxA, and 10:2 fluorotelomers can be degraded/metabolized to PFDA and PFUnDA (Butt *et al.*, 2010). The toxicity of the intermediates of these transformations is greater than the toxicity of the final product, the PFCAs; the intermediate stage of the conversion of 6:2 FTOH was surprisingly more toxic than the intermediates of the conversion of 8:2 FTOH (Rand *et al.*, 2014).

The most important and best studied compound in this group is 6:2 fluorotelomer alcohol (6:2 FTOH) which shows acute toxicity in experimental animals (LD_{50} (o, r) of 1.75 g/kg bw). In accordance with this, 6:2 FTOH is self-classified as acute toxic cat. 4 in EU REACH.

In a 90-day sub-chronic study in rats exposed to 6:2 FTOH via gavage, 250 mg/kg bw/day resulted in fatal kidney damage. The NOAEL was determined to be 5 mg/kg bw/day in males and 125 mg/kg bw/day in females (Serex *et al.*, 2014).

Estrogenic effects of 6:2 FTOH have also been demonstrated in studies in human cell cultures (Maras *et al.*, 2006; Vanparys *et al.*, 2006).

A series of 6:2 fluorotelomers used as reactive intermediates are self-classified as skin and eye irritants. These are:

- 6:2 Fluorotelomer iodide,
- 6:2 Fluorotelomer sulfonyl chloride,
- 6:2 Fluorotelomer acrylate (6:2 FTAC) and
- 6:2 Fluorotelomer methacrylate (6:2 FTMAC).

6:2 FTAC and 6:2 FTMAC are, among other substances, important intermediates in the polymer industry, and were recently evaluated for toxicological properties in a report prepared by the consultant company ENVIRON (2014). The acute toxicity of the substances is negligible with oral LD_{50} in rats and mice of 2,000 to 5,000 mg/kg, but the substances irritate the eyes and skin. In a 28-day rat study with oral exposure to 25 mg 6:2 FTAC/kg bw/day, increased kidney weight was recorded, and the NOAEL was determined at 5 mg/kg bw/day. In a similar study, 6:2 FTMAC was demonstrated to affect the incisors and liver and kidney weight. The NOAEL was the same.

Data on 10:2 fluorotelomers are scarce. In a test with human cell cultures, 10:2 diPAP had only limited endocrine disruptor effects as opposed to 8:2 diPAP (Rosenmai *et al.*, 2013).

Technical mixtures of C_8-C_{14} perfluoralkyl ethyl alcohols are used (8:2 FTOH to 14:2 FTOH) as starting materials for the preparation of polyacrylates with perfluoroalkyl chains (among other substances). This mixture is self-classified as skin irritant and acute toxic, and contains the particularly hazardous 8:2 FTOH.

6.4 Exposure of children

The textile products for children most often containing PFASs are snowsuits, ski and all-weather jackets, outer pants and rainwear with accompanying mittens, caps and hats.

When the children wear the clothes, the direct exposure routes occur in particular from dermal contact with the substances in the textiles and skin absorption after release of the substances from contact with the clothes. This includes the use of mittens to wipe the face. The use of impregnated backpacks may also result in exposure. Activity and sweat production is likely to increase the absorption of the substances.

In an early study by industry scientists, it was assumed that dermal absorption of PFASs by contact to children's clothing would be 50 % of the content (Wasburn *et al.*, 2005). The assumed contact area for a 1-year old child was 4,500 cm^2, 5,200 cm^2 for a 4-year old and 11,200 cm^2 for a 12-year-old.

A less common, but direct exposure possibility is if the baby is sucking/licking on the clothes, including the mittens. Saliva can increase the transfer of PFASs. It was estimated by Wasburn *et al.* (2005) that the intake of PFASs by this route corresponded to the amount extracted in a migration test to saliva.

Transfer from hand to mouth may also occur. By this route, there is a possibility of direct oral uptake of PFASs, which are initially migrated from the products to the hand.

When the clothes are used, stored indoors or dried, dependent on the temperature there may be possible evaporation of volatile PFASs (e.g. fluorotelomers and sulfonamides) to indoor air. Textile dust is formed as well, wherein the less volatile PFASs (salts, carboxylates and sulfonates) concentrate. The more volatile substances may be condensed on the surface of the dust by a drop in temperature (Ahrens *et al.*, 2012). Children's clothes will contribute to the overall general PFAS indoor

exposure with indoor air and house dust. The quantitative importance is, however, difficult to determine.

Where impregnated clothing in greater quantities is stored, relatively high indoor concentrations of PFASs can be reached. This supposition was confirmed by air measurements in shops for sports and recreational equipment, where levels were measured up to 47, 286 and 58 ng/m3 of 6:2, 8:2 and 10:2 FTOH, respectively (Langer *et al.*, 2010 ; Schlummer *et al.*, 2013). Calculations also showed that exposures of the staff from indoor air wereof the same magnitude as the normal dietary intake.

Other studies showed that emissions from impregnated clothes for indoor air were dominated by 8:2 FTOH, and the hourly emission was between 1 and 47 ng 8:2 FTOH/kg clothes and 8-60 ng total FTOH/kg clothes (Bringewatt *et al.*, 2013).

The main contribution to exposure for indoor PFASs would normally be the use of impregnation products for shoes, curtains, tablecloths, blankets, mattresses, bedding and upholstery fabrics. Here exposure would predominantly be to FTOHs and other PFOS/PFOA precursors (including FTCAs and FTSAs), and not PFOS/PFOA themselves. During re-impregnation of clothing with spray products, there would also be the possibility of very high acute exposures from air where the process takes place (Feilberg *et al.*, 2008).

The exposure of children to PFAS in textiles should be seen in the context of the children's other, and in many cases higher, exposures to PFASs from the external environment (air, soil, water), food, cooking utensils, food packaging and drinking water. Danish children's average daily intake of PFOS and PFOA in the diet is estimated to be 1.8-7.2 ng/kg bw and 0.3-8.3 ng/kg bw, respectively (EFSA 2008; Lassen *et al.*, 2013).

Estimates for the intake of other PFASs are not available, but consumption of precursors (fluorotelomers, etc.) with the diet and drinking water is presumed to be less important, as the substances are not water-soluble and have low concentrations in the food, apart from PFAS originating from the packaging. Intake of PFASs in drinking water is relatively more important for children than adults because children drink more relative to their body volume. Sources of PFASs other than food and drinking water are estimated to correspond to up to half of the intake in total.

An estimate of the average exposure of the European population is shown in Table 13 below, which is also included in the initial health screening in section 3.1. This inventory does not take into account children's special circumstances, and does not include exposure to PFAS in consumer products such as all-weather clothing. Moreover, data are missing for many PFASs.

TABLE 13
EXPOSURE OF SELECTED PFASs FROM DIFFERENT SOURCES (FROMME *ET AL.*, 2009)

Exposure source	Daily intake (pg/kg bw)							
	PFOS		PFOA		ΣFTOHs		ΣFOSE/FOSA	
	Average	Max	Average	Max	Average	Max	Average	Max
Indoor air	0.9	0.9	4.7	4.7	38	105	460	2,050
Outdoor air	1.3	12	0.1	1.0	3.0	3.2	1.1	12
House dust	16	1,028	32	4,217	103	1,017	983	2,033
Food	2,817	11,483	1,500	4,483	-	-	217	6,866
Drinking water	22	87	23	130	-	-	-	-
Total intake	**2,857**	**12,611**	**1,560**	**8,836**	**144**	**1,125**	**1,661**	**10,962**

In theory, the use of the clothing outdoors could also give rise to outdoor air pollution. Inhalation of PFASs in the outdoor air is, however, usually considered to be a minor source of exposure (see Table 13) (Sasaki *et al.*, 2003).

6.5 Health risk assessment of the use of children's clothing treated with PFASs

6.5.1 Dermal exposure

The total concentration of the examined PFASs in the textile materials ranged from 18 to 407 µg/m². In the three materials with the highest concentrations (120 to 407 µg/m²), 8:2 FTOH accounted for more than 65 % of the concentration of PFASs; the rest were mainly the PFCAs where PFOA was dominant. C_8 substances constituted 80-90% of PFASs.

Since the impregnation with PFASs occurs on the outside of children's clothes, and as PFASs in general have a very low skin permeability (see section 6.2.3), it is estimated that skin exposure to PFASs from outerwear does not contribute significantly to the children's exposure to PFASs. The exposure will be limited to the hands touching the outside of the clothing and to mittens used to dry the face. In in practice it is possible to ignore this exposure as it is negligible.

This assessment is different from the assessment of Washburn *et al.* (2005), who did not distinguish between the inside and outside of the clothing, and assumed that dermal absorption of PFASs by contact with children's clothing in all cases would be 50 % of the content of PFASs, and assumed a contact area of 4,500 cm² for a 1-year old child, 5,200 cm² for a 4-year old and 11,200 cm² for a 12-year old.

Because of these conflicting assessments, theoretical "worst case" calculations of dermal absorption of PFASs from three products (infant sleeping bag, snowsuit and rain jacket) are provided here as well as the resulting Risk Characterization Ratio (RCR) which is the ratio between the amount absorbed (internal dose) and the derived no-effect level (DNEL):

RCR = exposure (quantity absorbed)/ DNEL (both in ng/kg bw/day)

The results are shown in Table 14, but the scenarios are considered unrealistic in practice.

The DNEL used in the tables is a DNEL for PFOA indicated in a presentation of Bernauer (2010) from the German Federal Institute for Risk Assessment (BfR). This DNEL of 80-170 ng/kg bw/day

is calculated on the basis of human studies and blood data, and is 2-10 times lower than the DNEL calculated from animal studies. DNEL for PFOA is used as worst-case in the present study, where part of the quantity of PFASs consists of other, presumably less dangerous substances.

TABLE 14
CALCULATION OF A THEORETICAL WORST-CASE SCENARIO FOR DERMAL EXPOSURE AND SKIN ABSORPTION

	Textile product		
	Infant sleeping bag	Snowsuit	Rain jacket
Age of the child (year)	1	4	12
Weight of the child (kg)	12	17	40
Contact area (cm^2)	4.500	5.200	11.200
PFAS concentration, max. extracted for 30 min. ($\mu g/m^2$) (from Table 8)	214	100	280
PFAS release (ng/cm^2)	21,4	10	28
Lifetime of the clothing (days)	730	730	1.095
Contact time (hours/day)	10	8	6
PFASs release ($ng/cm^2/day$)	0,0120[4]	0,0046	0,0064[5]
Max. average daily exposure during the lifetime of the product ng ng/kg bw	 55 4,6	 24 1,4	 72[6] 1,8[7]
Skin absorption (% max)	2	2	2
Max. average daily uptake ng ng/kg bw	 1,1 0,09	 0,5 0,03	 1,4[8] 0,04[9]
Derived No-Effect Level, DNEL (ng/kg bw/dag)	80-170	80-170	80-170
Risk Characterization Ratio (RCR)	0,0005-0,0011	0,0002-0,0004	0,0002-0,0004[10]

The calculated maximum average daily exposure of 1.4 to 4.6 ng/kg bw is relatively large compared to the total intake of PFOA + FTOHs for adults of approximately 10 ng/kg bw/day and the maximum average intake for children with the diet of 8.3 ng PFOA/kg bw, shown in Table 13 above (section 6.3). However, absorption through the skin (<2 % of the exposure) is 45 times less than the absorption of PFASs ingested with the diet (about 90 %). On the basis of the uptake, the RCR is calculated.

Furthermore, the chosen scenario is based on highly conservative values that lead to significant overestimation of the exposure and the risk:

- Maximum parameters for skin absorption and contact area.
- Skin adsorption tests were with solutions; dry contact is likely to produce less skin absorption.
- Only a small part of the body is expected to be exposed to the impregnated outside of the clothing.

[4] Corrected from 0,012
[5] Corrected from 0.0096
[6] Corrected from 108
[7] Corrected from 2,7
[8] Corrected from 2,7
[9] Corrected from 0,5
[10] Corrected from 0.003-0,006

- It is unlikely that sweat can extract as much PFASs as the organic solvents used for the chemical analysis of the material. The lower content of PFASs found in the saliva migrations fluid (more than 10 times lower) would be more realistic (see next section).

All in all, the exposure from dermal contact of impregnated clothes even in the worst case does not result in a contribution to the background load of any health significance, a fact illustrated by the very low RCR values of 0.0002 to 0.001. RCR should be ≥ 1 for the existence of a potential risk.

6.5.2 Oral exposure via saliva

As mentioned in section 5.2.3, eight children's textile materials with a relatively high content of PFASs were selected for migration tests using artificial saliva. The release was measured over three hours.

The composition of PFASs in the migration liquid was found to be significantly different from the composition of PFASs in textile materials. In the migration liquid, as expected, the more water-soluble acids dominated: PFCAs, PTCA and FTSAs. The content of non-water soluble FTOHs was generally low.

It appears from section 5.2.3 that the average concentration of PFASs in the artificial saliva was about 1 % of the PFAS content of the textile materials. For the group of PFCAs, the migrated quantity corresponded to approximately 6 % of the content of the materials. Specifically for PFOA the migration corresponded to about 8 %, meaning that total PFCAs and PFOA specifically represented about 55 % and 30 %, respectively, of the total content of PFASs in the migration liquid. The highest release rate for PFASs was 2.87 µg/m² of fabric. Of this, PFOA accounted for 1.46 µg/m², the other PFCAs accounted for 1.22 µg/m², 8:2 FTOH accounted for 0.15 µg/m², and all other PFASs accounted for 0.05 µg/m². This means that the C_8-fluorine compounds represented more than half of the exposure.

It appears that only a small part of the content of extractable PFASs in the fabric migrated to the artificial saliva. Therefore, it is uncertain as to whether the method of standardized artificial saliva consisting of ionized water with salts adjusted to pH 6.8 is realistic. The shortfall compared to natural saliva includes enzymes and mucin (glycoprotein). However, the artificial saliva is recognized and accepted for standardized migration tests, and it has been used for many of the Danish EPA consumer projects.

In a previous study of Wasburn *et al.* (2005) it was projected (but not demonstrated) that saliva can increase the transfer of PFASs when the baby sucks on clothing. It is estimated by the authors that the amount of PFASs emitted from the material in this way could be equivalent to the amount measured in the analysis extracts. This would require, however, that the saliva is as effective an extraction solvent as the organic solvents used, and that the baby sucks on the clothes constantly. Both seem unrealistic.

By the oral exposure route, only a minor part of the fabric is in contact with the mouth. In worst-case scenarios, however, it is assumed that the mouth is in contact with the entire surface as shown in Table 15.

TABLE 15
CALCULATION OF A THEORETICAL WORST-CASE SCENARIO FOR ORAL EXPOSURE AND UPTAKE OF PFASS VIA SALIVA

	Textile product		
	Infantsleeping bag	Snowsuit	Rain jacket
Age of the child (year)	1	4	12
Weight of the child (kg)	12	17	40
Contact area (cm^2)	4.500[11]	5.200[12]	11.200[13]
PFASs release to "saliva" ($\mu g/m^2$) (from Table 9)	2,67	1,88	5,48
PFASs release (ng/cm^2)	0,27	0,19	0,55
Lifetime of the clothing (days)	730	730	1095
Contact time (hours/day)	10	8	6
PFASs release ($ng/cm^2/day$)	0,00015	0,00009[14]	0,00013[15]
Max. average daily exposure during the lifetime of the product (ng) (ng/kg bw)	 0,7 0,06	 0,6 0,03	 1,4[16] 0,04[17]
Oral absorption (% max)	90	90	90
Max. average daily uptake (ng) (ng/kg bw)	 0,6 0,05	 0,4[18] 0,02[19]	 1,3[20] 0,03[21]
Derived No-Effect Level, DNEL, for PFOA (ng/kg bw/dag)	80-170	80-170	80-170
Risk Characterization Ratio (RCR)	0,0003-0,0006	0,0001-0,0003[22]	0,0002-0,0004[23]

The results show that even in a worst-case scenario the intake of PFASs via saliva is minimal and therefore of no health significance. This is confirmed by the low RCR at 0,0001 til 0,0006[24].

In addition, only a small portion of the clothing, such as sleeves and mittens, in fact will be exposed to saliva. Conversely, the use of artificial saliva probably results in less efficient extraction of the PFASs in the material.

6.5.3 Exposure via indoor air

Emission of PFASs to indoor air

Within the framework of the project, the release of substances into the air was not prioritised as an area to examine, as this has already been investigated in a number of foreign studies.

[11] Corrected from 4500
[12] Corrected from 5200
[13] Corrected from 11200
[14] Corrected from 0,00011
[15] Corrected from 0,00031
[16] Corrected from 3,5
[17] Corrected from 0,09
[18] Corrected from 0,5
[19] Corrected from 0,03
[20] Corrected from 3,2
[21] Corrected from 0,08
[22] Corrected from 0,0002-0,0004
[23] Corrected from 0,0005-0,001
[24] Corrected from This is confirmed by the low RCR at 0,0001 til 0,0006

In a recent German study the release during over five days of the volatile PFASs 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, EtFOSE and MeFOSE from four jackets was measured by flow tests (Knepper *et al.*, 2014). Reference is made to the report for details of the methods used.

Significant differences in the emission rates for the three FTOHs were observed, as shown in Table 16. The extractable content of 10:2 FTOH (determined by solvent extraction) was emitted in the range of 27-70 % in the course of five days, while only 7-16 % of 8:2 FTOH was emitted during that period. The authors of the study are surprised that the release rate of 10:2 FTOH (expressed as percentage of the content in the clothes) is significantly higher than the release of 8:2 FTOH, since the vapour pressure of 10:2 FTOH (13.3 Pa) is significantly lower than the vapour pressure of 8:2 FTOH (45.1 Pa).

Two of the products had PTFE membranes (it is not specified whether they were also coated with PFAS-based surface coatings). The release expressed as a percentage of the content was greater from these than from the other two jackets, and from one of the jackets, the release was more than 10 times higher than the release from the other jackets.

For two of the jackets (J2 and J8) the experiment continued for two more days. The release of 6:2 FTOH continued at almost the same daily rate, while the rates for 8:2 FTOH and 10:2 FTOH decreased significantly compared to the release the first five days.

TABLE 16
RELEASE OF FTOHs TO AIR FROM FOUR JACKETS OVER A PERIOD OF 5 DAYS (KNEPPER *ET AL.*, 2014)

Sample No	With PFTE membrane	Release to air over a period of 5 days (average of two analyse)					
		6:2 FTOH		8:2 FTOH		10:2 FTOH	
		µg/m²	% of the content of the clothing	µg/m²	% of the content of the clothing	µg/m²	% of the content of the clothing
J2		0.71	53.7	3.46	8.87	5.69	40.4
J8	X	<dg	-	5.83	16.0	7.95	69.7
J10		<dg	-	4.26	6.51	2.74	27.2
J14	X	<dg	-	90.6	17.6	110	60.5

* Standard deviations are not reproduced here, but are generally small compared to the mean.
<dg = below detection level. Quantification level was 3 times the detection level, which depending on the analysed substances, varied from 0.1 to 5.2 ng/L in the sampling liquid (figures not indicated in µg/m²).

The release of EtFOSE and MeFOSE were below the quantification level from all jackets.

Other studies have also shown that the emission from impregnated clothes to indoor air was dominated by 8:2 FTOH. Bringewatt *et al.* (2013) found that the emission within one hour was between 1 and 47 ng 8:2 FTOH/kg clothes and 8-60 ng total FTOHs/kg load.

In contrast to the above results, it was found in another new German study (Dreyer *et al.*, 2014) that the emission of 6:2 FTOH was higher than the emission of 8:2 FTOH in six of the eight samples tested. The issue was expressed as the total emissions from an outdoor jacket per day in this study (surface not specified). The emission of 6:2 FTOH from the eight jackets ranged from 17 to 8,798 ng/day (average 3,301 ng/day), while the emission of 8:2 FTOH ranged from 47 to 803 ng/day (average 277 ng/day), and 10:2 FTOH varied from 11 to 803 ng/day (mean 138 ng/day). The emission of the fluorotelomer carboxylic acids 6:2 FTCA, 6:2 FTCA and 6:2 FTCA ranged from <0.6 to 74 ng/day, with the exception of one sample, where the emission of 8:2 FTAC was measured at 556 ng/day and the emission of 10:2 FTCA to 597 ng/day. The emission of 10:2 FTOH was also exceptionally high in this sample. The emissions of MeFOSA, EtFOSA, MeFOSE and EtFOSE were below the detection limit for all samples.

Release during the entire life cycle of the products
A difficulty in the interpretation of the results is that release of PFASs from the textiles over time is not measured. As in the case of volatile substances, it is expected that the substances evaporate over a period of time and it may significantly affect the results when the products are produced in relation to the time the sample is taken. As pointed out by Knepper *et al.* (2014), there are no results showing the quantities of volatile PFASs that are released immediately after production and during transport of the products.

When a significant proportion of FTOHs, in products that can be many months old, are released over five days, it must be anticipated that significantly higher quantities have been released earlier in the life cycle of the products. Analyses of impregnating agents indicated in Appendix 4 show that the concentrations of the volatile 8:2 FTOH and 10:2 FTOH was of the order of 1000 times greater than the concentrations of PFCAs. In the tested fabrics, FTOHs concentrations are typically less than one tenth of the concentration of PFCAs. The analysed impregnation agents may be different than the agents that have been used to produce the studied textiles, but the results indicate that there may be a significant emission of volatile PFASs early in the lifecycle of the treated products. In addition, FTOHs will probably be continuously formed by degradation of the perfluorinated side chains. The total emissions over the products' life cycle may therefore be significantly higher than the concentrations of the non-bound substances that can be measured by analysis of the textiles.

The background exposure via indoor air is partially covered by the data in Table 13, where children are assumed to have maximum exposure.

As PFCAs and PFSAs are not volatile, the PFASs in the air are dominated by FTOH derivatives and possibly PFSA precursors, while the more polar PFASs may also be concentrated in house dust - probably mainly formed by the decomposition of the FTOH derivatives.

As concern the impregnated children's clothing, a special case would be the indoor environment in schools and institutions, which can be influenced by high volume of impregnated outdoor wear kept indoors, especially in winter. The indoor air in institutions was not investigated during this project by specific measurements.

Foreign studies of emissions from impregnated clothes to indoor air measured releases in the range of 1-47 ng 8:2 FTOH/kg clothing/hr and 8-60 ng total FTOHs/kg clothing/hour (Bringewatt *et al.*, 2013).

Knepper *et al.* (2014) measured the release of PFASs over five days to indoor air from four jackets and reported the results in µg/m² and in percentage released from the clothes. For 8:2 FTOH, the release was 3-90 µg/m² and 6-17 % of the content of the clothing.

In Dreyer *et al.* (2014), the total daily emissions to air of 8:2 FTOH from an outdoor jacket averaged 227 ng. The actual concentrations in indoor air depend on the room size and ventilation.

Health risk assessment of exposure via indoor air
In the absence of specific indoor air measurements where outerwear is stored in schools and institutions, an absolute "worst-case" can be estimated using the measured concentrations of FTOHs in the indoor air of outdoor stores of about 400 ng FTOHs/m³ (Schlummer *et al.*, 2013). In Table 17, exposure via indoor air is estimated on the basis of these measurements and a RCR was calculated as the maximum average annual daily uptake in ng/kg bw divided by the DNEL.

TABLE 17
CALCULATION OF A THEORETICAL ABSOLUTE WORST-CASE SCENARIO OF EXPOSURE AND UPTAKE OF PFASs VIA MEASURED INDOOR AIR.

Institution	Kindergarten	School
Age of the child (year)	4	12
Weight of the child (kg)	17	40
Ventilation rate (m^3/d)	8	14
Concentration of PFASs in indoor air (ng/m^3)	400	400
Intake pr. exposure day (ng)	3,200	5,600
Contact time (days/year) (winter semester)	150 of 365	100 of 365
Contact time (hours/day)	10 of 24	8 of 24
Actual exposure time (days/year)	62	34
Max. average inhalation (ng/dag)	543	522
Uptake % max of inhaled	90	90
Max. average daily inhalation (ng/kg bw)	32	13
Max. average daily uptake (ng) (ng/kg bw)	 488 29	 470 12
Derived No-Effect Level, DNEL (ng/kg bw/dag)	80-170	80-170
Risk Characterization Ratio (RCR)	**0.17-0.36**	**0.07-0.15**

The daily uptake via the indoor air in this example is very significant, and the calculated RCR of up to 0.36 is very high, but still less than one. The scenario is extreme, as large amounts of newly unpacked PFAS-impregnated textiles are stored in the premises in outdoor stores.

Another possible, and probably more realistic, calculation is based on the expected amount of impregnated textiles in the rooms where the children are located. Data from Bringewatt *et al.* (2013) for the releases of PFASs from impregnated clothing of 47 ng per kg clothing and per hour can be used for the calculations. This is a worst-case scenario, since, as mentioned above, Dreyer *et al.* (2014) measured an evaporation of 227 ng FTOHs/day as the maximum from an impregnated jacket. This result corresponds to <10 ng/hour. The results are given in Table 18.

TABLE 18
CALCULATION OF REALISTIC WORST-CASE SCENARIO OF EXPOSURE AND UPTAKE PFASS VIA INDOOR AIR BASED ON EMISSION FROM CLOTHES

Institution	Kindergarten	School
Age of the child (year)	4	12
Weight of the child (kg)	17	40
Ventilation rate (m^3/d)	8	14
Volume of the room (m^3)	400	4,000
Emission rate for PFASs (ng/kg clothing/hour)	47	47
Number of outerwear of 0.1 kg	40	400
Weight of the clothing (kg)	4	40
Emission rate (ng/time)	188	1.880
Contact time (days/year) (winter semester)	150 of 365	100 of 365
Contact time (hours/day)	10 of 24	8 of 24
Average emission rate (ng/dag)	1,880	15,040
Concentration in the room (ng/m^3)	4.7	3.8
Air exchange (per hour)	0	0
Intake pr. exposure day (ng)	38	53
Actual exposure time (days/year)	62	34
Max. average inhalation (ng/dag)	6.5	4.9
Uptake % max of inhaled (%)	90	90
Max. average daily inhalation (ng/kg bw)	0.54	0.12
Max. average daily uptake (ng) (ng/kg bw)	 5.8 0.49	 4.4 0.11
Derived No-Effect Level, DNEL (ng/kg bw/dag)	80-170	80-170
Risk Characterization Ratio (RCR)	0.002-0.006	0.0006-0.0013

These calculations in Table 18 give much lower results than the calculations in Table 17. Evaporation from the clothes gives a small contribution to children's exposure to PFASs, resulting in low RCRs at 0.0006 to 0.002 - far below one. The calculation does not take into account that in practice there would be air circulation reducing the exposure. Furthermore, it is not taken into account that wet outerwear may be placed in a drying cabinet. The parameters used for e.g. room sizes, number of children and amount of clothes are, however, roughly estimated and uncertain.

6.5.4 Total uptake of PFASs released from clothing

If the "realistic worst case" contribution to the total absorption from dermal and oral exposure to PFASs in a snowsuit (0.03 + 0, 03 ng/kg bw/day) is added to the contribution from a 4-year-old's inhalation in institutions (0.49 ng/kg bw/day), the overall uptake can be estimated at 0.55 ng/kg bw/day. When compared with a DNEL (Derived No-effect Level) of 80-170 ng/kg bw/day, a total risk characterization ratio (RCR) of 0.003 to 0.008 can be estimated. The value is far below 1. If the RCR is greater than 1, there is a risk of health effects. In the calculation it is assumed that the child is wearing a snowsuit all day. If, alternatively, for part of the day the child uses an infant sleeping bag (small children), rainwear and mittens, the RCR values would still be very low. Danish children's average daily intake of PFOS and PFOA with the diet is estimated to be 1.8-7.2 ng/kg bw/day and 0.3-8.3 ng/kg bw/day, respectively (the two substances jointly form the majority of the total intake of PFASs).

The total intake in the indoor environment, which only includes the contribution from the clothes in institutions in this study, could therefore potentially be of the same magnitude as the intake of PFOA with the diet. Besides the described exposure pathways, relevant exposure pathways could include air inside a car or a tent/sleeping bag and indoor air/dust in the home.

6.6 Summary and conclusion

Exposure to PFASs
When exposed to PFASs, almost complete absorption occurs after oral ingestion or inhalation, while the substances hardly penetrate intact skin. The PFASs taken up is bound to albumin in the blood and is distributed to most organs, but in most cases, the primary target organ is the liver.

The residence time in the blood and distribution to other tissues and organs depends on the fabric, the extent of the exposure and the animal species. The residence time in the blood increases with chain length; sulfonic acids have a longer residence time than carboxylic acids. The residence time is relatively short in rodents, slightly longer in monkeys and very long in humans. The residence time is also longer in males than in females.

The residence time in the blood is partly associated with the ease of PFASs excretion. The acids are excreted in the urine by the kidneys. This excretion occurs in female rats more quickly and efficiently than in males who have more efficient re-absorption in the kidney. The difference is determined by the sex hormones. Whether the same gender differences in excretion in humans exist is not known. Humans have the most efficient re-absorption (99 %) in the kidney, which means that humans have difficulties in excreting the substances, which instead will accumulate in the organs. This factor means that a risk assessment cannot be based on results from animal tests, as is usually done.

Health effects
Both in animal tests in which each substance is tested, and in population studies in which participants are exposed to a mixture of PFASs, PFASs affect fatty acid metabolism with increased risk of onset of liver injury, increased cholesterol and low-density lipoproteins in the blood, obesity and effects on the insulin balance. An association between high concentrations of PFOS and PFOA in blood and thyroid disorders and endocrine disruption have also been demonstrated. Effects on hormone systems are also observed in animal studies.

A possible association between PFOS and PFOA in the blood and women's difficulties in becoming pregnant, endometriosis and earlier onset of menopause have been observed. The women most exposed to PFOA, who nevertheless become pregnant, give birth to babies with lower birth weight and birth length. Apparently there is also an effect of PFASs on male fertility in the form of reduced sperm count and later onset of puberty.

Occupational studies, population studies and animal tests suggest that certain PFASs can induce hepatic, pancreatic, bladder, kidney, testicular, prostate and breast cancer. All of these cancers are largely hormone-related. IARC has recently classified PFOA as possibly carcinogenic to humans (Group 2B).

Other population studies suggest that PFASs can influence the immune system in children and induce asthma and cause hyperactivity (ADHD).

There are also studies that suggest a link between PFOS and PFOA in the blood and chronic renal disease. In view of the kidney's central position in the body's detoxification of PFASs, this effect appears plausible.

Exposure of children
For children's textiles, the present study shows that the main PFASs to which the children may be exposed in Denmark are PFOA and 8:2 FTOH, a precursor of PFOA.

The textile products for children which often contain PFASs are snowsuits, ski gear and all-weather clothing in the form of jackets, pants and waterproof clothing with accompanying mittens, caps and hats. When the children have the clothes on, the direct exposure pathways occur from dermal contact with the substances in textiles and subsequent skin absorption. Activity and sweat production is likely to increase the absorption of substances.

Since the impregnation with PFASs occurs on the outside of children's clothes, and because PFASs generally have a very low (<2 %) skin permeability, it is estimated that skin exposure to PFASs from outerwear confined to the hands touching the outer side of the clothing and any mittens used to wipe the face has no significant impact on the children's total exposure to PFASs. This assessment is supported by worst-case calculations for three scenarios of skin exposure/uptake for children of 1, 4 and 12 years, respectively. The calculations show a maximum average daily absorption ranging from 0.03 to 0.09 ng/kg bw, about 100 times lower than the intake with the diet.

In addition, PFASs can be transmitted orally with saliva when the child chews or sucks on the clothes. This was verified by extraction of PFASs from the textile materials with artificial saliva. The extracted PFASs were particularly the water-soluble acids with PFOA as the main substance. Despite the almost complete (> 90 %) uptake of PFOA in the gastrointestinal tract, the worst case calculations showed that the daily intake of PFASs via saliva is insignificant compared with other sources, at a level ranging from 0.03 to 0.08 ng/kg bw/day for the three scenarios. In addition, only a small part of the clothing (sleeves, gloves) would actually be exposed to the saliva. Conversely, the use of artificial saliva, which lacks some natural components, probably results in lesser extraction of the PFASs in the material.

When the clothes are used, stored indoors and dried, depending on the temperature, volatile PFASs (e.g. fluorotelomers and sulfonamides) may evaporate to indoor air. Furthermore, textile dust is formed wherein the less volatile PFASs (salts, carboxylates and sulfonates) are concentrated, and on which the more volatile substances may be condensed on the surface by a drop in temperature.

In the absence of actual measurements of indoor air containing PFASs in institutions and schools where children stay indoors especially during the winter months, it is difficult to estimate children's exposure to PFASs in the indoor air with great certainty. The presence of sources other than children's clothing is sparsely studied. Based on the measurement results from foreign outdoor shops selling outdoor clothing, two scenarios were established, which result in a maximum average daily uptake of respectively 29 ng/kg bw for a 4-year old and 12 ng/kg bw for a 12-year-old. These calculated contributions result in significant exposures and intakes far greater than the intake of PFASs with food. However, the scenarios are extreme and theoretical. Institutions for children are likely to have lower volumes of impregnated textiles than an outdoor clothing store.

Therefore, alternative realistic worst-case exposure calculations based on data on the evaporation of PFASs from the clothes were made. The result in this scenario was a maximum average daily uptake of 0.49 ng/kg bw for the 4-year old and 0.11 ng/kg bw for the 12 year old. These calculations yield much lower total intake with a small contribution to children's exposure to PFASs. The scenario does not take into account that in practice there would be air circulation, reducing the exposure.

Several of the parameters used in the exposure calculations for indoor air are estimated and particularly uncertain. Furthermore, there is great uncertainty about the presence of other sources of

PFASs to the indoor air. It is therefore proposed to carry out actual indoor air measurements in rooms where children stay indoors in winter and their outerwear is stored.
An important contribution to exposure to indoor PFASs could be application of impregnating sprays for shoes, curtains, tablecloths, blankets, mattresses, bedding and upholstery fabrics. It will require a specific study to quantify the exposures in comparison to the exposure to PFASs in children's clothing more accurately.

The study shows that children's direct exposure to PFASs from their winter clothing results in negligible exposures to PFASs and low RCR values (even when the contributions are added together), and they are thus assessed not in themselves to cause health problems.

However, the drying and storage of winter clothing indoors in schools and institutions may possibly in the worst case cause significant indoor air exposures and high RCR, thereby contributing to the health problems that can result from the combined exposure to PFASs.

7. Environmental assessment

7.1 Introduction

In recent years, considerable attention has been paid to the perfluorinated substances (PFASs), also in the environmental context, because they have proven to be highly resistant to degradation in nature, can be transported over long distances and detected everywhere in the environment (e.g. the Arctic) and may accumulate in food webs.

Some PFASs are included in DEPA's list of undesirable substances (LOUS) and recently, as part of the general surveys of the substances on this list, a literature-based review of the substances' behaviour, fate and effects in the environment has been conducted (Lassen *et al.*, 2013). The LOUS survey is followed up by several other ongoing or recently completed projects dealing with various health and environmental aspects of the PFASs and their possible alternatives.

The environmental description and the assessment below are, to a large extent, based on the results of those projects that included the collection and assessment of the latest knowledge in the field. There are no specific environmental data on many PFASs, so the assessment must essentially be based on extrapolation/"read across" of data for the most studied substances within the group: PFOS and PFOA.

7.2 Environmental classification

PFOS and its derivatives are the only substances that have a harmonized classification in accordance with the EU CLP Regulation. With regard to environmental properties they are classified as Aquatic Chronic 2, "Toxic to aquatic life with long lasting effects" (H411). For a number of other substances, self-classifications are made; however, they largely address the substance health properties.

7.3 Behaviour and environmental fate

There is general agreement internationally that perfluorinated substances are extremely persistent in the environment as a result of the inherent strength of the chemical bond between the fluorine and carbon atoms in the perfluorinated chain of the substances; that is, the portion of the molecules where all atoms of the carbon chain are fully substituted by fluorine molecules (e.g. Brooke *et al.*, 2004; US EPA, 2009; Lassen *et al.*, 2013). The perfluorinated part of PFASs is considered, almost regardless of the chain length, to be non-degradable under realistic environmental conditions.

7.3.1 PFOS and precursors

PFOS is biodegradable neither aerobically nor anaerobically. It does not degrade abiotically by hydrolysis and does so only marginally by photolysis. The calculated half-life is 41 years by hydrolysis and 3.7 years by photolysis (UNEP, 2006). For degradation in the atmosphere based on model calculations, the half-life of PFOS is estimated at 114 days, indicating that even in the atmosphere there would not be significant degradation (Environment Agency, 2004). The volatility of PFOS is also low. The substance binds strongly to soil, sediment and sludge (Jensen *et al.*, 2012).

PFOS bioaccumulates, but via a different mechanism than the general one for bioaccumulative substance; the substance therefore does not meet the EU criteria for bioaccumulation (B) and a PBT

assessment. Bioaccumulation factors, BCF for fish in the range of 2800-3100, have been found. Monitoring studies in the Arctic have shown biomagnification of PFOS in food webs. Thus, for top predators such as seals, bio-magnification factors (BMF) at> 160 have been found (UNEP, 2006). The potential for bio-magnification is higher in marine food webs than in terrestrial food webs (Jensen *et al.*, 2012).

The substances EtFOSE and MeFOSE, precursors for PFOS, may undergo primary degradation, both biologically and by hydrolysis, and are thereby transformed into PFOS. The calculated half-life of EtFOSE in the atmosphere (half-life 16 hours) is shorter than the half-life of PFOS because it can undergo primary degradation, but will ultimately be converted to PFOS (Brooke *et al.*, 2004).

7.3.2 PFOA and precursors

PFOA is similar to PFOS in terms of behaviour and fate in the environment. The substance is very persistent and does not decompose under environmentally relevant conditions. This statement applies to both biotic and abiotic degradation. PFOA is more water-soluble than PFOS and may be transported over long distances to an even greater extent than PFOS via water systems, including ocean currents (Environment Canada, 2012b).

The presence of PFOA in the environment, far from potential sources, may be due to transport of the substance itself, but may also be caused by long-distance transport of more volatile precursors (Ellis *et al.*, 2004). Such precursors could be FTOHs, fluorotelomer iodides and fluorotelomer olefins and/alkenes.

PFOS monitoring studies have demonstrated the presence of PFOA in top predators in Arctic food webs, albeit at a slightly lower level than for PFOS. PFOA is found not to significantly bioaccumulate in fish in laboratory tests (Environment Canada, 2012b).

7.3.3 Long-chain perfluorinated carboxylic acids

For the four long-chain PFCAs (C11-C14) (PFUnDA, PFDoDA, PFTrDA and PFTeDA), Kjølholt *et al.* (2014) found that only a few specific studies on the four substances were available, and that the assessments of the substances in the prepared REACH Annex XV dossiers are based on read-across from other related compounds, including PFOA, to a significant extent. For all four substances it was concluded that they are vPvB substances and are therefore included on the candidate list under REACH.

Arctic monitoring studies have shown that a number of C_{14}-C_{15} PFCAs are also widely distributed in fish, invertebrates and top predators (Lassen *et al.*, 2013).

7.3.4 Short-chain perfluorinated carboxylic and sulfonic acids

As a result of growing concerns about the persistence and bioaccumulative properties of the long-chain perfluorinated substances (including PFOA and PFOS), a number of alternatives have been introduced including perfluorinated carboxylic and sulfonic acids with shorter chain lengths, especially C_4-based substances as PFBS. These substances have been demonstrated to be non-bioaccumulative, as they are rapidly excreted from the organisms in which they are taken up (Rayne *et al.*, 2009). However, they are persistent in the environment (Buck *et al.*, 2011).

7.3.5 Fluorotelomer alcohols

Fluorotelomer alcohols (FTOHs) such as 6:2 FTOH exhibit a higher degree of primary reactivity than the corresponding carboxylic acids, but are converted microbially in the environment to either the corresponding carboxylic acid or, where appropriate, to a carboxylic acid with a slightly longer chain length. It was thus found that 6:2 FTOH can degrade to PFHpA (heptanoate) or PFHxA (hexanoate), but no further (Buck *et al.*, 2011).

7.3.6 Polyfluorinated acrylates/methacrylates

Kjølholt *et al.* (2014) project that these esters would be hydrolyzed relatively quickly to the corresponding fluorotelomer alcohols and further on to short-chain perfluoroalkyl carboxylic acids which do not degrade further. Since the perfluoroalkyl part of the molecules is highly stable, it is the substances are not seen as biodegradable. Some of the substances are persistent (P) or very persistent (vP) according to the REACH criteria but none of them meet the criteria for bioaccumulation (B) or toxicity (T).

7.4 Environmental effects

In terms of environmental effects, a reasonable number of data are available for the most common and most frequently found PFASs, the C_8 acids PFOS and PFOA, while data for other PFASs and short-chain analogues of carboxylic and sulfonic acids are scarce or absent. Estimates of toxicity in the environment (almost exclusively for the aquatic environment) are therefore made typically by extrapolation/"read across" from data for PFOS/PFOA.

7.4.1 PFOS and precursors

The acute toxicity of PFOS to aquatic organisms is moderate with reported LC_{50} values for fish in the range from 4.7 to 133 mg/L, while for *Daphnia* and other invertebrates, EC_{50} values of 2.7 to 223 mg/L are reported. Algae seem to be a bit less sensitive (Brooke *et al.*, 2004). The lowest chronic NOEC (No Observed Effect Concentration) to aquatic invertebrates is found to be 0.25 mg/L and for fish, 0.3 mg/L (Brooke *et al.*, 2004).

An overview of the most sensitive endpoints for PFOS in various aquatic organism groups is shown in Table 19.

TABLE 19
OVERVIEW OF THE MOST SENSITIVE ACUTE AND CHRONIC ENDPOINTS FOR PFOS IN DIFFERENT AQUATIC ORGANISM GROUPS (BROOKE *ET AL.*, 2004)

Acute	Fish	*Pimephales promelas* (minnow) (96-h): LC_{50} = 4.7 mg/L *Oncorhynchus mykiss* (rainbow trout) (96-h): LC_{50} = 13.7 mg/L
	Invertebrates	*Daphnia magna* (48-h): EC_{50} = 27 mg/L *Mysidopsis bahia* (marine) (96-h): LC_{50} = 3.6 mg/L
	Algae	*Selenastrum capricornutum* (96-h): EC_{50} = 126 mg/L *Skeletonema costatum* (marine species) (96-h): EC_{50} > 3.2 mg/L
Chronical	Fish	*Pimephales promelas* (minnow) (42-days): $NOEC_{survival}$ = 0.3 mg/L
	Invertebrates	*Daphnia magna* (28-dage): $NOEC_{reproduction}$ = 7 mg/L *Mysidopsis bahia* (marine species) (35-days): $NOEC_{reproduction}$ = 0.25 mg/L
	Algae	*Selenastrum capricornutum* (96-h): NOEC=44 mg/L *Skeletonema costalum* (marine species) (96-h): NOEC>3.2 mg/L *Lemna gibba* (duckweed) (7-days): NOEC = 15.1 mg/L

7.4.2 PFOA and precursors

The toxicity of PFOA in the water environment is lower than the toxicity of PFOS. There are reports on acute toxicity to fish (LC50) in the range from 70 to 2,470 mg/L. On average, PFOA seems to be about 10 times less toxic to the aquatic environment than PFOS. The lowest reported chronic NOEC is 2.0 mg/L for the fresh water alga *P. subcapitata*. There are few findings concerning organisms in the soil, but in this compartment as well, PFOA appears to be less toxic than PFOS (Jensen *et al.*, 2012).

In some studies, a potential for endocrine disrupting effects of PFOA in fish is found. The thyroid hormone synthesis is thus hampered in the freshwater fish minnow at concentrations of 3-30 mg/L (although this is significantly higher than the concentrations that can be expected in the environment) (Environment Canada, 2012b).

7.4.3 Long-chain perfluorinated carboxylic acids

The toxicity of the long-chain PFCAs in the aquatic environment is found to be moderate to low, with EC_{50}/LC_{50} values ranging from 8.8 to 285 mg/L (Environment Canada, 2012b).

7.4.4 Short-chain perfluorinated carboxylic acids

These substances are shown not to be bioaccumulative, as they are rapidly eliminated from the organisms in which they are taken up, but no data on their toxicity to aquatic organisms exist.

7.4.5 Fluorotelomer alcohols

For the short-chain fluorotelomer alcohol 5H 4:1 FTOH, a short-term (4.5 hours) EC_{50} (inhibition of photosynthesis) to algae (*P. subcapitata*) of 4.85 mM or about 1,125 mg/l was reported (Ding *et al.*, 2012).

7.4.6 Polyfluorinated acrylates/methacrylates

The substances' acute toxicity to aquatic organisms varies from high to moderate. Relatively, the toxicity is as follows: 6:2 FTMAC > 6:2 FTAC > MeFBSE acrylate > PFO-TeAA (Kjølholt *et al.*, 2014).

7.4.7 Summary - environmental effects of PFASs

It is shown in the above that although overall limited data on the environmental toxicity of PFASs are available, there are, however, a number of data for PFOS, which must be considered the most toxic and environmentally problematic (of the most frequently detected PFASs).

Some data on the acute toxicity of PFOS to aquatic organisms exist for three trophic levels and similar chronic toxicity data for the same groups of organisms are available (fish, invertebrates and algae). Invertebrates have been shown to be the most sensitive group with a lowest NOEC = 0.25 mg/L. From this value, a PNEC (Predicted No Effect Concentration) of 0.025 mg/L can be calculated as a safety factor (assessment factor, AF) of only 10 is applied to the calculation in accordance with the REACH guideline No. R.10 (ECHA, 2008).

7.5 Environmental exposure

In the chapter on health assessment (Chapter 6), it is described how PFAS-based impregnation agents could be released from impregnated textiles in the use phase to humans and the environment, respectively.

Release of PFASs to the air would take place very slowly through migration to and evaporation from the surface of the impregnated fabric. Some substances have such low vapour pressure that the release would be minimal. The release would, moreover, increase proportionally with increases in the ambient temperature.

In addition, PFASs could be released from the textiles during use when exposed to rain or, to a lesser extent, by wear and tear. It is considered to be primarily the soil environment that would be exposed to these types of release.

Finally, PFAS-impregnated clothing could be released to the sewage system through the laundry. The immediate assessment is that most PFASs will primarily end in the sludge phase at treatment

plants, but a certain proportion of the more water-soluble substances could go through the treatment process and end up in the aquatic environment. This environmental exposure route is considered to be the most comparatively important, even if impregnated textiles are usually washed considerably less frequently than other clothing.

Environmental exposure in the waste disposal phase of the impregnated textile is projected to be low, as the textile products in Denmark ultimately are expected to be disposed of to waste incineration plants where efficient combustion at high temperature and the purification of flue gas, happening subsequently, would ensure that only negligible amounts of PFASs are released to the atmosphere. It cannot be ruled out that very low residue concentrations could be detected in slag and flue gas cleaning products.

Clothing exported for recycling would, as specified in Section 2.3, mainly be disposed of to landfills from which the PFASs would eventually be released.

7.6 Environmental risk assessment

The environmental hazardousness of PFOS, PFOA and other PFASs is based primarily on the substances' persistence and potential for bioaccumulation, not least among top predators in the marine food webs. Furthermore, for some substances the potential for endocrine disrupting effects have been shown, although these are likely only expressed at higher concentrations than are normally found in the environment. It seems unlikely that short-term effects could occur at the concentrations it is realistic to find in the environment.

It is estimated that the greatest environmental impact with PFASs from impregnated textiles (clothing items incl. footwear) for children would occur in connection with the washing of textiles and subsequent discharge of laundry water to the sewage system, from which the remains in sludge and treated wastewater could end up in soil or streams and/or coastal marine waters. A more detailed assessment will focus on the possible impact on fauna associated with aquatic environments.

The release to the atmosphere from impregnated textiles could play a role in the long-distance transport of bioaccumulative PFAS detected in Arctic areas. The levels found are discussed further below.

7.6.1 Exposure via wastewater

During the use phase of PFAS-impregnated clothing, laundry and thus domestic sewage are supposed to be the main sources and release pathways leading to environmental exposure for PFASs (excepting FTOHs and other substances with a high vapour pressure). Laundry water from consumers containing PFASs is discharged predominantly to the public sewage system and transported to a wastewater treatment plant in Denmark (and Europe). In the plant the PFAS would partly be degraded of PFASs, forming a distribution of the substances between the water phase and solid phase (sludge). The treated wastewater is usually discharged to a stream or directly to the sea, while the sludge in Denmark is typically either incinerated or applied to agricultural land.

In accordance with ECHA's guidance for environmental exposure (ECHA, 2012b) for the assessment of environmental exposure for laundry, a standard EU wastewater treatment plant with a capacity of 10,000 PE (person equivalent) is used. Each PE contributes 200 litres of water/day and 0.11 kg sludge/day. That total for a wastewater treatment plant of this size is 2,000,000 litres of water/day and 1.100 kg sludge/day. In the current context, the 10,000 PE are assumed to be distributed over 2,500 households, each consisting of four people.

The daily discharge (U_d) of each studied PFASs (PFASsx) with sewage from a standard household/family is in principle estimated for each PFASs by the following formula:

$$U_d = N * M * Q_{PFASsx} * C_{clothing,PFASsx} * R$$

where

U_d is the daily discharge of $PFASs_x$ from clothing wash per household,
N is the number of wash per day,
M is the weight of the clothing per wash,
Q_{PFASsx} is the percentage (%) of the clothing which includes the $PFASs_X$,
$C_{clothing,PFASsx}$ is the concentration (%) of $PFASs_X$ in the treated clothing,
R is the percentage (%) of $PFASs_X$ released per wash.

Worst case exposure of the recipient - For the assessment of the exposure via wastewater both discharges from clothing used by both adults and children will be included, so as to obtain an estimate of the total emissions from the use of PFASs in clothing.

For an initial, conservative estimate of the amount of PFASs derived from the laundry it will be assumed that all individuals in the catchment area of a standard treatment plant owns a set of PFAS-impregnated rainwear or other outerwear, i.e. 10,000 sets of waterproof clothing/outerwear. Furthermore, it is assumed that each set of rainwear/overcoats have a combined outer surface (jacket + pants) of 2 m² and are cleaned once a year. In addition, snowsuits and jackets for children are washed more often; it is assumed that the clothes are washed every month. It is assumed that ¼ of the people in the catchment have snowsuits and gloves, which overall results in an additional 1 m² for 1/4 of the persons.

The present study found a smaller release rate by washing than the German studies, in which it is found that the quantities released by washing may exceed the quantities found by analysing the content of non-polymeric PFASs in the clothing. For a conservative estimate, the higher release rates found in German studies will be applied here.

On this basis, it will be assumed that by each wash of rainwear/outerwear an amount equal to the total extractable content will be released (i.e. annually, an amount corresponding to the extractable quantity is released to waste water). It is uncertain how much can be delivered in the long run, but it will be assumed that the "worst case" from children's clothes washed every month would show that over a year, the released quantity could be 10 times the extractable content.

The clothes with the highest total content of PFASs in the present study (Table 8, F20; Mittens) contain approximately 400 µg/m², much of which is made up of FTOHs (8:2 FTOH) (the highest content of PFASs in rainwear/snowsuits was 280 µg/m², again predominantly in the form of 8:2 FTOH).

The total annual emissions of PFASs to wastewater from washing, U_y, is determined by the following formula, which is divided into two parts: the first part of the formula calculates the emissions of all persons in the catchment area of the treatment plant and the second part calculates the additional contribution from clothes from children (expected to be washed more often than the clothes for adults):

$$U_y = (C_{clothing,\ m2} * A_{Å} * N * PE)_{all} + (C_{clothing,\ m2} * A_{Å} * N * PE)_{children}\ (\mu g/year)$$

where

U_y is the total annual discharge of $PFASs_x$ from washing of the clothing to the waste water treatment plant,
$C_{clothing,\ m2}$ is the concentration of PFASs in the clothing in µg per m²,
$A_{clothing}$ is the surface area of the PFAS-containing clothing in m² per piece of clothing,
N is the annual number of wash for PFAS-containing clothing (on average),
PE is the number of persons in the catchment area of the waste water treatment plant which have PFAS-containing clothing.

The resulting concentration on the waste water, $C_{PFASs,\ spv.}$, subsequently can be estimated from :

$$C_{PFASs,\ ww.} = U_{Å}/(V_{ww} * 1000)\ \ (\mu g/L)$$

where

V_{ww} is the total annual volume of waste water going to the waste water treatment plant in m³.

In the specific case the following values are applied:

$C_{clothing,\ m2}$	= 400 µg/m²,
$A_{clothing}$	= 2 m² for clothing for adult and 1 m² for children's clothing,
N	= 1 for clothing for adult and 10 (extra) for children's clothing,
PE	= 10,000 for "all" and 2,500 for "children",
$V_{spv.}$	= 365 days * 2,000 m³/day = 730.000 m³/year.

The result is:

$$U_y = (400 * 2 * 1 * 10.000)_{all} + (400 * 1 * 10 * 2500)_{children} = 18.000.000\ \mu g/year$$

and

$$C_{PFASs,\ ww} = 18.000.000/(730.000 * 1000) = 0{,}025\ \mu g/L$$

The concentration of PFASs going to the treatment plant is calculated at 0.025 µg/L on this basis, which in a worst case scenario is not degraded or detained in the treatment plant, but discharged to the environment and thus, in this rough calculation, becomes the "predicted environmental concentration" (PEC).

In section 7.4.7, a PNEC of 25 µg/L for toxicity of the substance PFOS in the aquatic environment is calculated. PFOS is considered to be approximately 10 times more toxic than PFOA, and is probably also more toxic than the other PFASs. If the PEC/PNEC ratio (risk quotient) is greater than 1, there is a risk of ecotoxicological effects, but it is seen that the PEC/PNEC even in this conservative scenario, in which the toxicity of all PFASs are set equal to the toxicity of PFOS, is far below 1, namely 0.025/25 = 0.001. There is therefore no need for further refinement of the risk calculation for exposure via wastewater.

Further transport of substances - In the classical sense, there does not appear to be any significant environmental risk associated with the discharge of PFASs from textile washing to wastewater and subsequently to the aquatic environment. However, there are indications in the literature on endocrine disrupting effects of some PFASs (Environment Canada, 2012b) which, however, are not sufficiently substantiated to be evaluated here. The longer chain PFASs can bioaccumulate and biomagnify in top predators, which ultimately may have some effect when the contribution from the washing of textiles is aggregated with emissions from a variety of other sources. On the present basis, it is not possible to assess what the effects of substances released from the washing of textiles could be higher up in the food web, but the following, the quantities discharged with the laundry water as compared to emissions from other sources will be assessed based on approximate calculations.

Comparison with other sources - If the above conservative quantitative calculations based on the analysis results of this study are scaled up to the whole of Denmark (approximately 5,000,000 PE), a total emitted quantity of 9 kg PFASs per year that can be estimated as a "worst case". The actual emissions must be expected to be considerably less. An alternative calculation can be obtained based on the estimated content of PFAS-based impregnating agents in clothes sold in Denmark. In Jensen *et al.* (2008) it is estimated that the total content of PFASs in clothing sold in Denmark is 745 to 18,000 kg (incl. polymeric PFASs). In the present study, it is shown that the extractable PFASs are on average around 0.04 % of the total fluorine content in the textiles. There is some uncertainty in relation to how much could be released as compared to the extractable content; studies show that the release could be higher than the extractable content. If it is assumed that between 0.02 % and 0.06 % of the content of PFAS-based impregnating agents could be released as non-polymeric PFASs to the laundry water, the total release would range from 0.1 to 11 kg PFASs. This indicates that the actual quantities are likely somewhat lower than the quantities estimated as the "worst case".

As described above, the "worst case" concentrations in the influent to the wastewater treatment plant of PFASs derived from textiles is estimated at 0.025 µg/L. These calculations are performed as the "worst case", which has been calculated with the highest measured concentration of PFAS in the clothes. If instead the average of the measurements is used for the calculations, the concentration at the inlet water could be estimated to be about 0.007 µg/L. The actual measured concentrations in the influent to the wastewater treatment plant are slightly higher. Monitoring of PFASs in the inlet to municipal wastewater treatment plants found total concentrations of PFASs ranging from 0.010 to 0.075 µg/L (Beach *et al.*, 2006). The dominant PFASs in the inlet to the municipal wastewater treatment plants were PFOA (median of about 0,020 µg/L), PFHxA (0.008 µg/L), PFOS (0.004 µg/L) and PFNA (0.003 µg/L) (median values are read on a figure as the source only indicates the median values in tabular form for municipal and industrial wastewater treatment plants together). The results indicate that textiles may potentially be significant sources of PFASs to municipal wastewater treatment plants, but that there probably are other significant sources.

In reality, a substantial fraction of the volatile PFASs in the wastewater is released into the air by sewage treatment. Ahrens *et al.* (2011) calculates, based on air measurements over a wastewater treatment plant in Canada, a total release of PFASs to the air of 2.6 kg/year, with FTOHs as the dominant PFASs. The paper does not specify how much PFAS is emitted through other routes.

Foreign studies - Knepper *et al.* (2014) calculates the total amount of PFOA emitted with laundry water from the washing of textiles in Germany at 0.27 kg/year in one of three scenarios, in which the maximum emission was assumed. The total discharged amount of nine PFASs with the largest content in textiles is determined to be 0.76 kg/year in the same scenario (i.e. considerably smaller quantities than has been assumed in this "worst case" scenario). By comparing the annual transport in the Rhine, which is estimated at 464 kg PFOA, with the possible release of laundry water from 80

million people in the catchment area, it is calculated that emissions from washing contributes 0.05 % of the total input to the river.

Based on measured concentrations of PFOA in discharges from wastewater treatment plants that treat domestic sewage of 0.020 µg/L (which corresponds very well to what is found in Denmark, where PFOA represent the largest share of PFASs in the inlet), the authors calculate total annual releases of approximately 100 kg PFOA from wastewater treatment plants. The authors estimated that 0.25 % of these may originate from the laundry water from the textile washing (the 0.27 kg/year mentioned above). The authors do not indicate what could be the source of the remaining amount. The authors calculate the total amount of non-polymeric PFASs in textiles sold in Germany to be 30-75 kg/year under the assumption that the non-polymeric PFASs constitutes 0.1 % of the fluorine content of the PFAS-based impregnating agents. Given that the study finds that the release of PFOA is greater than the extractable content, it does not appear to be well documented as to why less than 1 % of the content of non-polymeric PFASs would end up in wastewater.

7.6.2 Exposure via air

Many PFASs, including the sulfonic acid PFOS, the carboxylic acid PFOA and the other PFSAs/PFCAs, have such low vapour pressure that release to the atmosphere is considered to be a minor pathway for releases to the environment as compared to releases by waste water. However, there are a number of PFASs, especially fluorotelomer alcohols (FTOHs), EtFOSE and MeFOSE that have higher vapour pressure, and their release to air is considered a major release pathway, as detailed in section 6.5.3.

Ellis *et al.* (2004) have provided some evidence from laboratory tests that long-range transport of PFASs can take place in the form of FTOHs, which are subsequently converted to the corresponding acids. Zhao *et al.* (2012) measured the prevalence of PFASs on transects along the east coast of Greenland and in the Atlantic from Europe to Antarctica, and also indicates long-range transport through the atmosphere as part of the explanation for those PFASs found in the Arctic, although long-range transport by ocean currents is regarded as more important. Emissions of FTOHs to the northern hemisphere are based on concentrations in the atmosphere; the half-lives of the substances are estimated to be around 100-1,000 t/year (Ellis *et al.*, 2003). If Denmark's contribution is considered to be in the range of 0.1-1 % of this total, it would be equivalent to 0.1 to 10 t/year from Denmark (Denmark demographically accounts for approximately 1 % of the EU and 0.1 % of the population of the northern hemisphere).

Contributions from children's clothing - As demonstrated in German studies over a five-day period, some 7-18 % (average 12 %) of the extractable content of 8:2 FTOH was released, while the release rates for 10:2 FTOH and 6:2 FTOH were 17-70 % (average 50 %) and 0-54 % (average 13 %), respectively. The release of volatile EtFOSE and MeFOSE was below the detection limit. With these rates it is expected that all the non-polymeric FTOHs is released during the useful life of the clothing. An important question is whether the clothes contained significantly higher amounts FTOHs immediately after production than at the time the sample was taken , likely several months after manufacture of the clothing. Furthermore, it is a question as to what extent FTOHs is formed during the useful life of the clothing. If it is assumed that the concentration of FTOHs in the clothing decrease exponentially and that measurements are made 3 months after manufacture, the concentrations immediately after manufacture would have been 10-50 times greater than the measured concentrations. There are no data to show whether this is so. An initial "worst case" estimate can be established based on the following assumptions:

- The number of snowsuits sold corresponds to the number of children born in the last 7 years (Statistics Denmark, 2013): Approximately 430,000 pieces. (note that this is slightly higher than estimated in section 2.2.3, which also takes reuse into account),
- Average FTOHs concentration: 110 µg/m^2,

- Surface area per. suit: 1 m^2/item, and
- Percentage of suits with PFASs: 30 %.

The total content is calculated to be 430,000 pieces x 110 µg FTOHs/m^2 x 1 m^2/pcs x 30 % = 0.14 kg FTOHs.

Besides snowsuits there is also a significant consumption associated with gloves, rainwear, jackets, etc. and in total, the amount of FTOHs is estimated to be 0.2 kg FTOHs. If it is roughly assumed that 10 times the extractable content could vaporize, the total reaches an amount of 2 kg FTOHs. Compared with the estimated contribution from Denmark at 0.1 to 10 t/year, it is in any case a small contribution that would come from children's clothes.

Another source could be the release of PFASs later in the life cycle of the textile by the decomposition of PFAS-based polymers. In a publication that discusses uncertainties and shortcomings of an inventory of the global emissions of PFASs (Wang *et al.*, 2014b), a number of scenarios have been prepared on the extent of releases from the biodegradation of PFAS-based polymers (particularly in textiles). It is anticipated that PFASs are continuously formed from the biodegradation of the products during use. The scenarios differ in two parameters: the life of the clothes and half-lives for biodegradation (only emissions from products in use are considered). The scenarios show that emissions of FOSA/FOSE (total substances groups) and different FTOHs (6:2 to 14:2) from the decomposition of PFAS-based polymers may well be of the same magnitude as the releases of these substances from all other sources combined. For 8:2 FTOH the scenarios estimate, for example, a total global emission in 2015 from the decomposition of PFAS-based polymers of between 100 and 350 tons/year, depending on the model parameters. It is not estimated how much of this total would result from the use of polymers in textiles, but it must be considered to be a substantial part. In the scenarios it is assumed that the products have a lifetime of between five and ten years, which is realistic for textiles for adults but probably too long for textiles for children. These calculations clearly indicate that textiles could be a significant source of emissions of volatile PFASs. If the assumptions in these scenarios are correct, releases from the textiles over the total use period could be significantly greater than estimated above.

In Denmark, the clothes after disposal (if not exported for reuse) are incinerated, but this is not the case in most other EU Member States. There are no results available demonstrating to what extent PFASs are released from the clothes by decomposition in a landfill, but the potential is very large compared to the release of non-polymeric PFASs in the course of the useful life of the clothing. Ahrens *et al.* (2011) have shown that the air above two investigated landfills in Canada contained 3-30-fold higher concentrations of PFASs (primary FTOHs) than the reference locations, and calculated a total emission of PFASs into the air from the two landfills of respectively 0.1 and 1 kg/year.

Foreign studies

German studies estimate a total release of PFASs from outdoor wear of about 24 kg/year in Germany, based on measurements of FTOHs in outdoor wear (Knepper *et al.*, 2014). *Per capita* it corresponds roughly to the above calculations for Denmark of 0.2 kg/year. Assuming that the highest measured value could represent the concentration immediately after production, the authors estimate that potentially 110 kg could be emitted. The authors also carried out a calculation which assumes that the products immediately after manufacture contained the maximum value for the Blue Sign mark of 50 mg/kg. In that case, the total emissions could correspond to 1,000 kg (approximately 50 times the emissions calculated based on actual measurements). The authors conclude that outdoor wear, at worst, could provide a significant contribution to the releases of FTOHs, but more knowledge is needed on the concentrations in the clothing earlier in its life cycle.

A new study on global sources of PFASs estimates, *inter alia*, the sources of global emissions of C_4-C_{14} PFCAs (Wang *et al.*, 2014a). Major sources are the production of fluoropolymers using the sodium salt of PFOA and PFNA followed by formation of PFCAs by the decomposition of discharged fluorotelomers (including fluorotelomers derived from fluorotelomer-based impregnating agents). The total formation based on the emissions of fluorotelomer for the period 2003-2015 was calculated at 13 to 1,902 tonnes for the period, which illustrates the considerable uncertainties that exist on the size of this source. Potentially (if the highest value is correct) the degradation of fluorotelomers may be responsible for almost half of the emissions.

7.6.3 Exposure in the soil environment

The direct exposure of the soil environment is assumed to be insignificant, and no attempt to estimate it has been done.

As a worst-case exposure of the soil environment by application of sewage sludge, it is assumed that all PFASs from the washing of textiles discharged to a wastewater treatment plant would be distributed to the sludge phase, after which the produced sludge is applied to farmland.

If it is conservatively assumed that all of the estimated annual "worst case" amount PFASs to wastewater of 5,600 mg follows the sludge, an annual average concentration in the sludge of 13.9 mg/tonne of sludge can be estimated.

If it is furthermore assumed that as a maximum, 7 tonnes/ha sewage sludge is applied on agricultural land and that the sludge is mixed homogeneously in the top 20 cm of soil, a soil concentration of approximately 30 ng/kg of soil = 0.000030 mg/kg of soil is obtained. This is such a low concentration that it is undoubtedly far below the level at which ecotoxicological effects would occur in the soil environment.

7.7 Summary and conclusion

Environmental effects of PFASs - Regarding environmental effects, there are a reasonable number of data for PFOS and PFOA, while data for other PFASs and short-chain homologues of carboxylic and sulfonic acids are scarce or absent. Estimates of the toxicity in the environment (almost exclusively the aquatic environment) are therefore typically made by extrapolation/"read across" from data for PFOS and PFOA.

The environmental toxicity of PFOS, PFOA and other PFASs is primarily due to the substances' persistence and potential for bioaccumulation, not least among top predators in marine food webs. There is a particular concern about the possible impact in the Arctic. Furthermore, for some substances, a potential for endocrine disrupting effects have been shown.

Environmental exposure via wastewater - On the basis of the calculated "worst case" discharges of PFASs with wastewater from washing clothes and PNEC (Predicted No Effect Concentration) values for PFOS, the most toxic of the described PFASs, a risk characterization ratio (RCR) of 0.001 is calculated. If the RCR is greater than 1, there is a risk for ecotoxicological effects. In the classical sense, it appears that there is no significant environmental risk to aquatic organisms associated with the discharge of PFASs to wastewater from washing of textiles and further on into the aquatic environment. However, there are indications in the literature regarding endocrine disrupting effects of some PFASs which are not sufficiently substantiated to be evaluated here.

As mentioned above, the main concern relates to the effects on top predators, particularly in the Arctic. Discharges of wastewater from laundry could contribute to the total emission of PFASs circulating in the environment. In German studies it is estimated that the laundry water from textile washing should only constitute 0.25 % of the sources of PFOA in sewage. "Worst case" scenarios used in this study indicate that emissions from the laundry could potentially be a significantly larger

source. Very different results in terms of discharges from washing of textiles have been found in this and previous studies, and a significant uncertainty is related to whether PFASs can be released from the PFAS-based polymers in relation to washing. It is possible that the total quantities released in the life cycle of the clothing are significantly greater than about 0.04 % of the total fluorine content in the textiles which are available as unbound, extractable PFASs.

Environmental exposure via air - PFASs emitted to air can travel long distances. It is primarily the volatile PFASs, such as FTOHs, EtFOSE and MeFOSE that are distributed via the air. Of these, it would be FTOHs in particular which could be released from the clothes. The calculations show that even in the worst case, the release of FTOHs from children's clothing in use is unlikely to be a major source of FTOHs to outdoor air. The potential for releases of PFASs by the decomposition of impregnating agents in landfills are potentially greater and outdoor wear may be a major source of the demonstrated emission of PFASs from landfills in other countries. In Denmark, clothes (not reused) are incinerated at waste incineration plants where the PFASs are destroyed, and releases from the disposal phase would therefore be negligible.

The environmental exposure in the soil environment - PFASs in sewage sludge applied to farmland result in concentrations in the soil estimated to be far below the level at which ecotoxicological effects would occur in the soil environment.

8. Conclusions

Prevalence of PFASs in children's wear - The study shows that PFAS-based impregnation agents are used for various outdoor clothing for children and occur in 10-30 % of all snowsuits. The results show that the PFAS-based agents to a large extent are based on C_8 chemistry. Recent surveys of adult clothes indicate that there has been a shift from products based on C_8 chemistry to materials based on C_{4-6}-chemistry, but such a change is not seen for the children's clothes examined. On the other hand, many of the major manufacturers have completely replaced the PFAS-based waterproofing agents for children's clothing. The alternatives make the garment water resistant, but to a lesser extent oil repellent (as evidenced by the claims) compared to the PFAS-based agents. The survey shows that alternatives are available and that they are used by major market players, which must consider that the alternatives provide an acceptable degree of proofing of the clothes.

Polymeric and non-polymeric PFASs - The study shows that the non-polymeric PFASs account only for about 0.04 % (average) of the fluorine content of textile. The remaining part is supposed to be present in the polymers in the form of polyfluoroalkyl-based side chains. There is considerable uncertainty about the extent to which these side chains can be split off from the polymers in use, washing and disposal of the clothes. The total amounts of PFASs, given in the course of the product life cycle, may be considerably higher than those that can be immediately extracted from the products. Foreign studies have shown that large quantities of non-polymeric PFASs may be released through the oxidation of the impregnating agents.

Composition of PFASs – Overall, 22 products were analysed for total fluorine; of these 19 contained fluoride above the detection limit. Of these, 15 products were selected for analysis of PFASs. The products were primarily selected from the products in which were found the highest concentrations of total fluorine. The results showed that FTOHs and PFCAs accounted for most of the total content of PFASs, with the C_8 compounds PFOA and 8:2 FTOH being predominant. Although the number of products is modest, this is deemed unlikely to result in significant uncertainties in the conclusions, as the results of the present study are well in line with the findings of other studies of PFASs in outdoor wear.

Migration to saliva - Analyses of migration to the saliva showed that a relatively small proportion of the PFCs content was released to the artificial saliva during the migration period. There is some uncertainty as to whether the simulant that does not contain enzymes fully simulates the effect of real saliva. However, in the context of the conclusions that are later drawn on the basis of measurements, it is assessed that this uncertainty is not important as the risk characterization ratio (RCR) of exposure via saliva is calculated to be well below 1.

Wash - Analyses of releases of PFASs under simulated washing conditions were similar to the analyses of the migration to saliva, in that it was a relatively small part of the textile's extractable content of PFASs released to the laundry water. This finding is different from the results of foreign studies where it was found that the release by wash exceeded the extractable content. As is it is not possible to judge which of the studies best reflects the actual situation, the "worst case" exposure scenarios were based on the foreign studies that showed the highest release of PFASs. The uncertainty associated with the quantities released by washing does not affect the conclusions in relation to the immediate effect of discharged PFASs in the aquatic environment. However, they affect the

assessment of the extent to which textiles may contribute to the overall emissions of PFASs to the environment.

Health - On the basis of the health assessment, it is concluded that the direct exposure to skin and saliva from the use of outerwear is a negligible contribution to the exposure of children and a negligible health risk. This conclusion is so robust that the uncertainties in the data do not affect the conclusions significantly. On the other hand, the estimated exposure via indoor air may potentially be a significant source of exposure for children. Therefore, it cannot be ruled out that the overall exposure of PFASs in the clothes may be of public health significance. Due to uncertainties in the data used, no firm conclusions can be drawn, and it is therefore proposed to investigate the problem further by actual measurements in rooms used by many children.

Environment - Discharges via wastewater treatment plants assessed do not immediately give rise to effects on aquatic life in the recipients. There is uncertainty about the possible endocrine disrupting effects as data are sparse. The main effects of PFASs in the environment, however, are effects on predators higher in food webs. There is some uncertainty about the extent to which PFASs from textiles discharged with the laundry water or released directly to the atmosphere may contribute to the overall emissions of these substances. "Worst case" calculations indicate that especially the direct discharges of PFASs to the atmosphere may contribute significantly, but there is limited knowledge about the quantities emitted during the product's entire lifecycle. If it is assumed that the amount emitted to the atmosphere corresponds to the extractable content, the total contribution would be modest. But it is possible that far larger amounts of non-polymeric PFASs are emitted shortly after the production of textiles, and it is also possible that later on in the product's useful life, substantial amounts of PFASs could be released from hydrolysis of the polyfluorinated side chains. Without this knowledge it is difficult to assess how much the textiles could contribute to these issues. Swiss studies suggest that use of PFASs in textiles may well be among the largest sources of emissions of PFASs in Switzerland.

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Appendix 1: Substances and substance groups mentioned in the report organised by abbreviation

The table below indicates the chemical name, CAS number, chain length, group name and group-abbreviation for all substances listed in the report.

The names and abbreviations of substances and groups are based on the recommended nomenclature in Buck *et al.* (2011). Substances that are analysed in this study are marked with an asterisk.

Note that the specified chain length is not in all cases the length of the perfluorinated carbon chain substance. The carboxylic acids will be in the chain being a carbon atom that is not perfluorinated. For instance, PFOA has a carbon chain of 8 carbon atoms and is usually expressed as C8 technology, but it is only 7 carbon atoms that are perfluorinated. The length of the perfluorinated chain is indicated in parentheses. For the telomers, the chain length indicated by the length of the perfluorinated part, and there will therefore be two C atoms which are not fluorinated. 6:2 FTOH is therefore consistently with common practice in the literature described as C_6, although the total chain length is 8 C atoms. This notation is used on chain length to make it easier to see which substances can act as precursors for others.

For acids the same abbreviations are used for both the acid form and acid anion, for example PFOS is used for both perfluorooctane sulfonic acid and the perfluorooctane sulfonate anion. In the following table only the acid form is indicated.

Abbreviation	Chemical name (according to Buck *et al.*, 2014)	CAS No.	Chain length **	Group name	Group abbreviation
4:2 FTAC	4:2 Fluorotelomer acrylate	52591-27-2	C4	n:2 Fluorotelomer acrylates	n:2 FTACs
4:2 FTOH *	4:2 Fluorotelomer alcohol	2043-47-2	C4	n:2 Fluorotelomer alcohols	n:2 FTOHs
4:2 FTSA *	4:2 Fluorotelomer sulfonic acid	757124-72-4	C4	n:2 Fluorotelomer sulfonic acids	n:2 FTSAs
6:2 diPAP	6:2 Fluorotelomer phosphate diester	57677-95-9	C6	Fluorotelomer phosphate diesters	diPAP
6:2 FTAC *	6:2 Fluorotelomer acrylate	65104-64-5	C6	n:2 Fluorotelomer acrylates	n:2 FTACs
6:2 FTCA *	6:2 Fluorotelomer carboxylic acid	53826-12-3	C6	Fluorotelomer (saturated) carboxylic acids	n:2 FTCAs
6:2 FTI	6:2 Fluorotelomer iodide	2043-57-4	C6	n:2 Fluorotelomer iodides	FTI
6:2 FTMAC	6:2 Fluorotelomer methacrylate	2144-53-8	C6	n:2 Fluorotelomer methacrylates	n:2 FTMACs
6:2 FTO	6:2 Fluorotelomer olefin	25291-17-2	C6	n:2 Fluorotelomer olefins	n:2 FTO
6:2 FTOH *	6:2 Fluorotelomer alcohol	647-42-7	C6	n:2 Fluorotelomer alcohols	n:2 FTOHs
6:2 FTSA *	6:2 Fluorotelomer sulfonic acid	27619-97-2	C6	n:2 Fluorotelomer sulfonic acids	n:2 FTSAs
6:2 FTUCA	6:2 Fluorotelomer carboxylic acid, unsaturated	70887-88-6	C6 (C5)	6:2 Fluorotelomer unsaturated carboxylic acids	FTUCA
8:2 diPAP	8:2 Fluorotelomer phosphate diesters	678-41-1 8	C8	Fluorotelomer phosphate diesters	diPAP

Abbreviation	Chemical name (according to Buck *et al.*, 2014)	CAS No.	Chain length **	Group name	Group abbreviation
8:2 FTAC *	8:2 Fluorotelomer acrylate	27905-45-9	C8	n:2 Fluorotelomer acrylates	n:2 FTAC
8:2 FTCA *	8:2 Fluorotelomer carboxylic acid	27854-31-5	C8	n:2 Fluorotelomer (saturated) carboxylic acids	n:2 FTCAs
8:2 FTMAC *	8:2 Fluorotelomer methacrylate	1996-88-9	C8	n:2 Fluorotelomer methacrylates	n:2 FTMACs
8:2 FTOH *	8:2 Fluorotelomer alcohol	678-39-7	C8	n:2 Fluorotelomer alcohols	n:2 FTOHs
8:2 FTSA *	8:2 Fluorotelomer sulfonic acid	39108-34-4	C8	n:2 Fluorotelomer sulfonic acids	n:2 FTSAs
8:2 FTUCA	8:2 Fluorotelomer carboxylic acid, unsaturated	70887-84-2	C8 (C7)	6:2 Fluorotelomer carboxylic acids, unsaturated	FTUCAs
10:2 FTAC *	10:2 Fluorotelomer acrylate	17741-60-5	C10	n:2 Fluorotelomer acrylates	n:2 FTACs
10:2 FTCA *	10:2 Fluorotelomer carboxylic acid	53826-13-4	C10	n:2 Fluorotelomer carboxylic acids, saturated	n:2 FTCAs
10:2 FTMAC *	10:2 Fluorotelomer methacrylate	2144-54-9	C10	n:2 Fluorotelomer methacrylates	n:2 FTMACs
10:2 FTOH *	10:2 Fluorotelomer alcohol	865-86-1	C10	n:2 Fluorotelomer alcohols	n:2 FTOHs
10:2 monoPAP	10:2 Fluorotelomer phosphate monoester	57678-05-4	C10	n:2 Polyfluoroalkyl phosphoric acid monoesters	monoPAP
APFN	Ammonium perfluorononanoate	4149-60-4	C9 (C8)	Perfluoroalkyl carboxylic acids and salts	PFCAs
APFO	Ammonium perfluorooctanoate	3825-26-1	C8 (C7)	Perfluoroalkyl carboxylic acids and salts	PFCAs
C8-PFPA	Perfluorooctyl phosphonic acid	40143-78-0	C8	Perfluorooctyl phosphonic acids	PFPAs
EtFASAs	*N*-Ethyl perfluoroalkane sulfonamides	Group	-	-	-
EtFASEs	*N*-Ethyl perfluoroalkane sulfonamidoethanols	Group	-	-	-
EtFOSA *	*N*-Ethyl-perfluorooctane sulfonamide	4151-50-2	C8	*N*-Ethyl perfluoralkane sulfonamides	EtFASAs
EtFOSE *	*N*-Ethyl-perfluorooctane sulfonamidethanol	1691-99-2	C8	*N*-Ethyl perfluoroalkane sulfonamidoethanols	EtFASEs
FASAs	Perfluoroalkane sulfonamides	Group	-	-	-
FOSA *	Perfluorooctane sulfonamide	754-91-6	C8	Perfluoroalkane sulfonamides	FASAs
FTACs	Fluorotelomer acrylates	Group	-	-	-
FTCAs	Fluorotelomer carboxylic acids	Group	-	-	-
FTMACs	Fluorotelomer methacrylates	Group	-	-	-
FTOs	Fluorotelomer olefins	Group	-	-	-
FTOHs	Fluorotelomer alcohols	Group	-	-	-
FTSAs	Fluorotelomer sulfonic acids	Group	-	-	-

Abbreviation	Chemical name (according to Buck *et al.*, 2014)	CAS No.	Chain length **	Group name	Group abbreviation
MeFASAs	*N*-Methyl perfluoroalkane sulfonamides	Group	-	-	-
MeFASEs	*N*-Methyl perfluoroalkane sulfonamidoethanols	Group	-	-	-
MeFBSE	*N*-Methylperfluorobutane sulfonamidethanol	34454-97-2	C4	*N*-Methyl perfluoroalkane sulfonamidoethanols	MeFASEs
MeFOSA *	*N*-Methylperfluoroctane sulfonamide	31506-32-8	C8	*N*-Methyl perfluoroalkane sulfonamides	MeFASAs
MeFOSE *	*N*-Methyl perfluorooctane sulfonamidoethanol	24448-09-7	C8	*N-Methyl perfluoroalkane sulfonamides*	MeFASEs
Na-PFOA	Sodium perfluorooctanoate	335-95-5	C8 (C7)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PASFs	Perfluoroalkane sulfonyl fluorides	Group			
PBSF	Perfluorobutane sulfonyl fluoride	375-72-4	C4		PASFs
PFASs	Polyfluoroalkyl substances	Group	-	-	-
PFBA	Perfluorobutanoic acid	375-22-4	C4 (C3)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFBI	Perfluorobutyl iodide	423-39-2	C4	Perfluoralkyl iodides	PFAI
PFBS *	Perfluorobutane sulfonic acid	375-73-5	C4	Perfluoroalkane sulfonic acids	PFSAs
PFCAs	Perfluoroalkyl carboxylic acids	Group	-	-	-
PFDA *	Perfluorodecanoic acid	335-76-2	C10 (C9)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFDoDA *	Perfluorododecanoic acid	307-55-1	C12 (C11)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFDS *	Perfluorodecane sulfonic acid	335-77-3	C10	Perfluoroalkane sulfonic acids	PFSAs
PFHpA *	Perfluoroheptanoic acid	375-85-9	C7 (C6)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFHpS *	Perfluoroheptane sulfonic acid	375-92-8	C7	Perfluoroalkane sulfonic acids	PFSAs
PFHxA *	Perfluorohexanoic acid	307-24-4	C6 (C5)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFHxDA	Perfluorohexadecanoic acid	67905-19-5	C16 (C15)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFHxS *	Perfluorohexane sulfonic acid	355-46-4	C6	Perfluoroalkane sulfonic acids	PFSAs
PFNA *	Perfluorononanoic acid	375-95-1	C9 (C8)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFOA *	Perfluorooctanoic acid	335-67-1	C8 (C7)	Perfluoroalkyl carboxylic acids and salts	PFCAs

Abbreviation	Chemical name (according to Buck *et al.*, 2014)	CAS No.	Chain length **	Group name	Group abbreviation
PFOS *	Perfluorooctane sulfonic acid	1763-23-1	C8	Perfluoroalkane sulfonic acids	PFSAs
PFO-TeAA	REACH registration:2-Propenoic acid, γ-ω-perfluor-C8-14-alkyl esters (PFO-TeAA) is a multi constituent substance consisting of alkyl esters with chain lengths of C8-C14. It can therefore also be declared as reaction mass of 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,10-heptadecafluorodecyl acrylate and 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,12,12,12-henicosafluorododecyl acrylate and 3,3,4,4,5,5,6,6,7,7,8,8,9,9,10,10,11,11,12,12,13,13,14,14,14-pentacosafluorotetradecyl acrylate.	85631-54-5	C8-C14	Fluorotelomer acrylates	No group abb.
PFPA *	Perfluoropentanoic acid	2706-90-3	C5 (C4)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFPeS *	Perfluoropentane sulfonic acid	2706-91-4	C5	Perfluoroalkane sulfonic acids	PFSAs
PFSAs	Perfluoroalkane sulfonic acids	Group	-	-	-
PFTeDA *	Perfluorotetradecanoic acid	376-06-7	C14 (C13)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFTrDA *	Perfluorotridecanoic acid	72629-94-8	C13 (C12)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFUnDA *	Perfluoroundecanoic acid	4234-23-5	C11 (C10)	Perfluoroalkyl carboxylic acids and salts	PFCAs
PFAAs	Perfluoroalkyl acids	Group	-	-	-
POSF	Perfluorooctane sulfonyl fluoride	307-35-7	C8	Perfluoroalkane sulfonyl fluorides	PASF

* Substances marked with an asterisk are included in the analysis program.
** For the carboxylic acids, the length of the perfluorinated chain length is indicated in parentheses.

Appendix 2: Other abbreviations and acronyms used in the report

ADHD	Attention deficit hyperactivity disorder
BCF	Bioconcentration factor
BfR	Bundesinstitut für Risikobewertung
BMF	Biomagnification factor
CAS	Chemical Abstract Service (chemical classification system)
CLP	Classification, Labelling and Packaging (Regulation (EC) No 1272/2008)
CMR	Carcinogenic, mutagenic, or toxic to reproduction
DEPA	Danish Environmental Protection Agency
EC_{50}	Concentration, where there is an effect on 50 % of the test organisms
ECHA	European Chemicals Agency
EC	European Community
EFSA	European Food Safety Authority
ESI	Negative electrospray ionisation
EU	European Union
GC	Gas chromatograph/Gas chromatography
HDL	Low-density lipoprotein
IARC	International Agency for Research on Cancer
JRC	Joint Research Centre
LC_{50}	Concentration, at which 50 % of the test organisms die
LDL	Low-density lipoprotein
LOUS	List of Undesirable Substances (of the Danish EPA)
MS	Mass spectrometry
MTBE	Methyl-*tert*-butyl ether
NHANES	National Health and Nutrition Examination Survey
NOAEL	No Observed Adverse Effect Level
NOEC	No Observed Effect Concentration
PBT	Persistent, bioaccumulative and toxic
PCI	Positive chemical ionisation
PE	Polyethylene
PE	Person Equivalent
PEC	Predicted Environmental Concentration
PNEC	Predicted No-effect Concentration
POP	Persistent organic pollutant
PTFE	Polytetrafluoroethylene
RCR	Risk Characterisation Ratio
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals (Regulation (EC) no. 1907/2006)
ROS	Reactive oxygen substances
TTR	Transthyretin
USA	United States of America
vPvB	Very persistent and very bioaccumulative

Appendix 3: Questionnaire to companies

Questions concerning the use of perfluorinated substances in textile products for children on the Danish market.

COWI and partners are preparing a survey of the use of perfluorinated substances in children's textile products on the Danish market for the Danish EPA. As a part of the project, companies are asked to support the survey by providing information on the topic. Please consider the following questions, which will presumably take 10 – 30 min to fill in depending on the amount of information and knowledge available.

Thank you very much in advance!

Your company:

Contact person:

Email:

Phone:

1. Which kind of **children's clothing** with water- and/or dirt-repelling properties do you sell? Are there any product brands within the group, which are branded/labelled as containing fluorine-based technology? If yes, please indicate the brand name(s) of the garment. Please tick both YES and NO if some products in a category contain the substance while others do not.

Garment in the assortment	*With fluorine technology for water and/or soil repellency?*			
	YES	NO	Don't know	If yes, which main brand(s)
☐ Snowsuits/cold weather suits (Danish: flyverdragter)	☐	☐	☐	
☐ Outdoor jackets and pants	☐	☐	☐	
☐ Rainwear	☐	☐	☐	
☐ Skiwear	☐	☐	☐	
☐ Gloves	☐	☐	☐	
☐ Bibs and pinafores	☐	☐	☐	
☐ Sports clothing	☐	☐	☐	
☐ Boots	☐	☐	☐	
☐ Other (please indicate):	☐	☐	☐	

2. Which kind of **childcare products** with water- and/or dirt-repelling properties do you sell? Are there any product brands within the group, which are branded/labelled as containing fluorine-based technology? If yes, please indicate the brand name(s) of the garment. Please tick both YES and NO if some products in a category contain the substance while others do not.

Childcare products	*With fluorine technology?*			
	YES	NO	Don't know	If yes, which brand(s)
☐ Car seats	☐	☐	☐	
☐ Buggies	☐	☐	☐	
☐ Cradle seats	☐	☐	☐	
☐ Carriers and baby slings	☐	☐	☐	
☐ Changing mats	☐	☐	☐	
☐ Other (please indicate):	☐	☐	☐	

3. Which kind of **accessories for children** with water- and/or dirt-repelling properties do you sell?

Accessories for children	*With fluorine technology?*			
	YES	NO	Don't know	If yes, which brand(s)
☐ Bags and backpacks	☐	☐	☐	
☐ Pencil cases	☐	☐	☐	
☐ Other (please indicate):	☐	☐	☐	

4. Textile products may be marked with certain trade names indicating the technology used for water and dirt repellency?
Are any of your children products branded with any of the following attributes?

☐ Teflon®
☐ Scotchgard®
☐ Unidyne®
☐ NanoTex®
☐ GreenShield®
☐ Lurotex® Duo System
☐ Crypton®
☐ Other (please indicate trade name):

5. Does your company have any restrictions of perfluorinated substances in textiles going beyond the general EU restriction on perfluorooctane sulfonates (PFOS) and related substances (the first five are covered by Oeko-Tex 100)?

☐Perfluorooctanoic acid (PFOA), concentration limit:
☐Perfluorotridecanoic acid(PFTrDA), concentration limit:
☐Perfluorododecanoic acid (PFDoDA), concentration limit:
☐Perfluoroundecanoic acid (PFUnDA), concentration limit:
☐Perfluorotetradecanoic acid (PTFEDA), concentration limit:
☐C8-based fluorotechnology, concentration limit:
☐Fluoroalkyl substances of any kind, concentration limit:
☐Other (please indicate substances):
☐Don't know

6. Do you know any of the fluorine compounds used to provide water and dirt repellency? Please indicate chemical name and/or CAS nr.

7. In which concentrations might the fluorinated substances be present in the finished textiles?

8. Compared to the total volume of sold outdoor textile products for children, how large is the turnover of products with water- and dirt-repelling properties? (We are aware of that it is largely impossible to provide a precise estimate on that, please provide a "qualified guess" if possible). Please choose at least 1 of the three quantification possibilities:

Weight in tons:	Number of articles:	Value of articles (DKK or EUR):
☐below 10 %	☐below 10 %	☐below 10 %
☐10 – 25 %	☐10 – 25 %	☐10 – 25 %
☐25 – 50 %	☐25 – 50 %	☐25 – 50 %
☐50 – 75 %	☐50 – 75 %	☐50 – 75 %
☐more than 75 %	☐more than 75 %	☐more than 75 %

Appendix 4: Fluorinated impregnation agents marketed for impregnation of fabrics.

Knowledge Centre for Smart Textiles at Creative Business Centre in 2012 prepared a list containing a number of surface treatment agents for textiles, including agents based on co-polymers with perfluorinated side chains (VIT, 2013). Information on PFAS-based agents is summarised in the table below where the information from the VIT survey is supplemented with information on other surface treatment agents obtained from the Internet.

Producer	Brand name/ product name	Fluorinated compounds (as indicated by the producer)	Application (as indicated by the producer)	Source
DuPont /Huntsman	Advanced Dual Action Teflon® fabric protector. Marketed via Huntsman as Oleophobol CP.	Fluoroalkyl acrylate copolymer dispersion.	Traditional textile finishing process (application to foulard - drying - curing). Teflon® fabric protector designation can only be achieved if the dye factory has a license from DuPont. Swipe	VIT, 2012
	Repel Teflon® fabric protector High performance Repel Teflon® fabric protector Advanced Dual Action Teflon® fabric protector Tri-Effects Teflon® fabric protector Marketed via Huntsman as Oleophobol CP®.	Fluoroalkyl acrylate copolymer dispersion.	Traditional textile finishing process (application to foulard - drying - curing). Teflon® fabric protector designation can only be achieved if the dye factory has a license from DuPont. Stain resistant. Tri-Effects Teflon® while also providing a moisture absorbing function on the inside	VIT, 2012
	Release Teflon® High Performance Release Teflon® Ultra Release Teflon®	Fluoroalkyl acrylate copolymer dispersion.	Traditional textile finishing process (application to foulard - drying - curing). Teflon® fabric protector designation can only be achieved if the dye factory has a license from DuPont. Dirt-repelling function	VIT, 2012

Producer	Brand name/ product name	Fluorinated compounds (as indicated by the producer)	Application (as indicated by the producer)	Source
			Ultra Release Teflon® fabric protector also prevents re-precipitation of dirt during washing	
BigSky Technologies LLC	GreenShield®	Fluoroalkyl acrylate and alkyl acrylate copolymers Supposedly environmentally friendly by 8-10 times less use of fluorocarbon.	Suitable for cotton, polyester, nylon, polypropylene and viscose. Supposedly permanent washing resistance (25 washes). Improved grip compared to conventional agents. Water-based formulation - traditional process (application, drying, curing). Dirt repellent Water and oil repellent	VIT, 2012
BASF	Lurotex Protector RL ECO ®	Fluorocarbon finishing with C_6-teknologi together with Perapret Booster XLR.	Traditional textile finishing process (application to foulard - drying - curing). Can be used on cotton and synthetic items. Good was resistance and soft grip. Reduces the need for washing. For clothing and home textiles. Stain is easier to wash out.	VIT, 2012
Pulcra Chemicals	Repellan KFC®	Perfluoroalkyl acryl polymer.	Can be used for materials of cellulose, synthetic and wool and blends. For clothing. Apply in conventional textile machinery. Does not affect grip and breathability. Improves the durability and service life. Stain resistant.	VIT, 2012
Rudolf Group	Rucostar® EEE6	Dendrimers of fluorocarbon, which reduce the amount of fluorocarbon by 50 % compared to conventional finishing with a better effect. Solvent free	Water repellent	VIT, 2012

Producer	Brand name/ product name	Fluorinated compounds (as indicated by the producer)	Application (as indicated by the producer)	Source
Daikin	Unidyne™	Fluoroalkyl acrylate copolymer	Unidyne is used for a number of different applications in various industries such as textiles, carpet, automotive, and medical device industries. Water and oil repellent.	Daikin, 2014
Rudolf Group	Bionic Finish®	By the use of starlike, branched polymers, water and oil-repellent effects with a simultaneously reduced fluorocarbon resin content can be obtained The fluorocarbon resins included are not specified	Water and dirt repellent impregnation of surfaces Note that Bionic Finish® Eco does not contain perfluorinated groups (Rudolf Group, 2014b)	Rudolf Group, 2014a
	RUCO-GUARD®	Water or solvent-based fluorocarbon polymers, fluorocarbon resins or boosters Based on C6-based fluorocarbon polymeric dispersions	For water, oil and soil-repellent impregnations of surfaces	Rudolf Group, 2014a
	RUCOSTAR® RUCO-COAT® RUCO-PROTECT® RUCOTEC® RUCO®	Water or solvent-based fluorocarbon products (some of the brands also available as fluorocarbon free) Based on C6-based fluorocarbon polymeric dispersions	For water, oil and soil-repellent impregnations of surfaces with reduced fluorocarbon portion	Rudolf Group, 2014a
Nano-Tex	Resist Spills™	C6-based "PFOA free" repellency solution. Each fibre has been fundamentally trans-formed through nanotechnology.	Repels liquids Outperforms conventional fabric treatments Provides long lasting protection Extends the life of the fabric Retains fabric's natural softness Allows fabric to breathe naturally	Nanotex, 2014

Producer	Brand name/ product name	Fluorinated compounds (as indicated by the producer)	Application (as indicated by the producer)	Source
			For apparel, home textiles, residential furniture, commercial interiors.	
	Resists Spills and Releases Stains™	C6-based "PFOA-free" dual action product. Each fibre has been fundamentally trans-formed through nanotechnology.	Repels spills Helps stains wash out easily Provides long lasting protection Extends the life of the fabric Retains fabric's natural softness Allows fabric to breathe naturally. For apparel and home textiles.	Nanotex, 2014
3M	Scotchgard™ Fabric Protector	Fluorochemical Urethane <3 %, CAS No confidential	Strong protection ensures that stains are easily removed Do not change the look, feel and breathability of textiles Safe to use on delicate fabrics like silk and wool	3M, 2014
Nicca	NK Guard S series	Fluorocarbon based water and oil repellent	PFOA-free water and oil repellent, gentle to environment (below detection level of 5 ppb) Does not contain formaldehyde and its derivatives Offers durable water and oil repellency for polyester, nylon and cotton Offers very soft hand to various type of fabrics	Nicca, 2014

Appendix 5: Perfluorinated and polyfluorinated intermediates/raw materials manufactured by Dupont

Brand name/ product name	Fluorinated compounds (as indicated by Dupont)	Application
Capstone™ 62-I	Perfluorohexyl ethyl iodide 85.0 % min (CAS No 2043-57-4; 6:2 FTI) Perfluorobutyl ethyl iodide 5.0 % max Perfluorooctyl ethyl iodide 2.0–10.0 % Perfluorodecyl ethyl iodide 6.0 % max Other perfluoroalkylethyl iodides 1.0 % max	Surfactants for dry-cleaning and fabric soil-proofing (US5610128) Urethanes for water- and oil-repellents (WO2006013791) Repellent polymer and surface treating agent (WO2005092937) Water and oil proof phosphate resin (JP2003096311) Water- and oil-repellent fluorine-modified silicones and their manufacture (JP09227685) Phosphonic acids useful as treatments for substrate surfaces (US6824882) Fluorosilicone resins (JP 3364355) Polyurethanes as soil-release agents (US3759874)
Capstone® 4-I	Perfluorobutyl iodide 90 % min (CAS No 423-39-2; PFBI) Perfluoroethyl iodide 2.0 % max Perfluorohexyl iodide 1.0 % max Perfluorohexane 8.5 % max Perfluorooctane 1.0 % max	Arylguanamine derivatives as water and oil repellents for textiles (JP03145477) Perfluoroalkylphenol stain resists for synthetic polymer fibres (WO2002055464) Fluoroalkyl triazines as water repellent (US6391948) Urethanes for water- and oil-repellents (WO006013791) Fluorinated polyurethanes as soil-release finishes for textiles (US3872058)
Capstone™ 62-AL	3,3,4,4,5,5,6,6,7,7,8,8,8-Tridecafluoro-1-octanol (CAS no. 647-42-7; 6:2 FTOH)	Not specified (assumd to be used for other intermediates)
Capstone™ 62-MA	2-Methyl-2-propenoic acid, 3,3,4,4,5,5,6,6,7,7,8,8,8-tridecafluorooctyl ester (CAS No 2144-53-8; 6:2 Fluorotelomer methacrylate)	Not specified DuPont™ Capstone™ 62-MA intermediate is a partially fluorinated methacrylate monomer, derived from a linear 6:2-FTOH

Appendix 6: Detection limits for PFASs in the different matrices

Substance	Detection limits Textile (µg/m²)	Detection limits Synthetic saliva (ng/L)	Detection limits Laundry water (ng/L)
Perfluoroalkane sulfonic acids (PFSAs)			
PFBS	0,001	0,54	1,1
PFPeS	0,001	*	*
PFHxS	0,002	0,98	0,97
PFHpS	0,001	0,72	2,38
PFOS	0,001	0,50	0,52
PFDS	0,002	0,68	0,25
Perfluoroalkane sulfonamides and - amidethanols			
PFOSA	0,006	2,5	0,91
MeFOSA	0,06	24	3,2
EtFOSA	0,06	24	3,3
MeFOSE	0,6	240	35,4
EtFOSE	0,3	120	16,0
Perfluoroalkyl carboxylic acids (PFCAs)			
PFPA	0,001	0,77	*
PFHxA	0,001	0,18	0,80
PFHpA	0,001	0,27	1,2
PFOA	0,001	0,51	2,2
PFNA	0,001	0,42	0,35
PFDA	0,001	0,23	0,71
PFUnDA	0,001	0,35	3,3
PFDoDA	0,002	0,87	0,59
PFTrDA	0,005	2,5	2,4
PFTeDA	0,007	3,2	0,37
Fluorotelomer alcohols (FTOHs)			
4:2 FTOH	1,0	400	*
6:2 FTOH	0,6	240	*
8:2 FTOH	0,5	200	44,1

Substance	Detection limits Textile (µg/m²)	Detection limits Synthetic saliva (ng/L)	Detection limits Laundry water (ng/L)
10:2 FTOH	0,09	36	6,3
Fluorotelomer carboxylic acids (FTCAs)			
6:2 FTCA	0,006	2,8	4,9
8:2 FTCA	0,009	3,9	5,7
10:2 FTCA	0,02	8,1	13,9
Fluorotelomer methacrylates (FTMACs)			
8:2 FTMAC	0,5	200	61,5
10:2 FTMAC	0,5	200	40,3
Fluorotelomer acrylates (FTACs)			
4:2 FTAC	1,0	400	*
6:2 FTAC	0,8	320	*
8:2 FTAC	0,6	240	61,5
10:2 FTAC	0,5	200	40,3
Fluorotelomer sulfonic acids (FTSAs)			
4:2 FTSA	0,001	0,12	*
6:2 FTSA	0,002	0,92	0,07
8:2 FTSA	0,001	0,25	0,50

Appendix 7: Recovery rates by analysis of PFASs in different matrices

Substance	Recovery (±RSD %) Textile	Recovery (±RSD %) Synthetic saliva	Recovery (±RSD %) Laundry water
Perfluoroalkane sulfonic acids (PFSAs)			
PFBS	69 (± 12)	57 (± 20)	27 (± 12)
PFPeS	4± 14)	105 (± 2)	*
PFHxS	96 (± 5)	97 (± 6)	9± 6)
PFHpS	117 (± 3)	94 (± 5)	1± 12)
PFOS	95 (±3)	10± 14)	1± 9)
PFDS	69 (±23)	7± 20)	19 (± 1)
Perfluoroalkane sulfonamides and amidethanols			
PFOSA	100 (± 6)	10± 7)	100 (± 5)
MeFOSA	85 (± 3)	99 (± 3)	94 (± 1)
EtFOSA	86 (± 2)	10± 5)	9± 1)
MeFOSE	89 (± 3)	99 (± 2)	85 (± 1)
EtFOSE	79 (± 4)	97 (± 2)	94 (±1)
Perfluoroalkyl carboxylic acids (PFCAs)			
PFPA	77 (± 15)	49 (± 21)	*
PFHxA	90 (± 14)	100 (± 3)	100 (± 1)
PFHpA	120 (± 11)	11± 6)	17 (± 9)
PFOA	10±12)	10± 6)	100 (± 3)
PFNA	108 (± 6)	99 (± 6)	97 (± 2)
PFDA	96 (± 11)	99 (± 5)	95 (± 5)
PFUnDA	98 (± 4)	10± 6)	89 (± 10)
PFDoDA	11± 14)	10± 6)	94 (± 3)
PFTrDA	10± 23)	75 (± 32)	98 (± 7)
PFTeDA	117 (± 15)	104 (± 10)	4± 16)
Fluorotelomer alcohols (FTOHs)			
4:2 FTOH	9± 9)	11± 2)	*
6:2 FTOH	10± 16)	90 (± 32)	*
8:2 FTOH	88 (± 5)	12±10)	57 (± 24)

Substance	Recovery (±RSD %) Textile	Recovery (±RSD %) Synthetic saliva	Recovery (±RSD %) Laundry water
10:2 FTOH	96 (± 10)	99 (± 3)	7± 10)
Fluorotelomer carboxylic acids (FTCAs)			
6:2 FTCA	99 (± 6)	9± 4)	95 (± 3)
8:2 FTCA	100 (± 9)	95 (± 3)	84 (± 3)
10:2 FTCA	9± 4)	9± 6)	59 (± 23)
Fluorotelomer methacrylates (FTMACs)			
8:2 FTMAC	106 (± 14)	127 (± 17)	4± 30)
10:2 FTMAC	12± 3)	13± 16)	6± 21)
Fluorotelomer acrylates (FTACs)			
4:2 FTAC	124 (± 3)	135 (± 12)	*
6:2 FTAC	138 (± 5)	174 (± 2)	*
8:2 FTAC	98 (± 2)	12± 10)	3± 33)
10:2 FTAC	107 (± 10)	129 (± 7)	5± 26)
Fluorotelomer sulfonic acids (FTSAs)			
4:2 FTSA	9± 6)	84 (± 12)	*
6:2 FTSA	116 (± 5)	115 (± 4)	12± 4)
8:2 FTSA	106 (± 14)	99 (± 3)	26 (± 4)

Appendix 8: Concentration of PFASs in textiles for children in µg/m²

No	F1		F3		F4		F5		F6		F7		F9	
Product name	Snowsuit		Softshell-suit		Gloves		Infant sleeping bag		Infant sleeping bag		Infant sleeping bag		Rain jacket	
Category	PFAS technology		Water/stain-resistant		PFAS technology		Water/stain-resistant		Water and dirt repellent		Teflon®		Water/stain-resistant	
Total-F µg/m³ (average)	60,000		128,000		58,000		21,000		42,000		365,000		32,000	
Perfluoroalkane sulfonic acids (PFSAs)														
PFBS	nd	0.006	0.004	0.004	nd	nd	0.003	0.002	nd	nd	nd	nd	nd	nd
PFPeS	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PFHxS	0.005	0.004	nd	nd	0.026	0.023	0.010	nd	nd	nd	nd	nd	nd	nd
PFHpS	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PFOS	0.055	0.062	0.009	0.009	0.033	0.028	0.24	0.17	0.002	0.002	nd	nd	0.051	0.044
PFDS	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Sum	**0.1**	**0.1**	**0.0**	**0.0**	**0.1**	**0.1**	**0.3**	**0.2**	**0.0**	**0.0**	**nd**	**nd**	**0.1**	**0.0**
Perfluoroalkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)														
FOSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.006	0.005
MeFOSA	0.011	0.002	0.008	0.005	0.007	nd	0.01	0.02	nd	0.008	nd	nd	nd	nd
EtFOSA	0.006	0.005	0.009	0.001	0.002	nd	0.001	0.01	nd	0.003	nd	nd	nd	nd
MeFOSE	0.03	0.02	0.03	0.04	nd	nd	nd	nd	nd	0.001	nd	nd	nd	nd
EtFOSE	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
Sum	**0.04**	**0.03**	**0.05**	**0.05**	**0.01**	**0.00**	**0.01**	**0.03**	**0.00**	**0.01**	**0.00**	**0.00**	**0.01**	**0.01**
Perfluoroalkyl carboxylic acids (PFCAs)														
PFPA	0.06	0.10	0.25	0.32	nd	0.014	0.085	0.058	nd	0.016	0.004	nd	nd	nd
PFHxA	0.84	1.13	2.92	3.59	0.22	0.19	0.27	0.22	0.02	0.030	0.17	0.12	0.047	0.044
PFHpA	0.77	1.21	1.90	2.54	0.022	0.016	0.18	0.14	0.016	0.021	0.064	0.026	0.031	0.031
PFOA	24.90	37.45	11.35	14.95	0.13	0.09	0.73	0.59	0.093	0.15	1.40	0.91	0.62	0.58
PFNA	0.31	0.78	2.32	3.62	0.023	0.017	0.14	0.12	0.019	0.11	0.063	0.038	3.62	3.20
PFDA	1.84	3.61	6.26	8.63	0.032	0.020	0.36	0.29	0.049	0.079	0.82	0.49	0.25	0.23

No	F1		F3		F4		F5		F6		F7		F9	
Product name	Snowsuit		Softshell-suit		Gloves		Infant sleeping bag		Infant sleeping bag		Infant sleeping bag		Rain jacket	
PFUnDA	0.11	0.29	0.77	1.18	0.001	0.003	0.066	0.056	0.001	0.026	0.038	0.047	1.16	0.95
PFDoDA	0.81	1.70	3.36	4.52	0.001	nd	0.15	0.11	0.006	0.013	0.56	0.37	0.025	0.022
PFTrDA	0.43	0.94	1.42	1.25	nd	nd	0.19	0.19	0.003	0.004	nd	nd	0.019	0.013
PFTeDA	4.69	8.65	9.77	8.86	0.024	0.014	1.21	0.87	0.48	0.66	0.018	0.21	0.079	0.068
Sum	**34.76**	**55.85**	**40.31**	**49.44**	**0.45**	**0.37**	**3.38**	**2.63**	**0.69**	**1.11**	**3.14**	**2.21**	**5.84**	**5.13**
Fluorotelomer carboxylic acids (FTCAs)														
6:2 FTCA	nd	nd	0.030	0.067	nd	nd	nd	nd	nd	nd	0.043	0.034	nd	nd
8:2 FTCA	0.22	0.23	0.13	0.16	nd	nd	0.053	0.051	0.065	0.12	1.33	1.03	0.31	0.33
10:2 FTCA	0.23	0.32	0.21	0.19	nd	nd	nd	nd	0.030	0.064	1.11	0.97	0.13	0.16
Sum	**0.45**	**0.55**	**0.38**	**0.41**	**nd**	**nd**	**0.05**	**0.05**	**0.10**	**0.18**	**2.48**	**2.03**	**0.44**	**0.49**
Fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs)														
8:2 FTMAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.37	nd	nd	nd
10:2 FTMAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.29	nd	nd	nd
4:2 FTAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.65	nd	nd	nd
6:2 FTAC	0.08	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.53	nd	nd	nd
8:2 FTAC	2.97	4.62	1.04	0.66	0.08	0.03	0.21	0.13	0.12	0.24	0.82	0.73	0.66	0.32
10:2 FTAC	1.00	1.44	0.28	0.24	nd	0.01	0.09	0.05	0.02	0.04	0.42	0.12	0.11	0.04
Sum	**4.05**	**6.06**	**1.32**	**0.90**	**0.08**	**0.05**	**0.30**	**0.18**	**0.14**	**0.28**	**3.08**	**0.85**	**0.77**	**0.36**
Fluorotelomer alcohols (FTOHs)														
4:2 FTOH	0.24	nd	0.07	nd	0.24	0.06	0.14	0.51	nd	nd	0.33	nd	nd	nd
6:2 FTOH	0.35	0.48	0.74	0.48	6.20	8.86	7.99	4.39	3.91	3.34	1.35	0.31	0.25	nd
8:2 FTOH	27.49	46.86	36.76	28.94	14.72	8.22	7.93	5.10	15.27	25.93	112.14	227.85	303.59	150.82
10:2 FTOH	10.18	13.86	9.94	0.67	4.10	2.39	2.14	1.52	3.68	6.41	25.63	39.50	58.43	33.12
Sum	**38.26**	**61.20**	**47.51**	**30.09**	**25.27**	**19.53**	**18.21**	**11.53**	**22.85**	**35.68**	**139.46**	**267.65**	**362.27**	**183.94**
Fluortelomersulfonates (FTSAs)														
4:2 FTSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.11	0.059	nd	nd
6:2 FTSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.58	0.25	nd	nd

No	F1		F3		F4		F5		F6		F7		F9	
Product name	Snowsuit		Softshell-suit		Gloves		Infant sleeping bag		Infant sleeping bag		Infant sleeping bag		Rain jacket	
8:2 FTSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	5.58	0.76	0.075	0.071
Sum	**nd**	**nd**	**nd**	**nd**	**nd**	**nd**	**nd**	**nd**	**nd**	**nd**	**6.27**	**1.07**	**0.08**	**0.07**
Sum PFASs	**77.63**	**123.75**	**89.58**	**80.90**	**25.87**	**20.00**	**22.20**	**14.60**	**23.78**	**37.26**	**154.42**	**273.80**	**369.45**	**190.03**

No	F10		F11		F15		F17		F18		F20		F21		F22	
Product name	Rain suit		Snow-suit		Snow-suit		Rain suit		Rain suit		Mittens		Infant sleeping bag		Mittens	
Category	Teflon®		PFAS technol-ogy		Water and dirt repellent		PFAS technol-ogy		PFAS technol-ogy		Teflon®		Water and dirt repellent		Water and dirt repellent	
Total-F µg/m³ aver-age)	115.000	32.000	149.000	19.000	27.000	193.000	11.000	49.000	115.000	32.000	149.000	19.000	27.000	193.000	11.000	
Perfluoroalkane sulfonic acids (PFSAs)																
PFBS	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PFPeS	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PFHxS	nd	nd	nd	nd	nd	nd	0.003	0.004	nd	nd	0.001	0.002	0.005	0.007	0.003	0.004
PFHpS	nd	nd	nd	nd	0.14	0.16	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
PFOS	nd	nd	nd	nd	0.054	0.067	nd	nd	0.001	0.002	0.006	0.007	0.003	0.002	0.02	0.02
PFDS	nd	nd	nd	nd	nd	nd	0.011	0.008	nd	nd	nd	nd	nd	nd	nd	nd
Sum	**nd**	**nd**	**nd**	**nd**	**0.2**	**0.2**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**	**0.0**
Perfluoroalkane sulfonamides and amidethanols (FASAs, MeFASAs, MeFASEs, EtFASAs, EtFASEs)																
FOSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
MeFOSA	nd	nd	nd	nd	0.007	0.007	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
EtFOSA	nd	nd	nd	nd	0.026	0.067	nd	nd	nd	nd	0.04	0.04	nd	nd	nd	nd
MeFOSE	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.06	0.04	nd	nd	nd	nd
EtFOSE	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.04	0.03	nd	nd	nd	nd

No	F10		F11		F15		F17		F18		F20		F21		F22	
Product name	Rain suit		Snow-suit		Snow-suit		Rain suit		Rain suit		Mittens		Infant sleeping bag		Mittens	
Sum	**0.00**	**0.00**	**0.00**	**0.00**	**0.03**	**0.07**	**0.00**	**0.00**	**0.00**	**0.00**	**0.14**	**0.11**	**0.00**	**0.00**	**0.00**	**0.00**
Perfluoroalkyl carboxylic acids (PFCAs)																
PFPA	0.021	0.016	nd	nd	0.11	0.12	0.03	0.007	0.012	0.02	0.011	0.006	0.004	0.012	0.03	0.04
PFHxA	0.22	0.22	0.003	0.004	0.75	0.70	0.01	0.01	0.13	0.14	0.08	0.06	0.005	0.004	0.50	0.58
PFHpA	0.034	0.049	0.005	0.004	0.44	0.31	0.007	0.007	0.03	0.04	0.02	0.02	0.006	0.005	0.16	0.24
PFOA	0.65	0.63	0.041	0.051	1.86	1.61	0.04	0.04	0.34	0.37	0.10	0.08	0.03	0.02	2.65	3.42
PFNA	0.040	0.037	0.084	0.093	0.73	0.54	0.002	0.002	0.03	0.03	0.04	0.04	0.03	0.03	0.15	0.27
PFDA	0.25	0.24	0.022	0.025	1.89	1.68	0.008	0.006	0.16	0.19	0.04	0.04	0.02	0.02	1.37	1.68
PFUnDA	0.016	0.014	0.027	0.029	0.21	0.13	0.001	0.001	0.01	0.01	0.007	0.008	0.01	0.01	0.06	0.10
PFDoDA	0.14	0.13	0.005	0.005	0.55	0.38	0.002	0.001	0.07	0.08	0.002	0.002	0.003	0.002	0.49	0.65
PFTrDA	0.039	0.012	0.006	0.009	0.089	0.056	0.001	0.002	0.008	0.008	0.002	0.002	0.004	0.002	0.06	0.09
PFTeDA	0.25	0.088	0.021	0.022	2.91	1.90	0.01	0.01	0.12	0.14	0.007	0.007	0.02	0.01	1.16	1.56
Sum	**1.66**	**1.43**	**0.21**	**0.24**	**9.53**	**7.42**	**0.11**	**0.08**	**0.91**	**1.02**	**0.31**	**0.26**	**0.14**	**0.12**	**6.62**	**8.63**
Fluorotelomer carboxylic acids (FTCAs)																
6:2 FTCA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.02	0.02	nd	nd	0.01	0.02
8:2 FTCA	0.074	0.063	0.10	0.11	0.19	0.18	0.03	0.03	0.04	0.03	0.07	0.11	0.06	0.05	0.04	0.14
10:2 FTCA	0.043	0.041	0.032	0.029	0.046	0.036	nd	nd	0.01	0.02	0.03	0.04	0.02	0.02	0.04	0.07
Sum	**0.12**	**0.10**	**0.14**	**0.14**	**0.24**	**0.22**	**0.03**	**0.03**	**0.05**	**0.05**	**0.11**	**0.17**	**0.08**	**0.07**	**0.10**	**0.23**
Fluorotelomer acrylates (FTACs) and fluorotelomer methacrylates (FTMACs)																
8:2 FTMAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
10:2 FTMAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
4:2 FTAC	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
6:2 FTAC	0.09	0.14	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
8:2 FTAC	4.34	5.28	0.91	0.63	0.75	0.48	0.45	0.50	0.24	0.24	2.92	2.52	0.83	0.60	0.82	1.37
10:2 FTAC	1.47	1.60	0.20	0.08	0.17	0.09	0.17	0.15	0.13	0.13	0.55	0.88	0.23	0.17	0.32	0.47
Sum	**5.90**	**7.02**	**1.11**	**0.71**	**0.92**	**0.57**	**0.62**	**0.65**	**0.37**	**0.37**	**3.47**	**3.40**	**1.06**	**0.77**	**1.14**	**1.84**

No	F10		F11		F15		F17		F18		F20		F21		F22	
Product name	Rain suit		Snow-suit		Snow-suit		Rain suit		Rain suit		Mittens		Infant sleeping bag		Mittens	
Fluorotelomer alcohols (FTOHs)																
4:2 FTOH	0.59	0.02	0.07	0.07	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
6:2 FTOH	0.13	0.83	0.40	nd	0.02	nd	0.50	0.39	0.65	0.24	93.93	72.17	3.46	1.90	1.33	2.20
8:2 FTOH	99.70	94.60	94.48	68.01	46.23	43.41	19.51	21.27	32.62	28.95	244.34	285.64	133.25	85.63	71.57	89.31
10:2 FTOH	22.65	21.07	20.84	13.28	13.49	15.04	5.72	5.38	7.49	6.57	48.50	60.58	26.69	18.52	15.40	18.69
Sum	**123.08**	**116.52**	**115.79**	**81.36**	**59.74**	**58.45**	**25.73**	**27.05**	**40.76**	**35.76**	**386.77**	**418.40**	**163.40**	**106.05**	**88.31**	**110.20**
Fluortelomersulfonater (FTSAs)																
4:2 FTSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
6:2 FTSA	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd
8:2 FTSA	0.023	0.022	0.030	0.024	0.026	0.023	0.02	0.014	0.02	0.02	0.03	0.02	0.03	0.02	0.02	0.03
Sum	**0.02**	**0.02**	**0.03**	**0.02**	**0.03**	**0.02**	**0.02**	**0.01**	**0.02**	**0.02**	**0.03**	**0.02**	**0.03**	**0.02**	**0.02**	**0.03**
Sum PFASs	**130.78**	**125.09**	**117.28**	**82.48**	**70.68**	**66.98**	**26.51**	**27.84**	**42.12**	**37.23**	**390.83**	**422.35**	**164.71**	**107.05**	**96.20**	**120.95**

Appendix 9: Overview of results of analyses of PFASs in previous studies identified during Phase 1 of the project

Fluorotelomer alcohols (FTOHs), concentration (µg/m²)									
Total FTOHs	4:2 FTOH	6:2 FTOH	8:2 FTOH	10:2 FTOH	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
<dg - 10,680					10/11	Ski and sporting clothing for children and adult		SFT, 2006	Most textiles have concentrations between 30 and 400 g/m², only a single jacket contained 10.7 mg/m². In 9 out of 11 products 8:2 FTOH was the substance found in the highest concentrations.
27.1 - 1,001					6/6	Jackets for all weather and anorak		Norges Naturvern-forbund, 2006	8:2 FTOH was the dominant FTOH in all products.
<dg – 464.2					8/14	Outdoor clothing for children and adult		Greenpeace, 2012	The finding that 6:2 FTOH was the dominating FTOH in more products (9 – 11, 12) indicates that more manufacturers have started to use this substance instead of 8:2 FTOH.
<dg	<dg	<dg	<dg	<dg		Greenpeace jacket	1	Greenpeace, 2012	China

Fluorotelomer alcohols (FTOHs), concentration (µg/m²)									
Total FTOHs	4:2 FTOH	6:2 FTOH	8:2 FTOH	10:2 FTOH	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
<dg	<dg	<dg	<dg	<dg		Children jacket	2	Greenpeace, 2012	Indonesia
418.5	<dg	<dg	229.5	189.0		Outdoor children jacket	3	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	4	Greenpeace, 2012	Vietnam
<dg	<dg	<dg	<dg	<dg		Ladies jacket	5	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	6	Greenpeace, 2012	Ukraine
<dg	<dg	<dg	<dg	<dg		Children rain trousers	7	Greenpeace, 2012	China
52	<dg	52.0	<dg	<dg		Ladies jacket	8	Greenpeace, 2012	China, sold as ”PFC-free”
123.0	<dg	72.00	30.00	21.00		Outdoor jacket	9	Greenpeace, 2012	China
99.0	<dg	99.00	<dg	<dg		Outdoor jacket	10	Greenpeace, 2012	China
17.6	<dg	17.6	<dg	<dg		Poncho for children	11	Greenpeace, 2012	China
40.6	<dg	<dg	40.6	<dg		Children jacket	12	Greenpeace, 2012	China
464.2	<dg	352	78.1	34.1		Ladies jacket	13	Greenpeace, 2012	China

Fluorotelomer alcohols (FTOHs), concentration (µg/m²)									
Total FTOHs	4:2 FTOH	6:2 FTOH	8:2 FTOH	10:2 FTOH	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
175.6	<dg	27.0	87.8	60.8		Ladies jacket	14	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
97.8	<dg	<dg	81.3	16.5		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
115.6	<dg	<dg	10.578	105		Ladies all-weather jacket	17	SFT, 2006	
285.0	<dg	<dg	154	131		Teflon tablecloth	18	SFT, 2006	DuPont Teflon®
42.2	<dg	<dg	31.8	10.4		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
86.8	<dg	<dg	62.3	24.5		Sporting jacket	20	SFT, 2006	
426.0	<dg	<dg	295	131		Over wear trousers for children	21	SFT, 2006	ReimaTec textile
206.6	<dg	<dg	140	66.6		Black snowsuit for children	22	SFT, 2006	
154.9	<dg	<dg	110	44.9		All-weather jacket	23	SFT, 2006	ProreTex textile

Fluorotelomer alcohols (FTOHs), concentration (µg/m²)									
Total FTOHs	4:2 FTOH	6:2 FTOH	8:2 FTOH	10:2 FTOH	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
33.0	<dg	<dg	24.9	8.08		Light-weight jacket	24	SFT, 2006	
168.3	<dg	<dg	125	43.3		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
91.0	<dg	<dg	50.4	40.6		All-weather jacket	26	Norges Naturvernforbund, 2006	FINE TEX“/DuPont™ Teflon“
384.8	<dg	12.8	224	148		All-weather jacket	27	Norges Naturvernforbund, 2006	Gore-Tex“ Pac Lite“
1001.1	<dg	<dg	954	47.1		All-weather jacket	28	Norges Naturvernforbund, 2006	HellyTech“
27.1	<dg	2.73	18.9	5.46		All-weather jacket	29	Norges Naturvernforbund, 2006	TCS WATER 2000

Detection limits: Greenpeace (6.8 – 179 µg/m²), SFT (0.48 – 2.0 µg/m²), Norges Naturforbund (0.50 - 0.54 µg/m²)

Fluorotelomer acrylates (FTA) and –olefins (FTolefin), concentration (µg/m²)									
Total	6:2 FTA	8:2 FTA	10:2 FTA	10:2 FTolefin	**Number > dg / total number**	**Type of textile product**	**Textile No.**	**Reference**	**Comments (e.g. country of origin or brand name)**
<dg – 78.3				-	11/14	Outdoor clothing for children and adult		Greenpeace, 2012	Intermediates in polymer production.
1.4	<dg	1.36	<dg	-		Greenpeace jacket	1	Greenpeace, 2012	China
10.1	<dg	10.13	<dg	-		Children jacket	2	Greenpeace, 2012	Indonesia
<dg	<dg	<dg	<dg	-		Outdoor children jacket	3	Greenpeace, 2012	China
19.5	<dg	19.5	<dg	-		Ladies jacket	4	Greenpeace, 2012	Vietnam
23.8	<dg	23.75	<dg	-		Ladies jacket	5	Greenpeace, 2012	China
6.1	<dg	6.09	<dg	-		Ladies jacket	6	Greenpeace, 2012	Ukraine
25.6	<dg	25.6	<dg	-		Children raintrousers	7	Greenpeace, 2012	China
20.8	20.8	<dg	<dg	-		Ladies jacket	8	Greenpeace, 2012	China, sold as ”PFC-free”
65.0	<dg	35	30	-		Outdoor jacket	9	Greenpeace, 2012	China
5.6	<dg	5.58	<dg	-		Outdoor jacket	10	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	-		Poncho for children	11	Greenpeace, 2012	China

<dg	<dg	<dg	<dg	-		Children jacket	12	Greenpeace, 2012	China
57.2	<dg	39.6	17.6	-		Ladies jacket	13	Greenpeace, 2012	China
78.4	<dg	58.1	20.3	-		Ladies jacket	14	Greenpeace, 2012	China
	-	-	-	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
	-	-	-	<dg		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
	-	-	-	<dg		Ladies all-weather jacket	17	SFT, 2006	
	-	-	-	<dg		Teflon tablecloth	18	SFT, 2006	DuPont Teflon
	-	-	-	<dg		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
	-	-	-	<dg		Sporting jacket	20	SFT, 2006	
	-	-	-	<dg		Outerwear trousers for children	21	SFT, 2006	ReimaTec textile
	-	-	-	<dg		Black snowsuit for children	22	SFT, 2006	
	-	-	-	<dg		All-weather jacket	23	SFT, 2006	ProreTex textile
	-	-	-	<dg		Light-weight jacket	24	SFT, 2006	

	-	-	-	<dg		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
	-	-	-	1,11		All-weather jacket	26	Norges Naturvern-forbund, 2006	FINE TEX“/DuPont™ Teflon“
	-	-	-	0,26		All-weather jacket	27	Norges Naturvern-forbund, 2006	Gore-Tex“ Pac Lite“
	-	-	-	0,85		All-weather jacket	28	Norges Naturvern-forbund, 2006	HellyTech“
	-	-	-	<dg		All-weather jacket	29	Norges Naturvern-forbund, 2006	TCS WATER 2000

Detection limits: Greenpeace (1.59 – 29.7 µg/m²), SFT (0.10 µg/m²), Norges Naturforbund (0.20 µg/m²)

Fluortelomersulfonates (FTSAs) and fluorotelomer -carboxylates (telomer acids, FTCAs), concentration (µg/m²)									
Total	6:2 FTSA	8:2 FTSA	6:2 FTCA	8:2 FTCA	**Number > dg / total number**	**Type of textile product**	**Textile No.**	**Reference**	**Comments (e.g. country of origin or brand name)**
<dg – 124.2					7/11	Ski and sporting clothing for children and adult		SFT, 2006	FTSAs can degrade to perfluoroalkyl sulfonates
<dg – 3,87					4/6	Jackets for all weather and anorak		Norges Naturvern-forbund, 2006	
<dg	<dg	<dg	<dg	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
<dg	<dg	<dg	<dg	<dg		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
2.72	<dg	<dg	<dg	2.72		Ladies all-water jacket	17	SFT, 2006	
5.56	<dg	<dg	3.72	1.84		Teflon tablecloth	18	SFT, 2006	DuPont Teflon
0.33	<dg	<dg	<dg	0.33		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
0.09	<dg	<dg	0.09	<dg		Sporting jacket	20	SFT, 2006	
0.38	0.38	<dg	<dg	<dg		Outerwear trousers for children	21	SFT, 2006	ReimaTec textile
<dg	<dg	<dg	<dg	<dg		Black snowsuit for children	22	SFT, 2006	

<dg	<dg	<dg	<dg	<dg		All-weather jacket	23	SFT, 2006	ProreTex textile
0.05	<dg	<dg	0.05	<dg		Light-weight jacket	24	SFT, 2006	
0.97	<dg	<dg	0.53	0.44		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
1.87	0.16	1.71	<dg	<dg		All-weather jacket	26	Norges Naturvern-for-bund, 2006	FINE TEX“/DuPont™ Teflon“
1.92	0.21	1.71	<dg	<dg		All-weather jacket	27	Norges Naturvern-for-bund, 2006	Gore-Tex“ Pac Lite“
3.87	0.34	3.53	<dg	<dg		All-weather jacket	28	Norges Naturvern-for-bund, 2006	HellyTech“
<dg	<dg	<dg	<dg	<dg		All-weather jacket	29	Norges Naturvern-for-bund, 2006	TCS WATER 2000

Detection limits: SFT (0.02 – 0.13 µg/m²), Norges Naturforbund (0.07 – 0.08 µg/m²)

Perfluorinated alkylsulfonates (PFS), concentration (µg/m²)									
Total	**PFBS**	**PFHxS**	**PFOS**	**PFDS**	**Number > dg / total number**	**Type of textile product**	**Textile No.**	**Reference**	**Comments (e.g. country of origin or brand name)**
0 – 30.5					9/11	Ski and sporting clothing for children and adult		SFT, 2006	
0.02 – 23.3					6/6	Jackets for all weather and anorak		Norges Naturvernforbund 2006	
<dg						Outdoor clothing for children and adult		Greenpeace, 2012	PFOS was not detected in samples from 2012, reflecting the ban from 2008
<dg	<dg	<dg	<dg	<dg		Greenpeace jacket	1	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Children jacket	2	Greenpeace, 2012	Indonesia
<dg	<dg	<dg	<dg	<dg		Outdoor children jacket	3	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	4	Greenpeace, 2012	Vietnam
<dg	<dg	<dg	<dg	<dg		Ladies jacket	5	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	6	Greenpeace, 2012	Ukraine
<dg	<dg	<dg	<dg	<dg		Children raintrousers	7	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	8	Greenpeace, 2012	China, sold as ”PFC-free”
<dg	<dg	<dg	<dg	<dg		Outdoor jacket	9	Greenpeace, 2012	China

<dg	<dg	<dg	<dg	<dg		Outdoor jacket	10	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Poncho for children	11	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Children jacket	12	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	13	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg		Ladies jacket	14	Greenpeace, 2012	China
0.06	<dg	0.04	0.02	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
<dg	<dg	<dg	<dg	<dg		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
30.5	0.02	0.38	30.1	<dg		Ladies all-weather jackets	17	SFT, 2006	
0.04	<dg	<dg	0.04	<dg		Teflon tablecloth	18	SFT, 2006	DuPont Teflon
0.03	<dg	<dg	0.03	<dg		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
<dg	<dg	<dg	<dg	<dg		Sporting jacket	20	SFT, 2006	
0.17	0.10	<dg	0.07	<dg		Outerwear trousers for children	21	SFT, 2006	ReimaTec textile
0.07	<dg	<dg	0.07	<dg		Black snowsuit for children	22	SFT, 2006	
0.07	<dg	<dg	0.07	<dg		All-weather jacket	23	SFT, 2006	ProreTex textile
0.06	<dg	<dg	0.06	<dg		Light-weight jacket	24	SFT, 2006	

0.26	<dg	0.02	0.24	<dg		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
0.02	0.02	<dg	<dg	<dg		All-weather jacket	26	Norges Naturvernforbund, 2006	FINE TEX“/DuPont™ Teflon“
0.28	0.12	<dg	0.16	<dg		All-weather jacket	27	Norges Naturvernforbund, 2006	Gore-Tex“ Pac Lite“
0.67	0.30	0.17	0.20	<dg		All-weather jacket	28	Norges Naturvernforbund, 2006	HellyTech“
0.44	0.08	0.12	0.24	<dg		All-weather jacket	29	Norges Naturvernforbund, 2006	TCS WATER 2000
<dg	<dg	<dg	<dg	<dg		Children jacket	30	Greenpeace, 2013	China
<dg	<dg	<dg	<dg	<dg		Children jacket	31	Greenpeace, 2013	China
<dg	<dg	<dg	<dg	<dg		Children jacket	32	Greenpeace, 2013	China

Detection limits: Greenpeace 2010 (0.042 – 0.277 µg/m²), SFT (0.01 µg/m²), Norges Naturforbund (0.02 – 0.04 µg/m²), Greenpeace 2010 (0.092 – 0.147 µg/m²)

Perfluoroalkyl carboxylic acids (PFCAs), concentration (µg/m²)															
Total	PFBA	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTeDA	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
2.97 - 170	-	-	-	-	-	-	-	-	-	-	11/11	Ski and sporting clothing for children and adult		SFT, 2006	
1.89 - 428	-	-	-	-	-	-	-	-	-	-	6/6	Jackets for all weather and anorak		Norges Naturvernforbund, 2006	
0.66 – 10.96	-	-	-	-	-	-	-	-	-	-	14/14	Outdoor clothing for children and adult		Greenpeace, 2012	PFOA could not be quantified in 6/14 samples
0.9	0.27	0.177	0.08	0.08	0.27	<dg	<dg	<dg	<dg	<dg		Greenpeace jacket	1	Greenpeace, 2012	China
5.1	0.22	0.37	0.47	0.24	2.01	0.26	1.02	0.08	0.46	<dg		Children jacket	2	Greenpeace, 2012	Indonesia
2.4	0.91	<dg	0.36	0.23	0.58	<dg	0.20	0.14	<dg	<dg		Outdoor children jacket	3	Greenpeace, 2012	China
0.5	0.20	<dg	<dg	<dg	0.24	<dg	0.08	<dg	<dg	<dg		Ladies jacket	4	Greenpeace, 2012	Vietnam
3.2	0.31	<dg	0.64	0.09	1.58	0.11	0.43	<dg	0.07	<dg		Ladies jacket	5	Greenpeace, 2012	China
0.6	0.37	<dg	<dg	<dg	0.20	<dg	<dg	<dg	<dg	<dg		Ladies jacket	6	Greenpeace, 2012	Ukraine

6.3	2.26	<dg	<dg	0.84	2.31	<dg	0.90	<dg	<dg	<dg		Children raintrousers	7	Greenpeace, 2012	China
1.2	0.58	<dg	0.30	<dg	0.29	<dg	<dg	<dg	<dg	<dg		Ladies jacket	8	Greenpeace, 2012	China, sold as ”PFC-free”
8.5	0.89	0.55	3.21	0.64	2.16	0.10	0.67	0.10	0.16	<dg		Outdoor jacket	9	Greenpeace, 2012	China
1.0	0.54	<dg	0.20	<dg	0.29	<dg	<dg	<dg	<dg	<dg		Outdoor jacket	10	Greenpeace, 2012	China
1.2	0.28	<dg	<dg	<dg	0.45	0.09	0.20	0.08	0.10	<dg		Poncho for children	11	Greenpeace, 2012	China
0.7	0.36	<dg	<dg	<dg	0.30	<dg	<dg	<dg	<dg	<dg		Children jacket	12	Greenpeace, 2012	China
5.4	0.53	1.34	0.58	0.93	0.65	0.62	0.36	0.23	0.16	<dg		Ladies jacket	13	Greenpeace, 2012	China
11.0	0.56	0.17	1.50	0.48	4.98	0.45	1.96	0.24	0.65	<dg		Ladies jacket	14	Greenpeace, 2012	China
6.3	0.50	<dg	1.45	0.30	3.08	0.14	1.37	<dg	<dg	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
3.0	<dg	<dg	<dg	<dg	0.42	2.03	0.37	0.15	<dg	<dg		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
34.9	4.5	2	5.46	5.18	13.7	2.21	0.45	0.34	0.85	0.16		Ladies all-weather jacket	17	SFT, 2006	
170.2	1.06	4.22	11.1	20.2	34.2	49.4	23.3	13.6	11.2	1.90		Teflon tablecloth	18	SFT, 2006	DuPont Teflon
10.8	<dg	<dg	4.94	0.05	1.21	0.49	3.53	0.16	0.37	<dg		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
18.9	<dg	<dg	6.06	1.59	9.26	<dg	<dg	0.56	1.18	0.21		Sporting jacket	20	SFT, 2006	

3.4	<dg	<dg	<dg	0.63	1.82	0.57	0.39	<dg	<dg	<dg		Outerwear trousers for children	21	SFT, 2006	ReimaTec textile
2.7	<dg	<dg	<dg	0.22	2.26	0.23	<dg	<dg	<dg	<dg		Black snowsuit for children	22	SFT, 2006	
7.0	<dg	<dg	<dg	0.19	1.73	2.44	1.41	0.76	0.22	0.22		All-weather jacket	23	SFT, 2006	ProreTex textile
17.6	0.30	<dg	2.64	0.78	8.23	1.51	2.98	0.18	0.84	0.13		Light-weight jacket	24	SFT, 2006	
94.7	2.62	2.94	24.0	10.5	32.7	6.27	13.1	0.70	1.83	0.06		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
16.4	<dg	<dg	2.25	2.82	4.76	5.75	0.40	0.42	<dg	<dg		All-weather jacket	26	Norges Naturvern-forbund, 2006	FINE TEX“/DuPont™ Teflon“
55.9	0.72	<dg	8.64	4.42	24.6	3.52	10.6	0.74	2.61	<dg		All-weather jacket	27	Norges Naturvern-forbund, 2006	Gore-Tex“ Pac Lite“
53.3	0.83	<dg	11.8	3.74	20.4	0.53	11.4	3.07	1.48	<dg		All-weather jacket	28	Norges Naturvern-forbund, 2006	HellyTech“
1.9	<dg	<dg	0.62	0.47	0.80	<dg	<dg	<dg	<dg	<dg		All-weather jacket	29	Norges Naturvern-forbund, 2006	TCS WATER 2000
<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg	<dg		Children jacket	30	Greenpeace, 2013	China
5.48	0.182	0.124	0.808	0.239	2.436	0.150	1.086	<dg	0.467	<dg		Children jacket	31	Greenpeace, 2013	China
<dg	<dg	<dg	<dg	<dg	0.317	<dg	<dg	<dg	<dg	<dg		Children jacket	32	Greenpeace, 2013	China

Detection limits: Greenpeace (0.042 – 0.804 µg/m²), SFT (0.03 – 0.23), Norges Naturforbund (0.20 – 0.50 µg/m²), Greenpeace 2010 (0.092 – 0.098 µg/m²)

Perfluoralkyl sulfonamides (PFASsA), concentration (µg/m²)										
Total	FOSA	MaFOSA	EtFOSA	MeFOSE	EtFOSE	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
<dg – 22.8						8/11	Ski and sporting clothing for children and adult		SFT, 2006	
1.43 - 107						6/6	Jackets for all weather and anorak		Norges Naturvernforbund 2006	
<dg						0/14	Outdoor clothing for children and adult		Greenpeace, 2012	
<dg	<dg	<dg	<dg	<dg	<dg		Greenpeace jacket	1	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Children jacket	2	Greenpeace, 2012	Indonesia
<dg	<dg	<dg	<dg	<dg	<dg		Outdoor children jacket	3	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	4	Greenpeace, 2012	Vietnam
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	5	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	6	Greenpeace, 2012	Ukraine
<dg	<dg	<dg	<dg	<dg	<dg		Children raintrousers	7	Greenpeace, 2012	China

Perfluoralkyl sulfonamides (PFASsA), concentration (µg/m²)										
Total	FOSA	MaFOSA	EtFOSA	MeFOSE	EtFOSE	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	8	Greenpeace, 2012	China, sold as "PFC-free"
<dg	<dg	<dg	<dg	<dg	<dg		Outdoor jacket	9	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Outdoor jacket	10	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Poncho for children	11	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Children jacket	12	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	13	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Ladies jacket	14	Greenpeace, 2012	China
<dg	<dg	<dg	<dg	<dg	<dg		Alpine trousers for children	15	SFT, 2006	AIR-FLO coating
0.41	<dg	<dg	<dg	0.35	0.06		Snowsuit for children	16	SFT, 2006	Ping Protection (finished polyester)
22.79	21.3	<dg	<dg	1.10	0.39		Ladies al-weather jacket	17	SFT, 2006	
0.03	0.03	<dg	<dg	<dg	<dg		Teflon tablecloth	18	SFT, 2006	DuPont Teflon

Perfluoralkyl sulfonamides (PFASsA), concentration (µg/m²)										
Total	FOSA	MaFOSA	EtFOSA	MeFOSE	EtFOSE	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
<dg	<dg	<dg	<dg	<dg	<dg		Alpine jacket	19	SFT, 2006	MaxAliento membrane from Maxland Technical Textile
<dg	<dg	<dg	<dg	<dg	<dg		Sporting jacket	20	SFT, 2006	
0.03	0.03	<dg	<dg	<dg	<dg		Over wear trousers for children	21	SFT, 2006	ReimaTec textile
0.04	<dg	<dg	0.04	<dg	<dg		Black snowsuit for children	22	SFT, 2006	
1.08	<dg	<dg	<dg	0.65	0.43		All-weather jacket	23	SFT, 2006	ProreTex textile
0.36	<dg	<dg	<dg	0.29	0.07		Light-weight jacket	24	SFT, 2006	
0.74	0.04	<dg	<dg	0.52	0.18		Sample of textile for outdoor use	25	SFT, 2006	G-1000 textile, according to Fjellräven the most used fabric in their product range
1.43	<dg	<dg	<dg	1.43	<dg		All-weather jacket	26	Norges Naturvern-forbund, 2006	FINE TEX“/DuPont™ Teflon“
14.13	0.07	0.16	<dg	13.9	<dg		All-weather jacket	27	Norges Naturvern-forbund, 2006	Gore-Tex“ Pac

Perfluoralkyl sulfonamides (PFASsA), concentration (µg/m²)										
Total	FOSA	MaFOSA	EtFOSA	MeFOSE	EtFOSE	Number > dg / total number	Type of textile product	Textile No.	Reference	Comments (e.g. country of origin or brand name)
										Lite“
6.1	0.06	<dg	<dg	6.04	<dg		All-weather jacket	28	Norges Naturvern-forbund, 2006	HellyTech“
4.3	<dg	<dg	<dg	4.30	<dg		All-weather jacket	29	Norges Naturvern-forbund, 2006	TCS WATER 2000
<dg	<dg	-	- -	-	-		Children jacket	30	Greenpeace, 2013	China
<dg	<dg	-	-	-	-		Children jacket	31	Greenpeace, 2013	China
<dg	<dg	-	-	-	-		Children jacket	32	Greenpeace, 2013	China

Detection limits: Greenpeace (0.85 – 29.7 µg/m²), SFT (0.01 – 0.06 µg/m²), Norges Naturforbund (0.03 – 0.40 µg/m²)

Polyfluoroalkyl substances (PFASs) in textiles for children

Polyfluoroalkyl substances (PFASs) is a large family of surfactants with different uses and environmental and health properties. There are major difference in how thoroughly the substances are tested for hazardous effects on human health and the environment. Among the substances studied most thoroughly, carcinogenic, toxic for reproduction and acute toxic effects have been observed. PFAS-based coatings are being used in garments and other textiles in order to make the materials water and dirt repellent.

The purpose of this report is to investigate which consumer products of textiles for children contain PFASs and analyse which PFASs that can be found in the materials. Further the extent to which PFASs may be released during wear and washing of textiles is investigated and it is assessed whether the release of the substances poses health and environmental risk. Relevant waste streams, waste amounts and fates of the substances in waste treatment are also assessed.

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