Are imported consumer products an important diffuse source of PFASs to the Norwegian environment?

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The aim of this study was to measure perfluoroalkyl substances in a selection of imported consumer products (n = 45) and estimate population normalized emission rates during the use phase. 6:2 and 8:2 perfluorotelomer alcohol (FTOH) were found in the highest concentrations ranging from <MDL to 374 and 163 µg m⁻² respectively. Concentrations of FTOHs were approximately 2–3 orders of magnitude higher than those of perfluorooctyl carboxylic acids (PFCAs). Although perfluorooctane sulfonate (PFOS) was detected in one carpet sample at 1.7 µg m⁻², the majority of samples complied with regulatory limits for PFOS in the EU. Population normalized emission rates of perfluorooctanoic acid, 6:2 FTOH and 8:2 FTOH from imported consumer products were estimated to be 6.6, 2130 and 197 g year⁻¹ capita⁻¹ respectively for the “intermediate” emission scenario. The results from this study suggest that emissions from imported products would have a small impact on the environmental concentrations of perfluoroalkyl acids on a regional scale.

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1. Introduction

Per- and polyfluoroalkyl substances (PFASs) are a large group of synthetic chemicals which have been manufactured for their unique amphiphilic properties and chemical stability (Kissa, 2001). Over the past decade PFASs have received considerable attention as a new group of persistent environmental contaminants with global distribution (Lindstrom et al., 2011). Long-chain perfluoroalkyl carboxylic acids (PFCAs, CₙF₂ₙ₊₁COOH, n ≥ 7) and perfluoroalkane sulfonic acids (PFASs, CₙF₂ₙ SO₃H, n ≥ 6) are of special concern due to their persistence (Kissa, 2001; Natarajana et al., 2005), bio-accumulation potential (Conder et al., 2008) and toxicity (Lau et al., 2007). Concern regarding the hazard profile of PFCAs and PFASs has led to a series of actions by regulators and the producing industry. In 2001, perfluorooctane sulfonate (PFOS) and related long-chain PFASs were phased out by the major global manufacturer in North America and Europe (3M Company, 2000; USEPA, 2000). Stewardship agreements have also been reached between the US Environmental Protection Agency, and leading global companies to reduce emissions and product content of perfluorooctanoic acid (PFOA) and related chemicals by 95% by 2010 and to work toward their elimination by 2015 (USEPA, 2006). PFOS has been included in the Stockholm Convention on Persistent Organic Pollutants as an Annex B substance, i.e., restricted in its use (UNEP, 2009) and more recently PFOA was identified as Substances of Very High Concern by the European Chemical Agency (ECHA, 2013).

The actions to reduce emissions of long-chain PFASs has lead to a substitution towards short-chain homologues which are considered to be less bioaccumulative (Conder et al., 2008) or PFASs with ether bonds inserted in the polyfluoroalkyl moiety which are perceived to be less persistent (Buck et al., 2011; Wang et al., 2013). However, a parallel trend is the continuous or increasing production of PFASs in other parts of the world than Europe and North America (Lindstrom et al., 2011; Lim et al., 2011; Xie et al., 2013). As Annex B of the Stockholm Convention has several exemptions for continuous use of PFOS, and other long-chain PFASs are still not recognized by international agreements (UNEP, 2009), these chemicals can still be used in similar applications as before the phase out actions took place. In China, the production of PFOS was estimated to increase from approximately 30 to 250 metric tons between 2001 and 2006 and has remained fairly constant over the

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The continuous production and use of PFOS and related chemicals indicates that import of consumer products may contribute to diffuse emissions of PFASs in countries where they are no longer produced. Release of unbound PFAS residuals from consumer products during the use phase has previously been identified as a source of PFCS and PFASs to the environment (Dinglasan-Paniliao and Mabury, 2006; Prevedouros et al., 2006; Paul et al., 2009) and a recent substance flow analysis performed in Switzerland reported that treated textiles and carpets (the majority from import) was a substantial source of PFOA in Switzerland (Buser and Morf, 2009).

The textile industry in China has been identified as the primary industrial sector of fluorinated finishing agents with approximately 10 000 metric tons of fluorinated finishing agents being used annually (Lim et al., 2011); but little is known about the abundance and composition of PFASs in China. Several monitoring studies have reported levels of residual PFASs in a variety of consumer products (Berger and Herzke, 2006; SFT, 2006; Norin and Schulze, 2007; Gewurtz et al., 2009; Herzke et al., 2012), but there are only a few studies specifically reporting the levels of PFAS residuals in consumer products of Chinese origin (Greenpeace, 2013; Brigden et al., 2013; Knepper et al., 2014).

The aim of this study was to explore the hypothesis that import of PFASs in consumer products from China appreciably adds to the environmental emissions in non-producing countries using Norway as a case study. China is the main origin of textiles purchased in Norway in 2007 (SSB, 2008/48). Furthermore, China is one of the countries with on-going production of long-chain PFASs (Wang et al., 2014). Thus, textiles, carpets, clothing, and food contact materials imported from China to Norway were specifically investigated to (i) measure extractable PFAS residuals in imported consumer products of Chinese origin, (ii) evaluate compliance with regulatory guidelines for PFAS content in consumer products and (iii) estimate the resulting emissions to air and water from the investigated product categories.

2. Methods

2.1. Sample collection

A total of 45 furniture textile, carpet, clothing and food contact material samples were purchased from three major retail stores in Tromsø and Trondheim during the period November 2012–February 2013 (Table S1). The sampling campaign was designed to screen for PFASs in a wide range of consumer products belonging to product categories which have previously been found to contain PFAS residuals (Berger and Herzke, 2006; SFT, 2006; Norin and Schulze, 2007; Gewurtz et al., 2009; Washburn et al., 2005; Begley et al., 2005; Herzke et al., 2012). A second selection criterion for the purchased product categories was that they were representative of products imported from China to Norway in large quantities (SSB, 2013). The individual products were randomly selected from a broad range of products without any prior knowledge of the treatment with PFAS in order to create a representative set of samples corresponding to the volumes of goods being imported to Norway from China. Outdoor clothing were excluded from the selection due to recent studies on these types of consumer products (Greenpeace, 2013; Brigden et al., 2013; Knepper et al., 2014). For this product category, concentration data from previous reports were used to calculate population normalized emission rates (see Section 2.3).

2.2. Chemical analysis

2.2.1. Chemicals

The 22 target analytes in this study were C₄–C₁₄ PFCS, C₄, C₆, C₈, C₁₀ PFASs, 4, 6, 8, 10 and 12 FTOHs, N-ethyl perfluorooctanoic acid (EtFOSA), N-Methyl perfluorooctanoic acid (MeFOSA), N-Ethyl perfluorooctanoic acid (EtFOSA) and N-Methyl perfluoroctanoic acid (MeFOSA). Mass-labeled internal standards were 1³C₄–PFCS, 1³O₂–PFASs 1³C₄–FTOHs, D₃–EtFOSA, D₃–MeFOSA, D₅–EtFOSA and D₅–MeFOSA (see also the supplementary material). 3,7-branched perfluorodecanoate (3,7 DMPFDOA) and 7:1 FTOH were used as volumetric standards to calculate the recovery of anionic and neutral PFASs respectively. All native and isotope labeled PFCA and PFSA standard compounds were purchased from Wellington Laboratories (Guelph, ON, Canada) in 2 μg mL⁻¹ solution mixtures. All solvents and reagents used in this work were of LiChrosolv grade.

2.2.2. Extraction and analysis

Two previously reported methods were used to extract and analyze ionophoric PFASs (PFCSs and PFASs) and neutral PFASs (FOSEs, FOSAs and FTOHs) separately (Herzke et al., 2012). Briefly, two squares (10 × 10 cm) of textile, carpet or food contact material were cut with a pair of scissors which had been rinsed in methanol. The sample was weighed, cut into small pieces and spiked with mass-labeled internal standards. Samples were extracted with methanol for ionic compounds and ethyl acetate for neutral PFASs two times for 15 min in an ultrasonic bath with vortex treatment in between. After centrifugation and solvent evaporation, an aliquot of 1 mL extract was transferred for dispersive clean up with ENVI-Carb (50 mg, 1 mL, 100–400 mesh, Supelco, USA) according to the method of Powley et al. (2005). Addition of glacial acetic acid (50 μL) to the ENVI-carb was used only for the ionic PFASs. Finally, a volume of 500 μL of the supernatant was transferred to a new glass vial and 2 ng of the volumetric standard was added and mixed thoroughly. For ionic PFASs, aliquots of 100 μL were diluted with 2 mL aqueous ammonium acetate solution (NH₄OAc, 50/50, v/v) prior to analysis whereas neutral PFASs were analyzed in ethyl acetate. PFCSs and PFASs were analyzed by ultra-high performance liquid chromatography–quadrupole mass-spectrometry (UPLC–QMS) whereas FOSEs, FOSAs and FTOHs were analyzed by gas chromatography–mass-spectrometry (GC–MS) in selected ion monitoring (SIM)-mode. Further details on the instrumental analysis are given in the supplementary material.

2.2.3. Quality assurance

The analytical method has previously been used for determination of extractable residuals in textiles (Berger and Herzke, 2006; Herzke et al., 2012). The NILU Tromsø laboratory has contributed in a number of inter-laboratory comparison studies for PFAS analysis with satisfactory results (Weiss et al., 2013). Recoveries of internal standards, procedural and instrumental blanks and method detection limits are regularly monitored as quality criteria for the analysis. Average recoveries for the internal standards ranged from 46 to 108% and 62 to 143% for ionic and neutral PFASs respectively (see also Table S2–S3). The recoveries indicated satisfactory extraction efficiency and only limited losses during sample preparation for the majority of analytes, whereas recoveries >100% for
some analytes also indicated a slight matrix enhancement of the ionization efficiency of the volumetric standards compared to the internal standards. A commonly observed problem in trace analysis of PFASs is contamination from the extraction and clean-up procedures. To circumvent this problem solvents and disposable laboratory equipment were routinely checked for PFSA contamination prior to use. Three procedural blanks were also included for every batch of 20 samples and 1 solvent injection blank was analyzed for every 10 samples injected. No blank contamination was detected, however, for any of the analytes. Within sample replicates were performed for 6 samples with low (<50%) recovery for one or several of the analytes which were extracted and analyzed in triplicate (Table S4–S5). Method detection limits (MDLs) were calculated on the basis of the lowest calibration standard giving 60% total average recovery. The MDLs for individual substances ranged from 0.005 to 0.010 µg m⁻² and 0.5 to 2 µg m⁻² for ionic and neutral PFASs respectively.

2.3. Calculation of population normalized emission rates from imported consumer products

The methodology to calculate population normalized emission rates (ER_capita) of PFASs to air and water respectively from imported consumer products was based on the following equation:

\[ ER_{a,w} = \frac{n \cdot Q \cdot EF_{a,w}}{P} \]

where \( C_i \) is the concentration of extractable PFASs in a specific consumer product category (µg kg⁻¹), \( Q \) is the quantity of that consumer product category being imported from China to Norway from China (kg year⁻¹), \( E_{F_{a,w}} \) is the emission factor (unitless) describing the transfer efficiency of different PFASs to air and water respectively and \( P \) is the population of Norway (capita). Extractable concentrations in the consumer product categories were derived from the chemical analysis (described above) with the exception of outdoor clothing. For these products, a data set of 17 measurements were derived from recently analyzed products (2012–2013) manufactured in China (Greenpeace, 2013; Brigiden et al., 2013; Knepper et al., 2014). Although the concentrations of PFASs in this paper are reported in µg m⁻², transformation to µg kg⁻¹ was performed by using the weight per surface area of consumer products (Table S1) to enable calculation of emission rates from import quantities. The quantities of different consumer product categories imported from China and population of Norway were derived from publicly available national statistics in Norway (SSB, 2013). The summed annual import of furniture textiles, carpets/mats, cotton and leather clothing and food packaging material and outdoor clothing was estimated to be 3920, 367, 10 700, 628, 11 600 metric tons respectively. For a more comprehensive summary of the import statistics see Table S6–S10 of the Supplementary Material. Emission factors of different PFASs were derived from a recent study (Knepper et al., 2014). The study of Knepper et al. demonstrated that extractable residuals of PFCCAs and PFSA microresidues in furniture textiles, food packaging, carpets and clothing is emitted during washing to municipal wastewater within a year of usage. In contrast to PFCCAs and PFSA microresidues, neutral PFASs (including FTOHs) typically have a low water solubility and a high vapor pressure (Kissa, 2001). A flow-through study showed that a significant part of the extractable FTOHs present in jackets was released to air during 5–days of forced ventilation (Knepper et al., 2014). The fraction of extractable chemical that was released to air was 53.7% and 12.4% for 6:2 FTOH and 8:2 FTOH respectively. During a period of one year of repeated use it may, however, be expected that a larger fraction of the FTOHs are released compared to five days of forced ventilation. For calculation of emissions of FTOHs from consumer products to air, we therefore used 95th percentile values (100% and 17.6% respectively) from the flow-through study (Knepper et al., 2014). The variability in concentrations within the consumer product categories were used to estimate the uncertainty range in population normalized emission rates. A low, intermediate and high scenario was used that the 5th, 50th and 95th percentile concentrations present in the products were generated to reflect the uncertainty range in emissions (see Tables S11–S12).

3. Results and discussion

3.1. Concentrations of extractable PFASs in consumer products

Overall, PFASs were detected in 69% of the samples with 6:2 FTOH, 8:2 FTOH and PFOA being the most frequently detected analytes. The extractable concentrations of PFCCAs and FTOHs are shown in Tables 1 and 2. PFSA microresidues were only detected in two samples (Table S13) whereas FOSAs and FOSAs were not detected in any of the products. The concentrations of individual target analytes displayed a large variation within and between the different consumer product groups.

The highest concentrations among the furniture textile samples were observed in the down quilt (T33), sofa pillow fabric (T23) and two curtain samples (T2 and J3) which contained individual PFCCAs in the range 0.14–1.022 µg m⁻². Although PFOA was the most frequently detected homologue, PFHpA was detected in the highest concentration. For all other furniture textiles, PFCCAs and PFSA were present at trace concentrations between MDL and 0.043 µg m⁻². The curtain sample (T2) also had relatively high concentrations of 8:2 FTOH (164 µg m⁻²) and 10:2 FTOH (60 µg m⁻²) while 6:2 FTOH was detected at relatively high concentrations in one pillow (J1; 374 µg m⁻²), one pillow case (T4; 242 µg m⁻²), one towel (T5; 70 µg m⁻²), one chair cover (T11; 26 µg m⁻²), two fleece blankets (T12 and J6; 18 and 30 µg m⁻²), one seat pillow (J4; 43 µg m⁻²) and one curtain sample (J3; 54 µg m⁻²). In carpet and mat samples, 6:2 FTOH was the most frequently detected homologue ranging between MDL and 74 µg m⁻² (median of 25 µg m⁻²). Concentrations of 4:2 FTOH and 8:2 FTOH close to the MDL were typically found in the same samples which contained 6:2 FTOH. PFOS was also detected in one carpet sample at 1.7 µg m⁻². PFAS microresidues were detected in all cotton and leather clothing (including one imitation leather jacket). However, the concentrations of extractable PFAS microresidues were generally close to the MDL, except for one cotton bath robe sample (J2) which contained 370 µg m⁻² of 6:2 FTOH. All food contact material samples contained detectable concentrations of PFCCAs; one paper plate (T26) and one paper cup (T29) sample contained elevated concentrations of PFOA (0.5 µg m⁻²). 8:2 FTOH was also detected in three of the food contact material samples, ranging up to 12.3 µg m⁻².

The primary use of PFASs in furniture textiles, carpets, mats and clothing is to impart water and stain repellency. Although the active coating used for surface treatment are typically polymeric
materials (Buck et al., 2011), low molecular weight PFASs, including FTIHs and PFCAs, may be present as unreacted starting materials or impurities from the production process (Dinglasan-Panililo and Mabury, 2006; Washburn et al., 2005; Mawn et al., 2005; Larsen et al., 2006; Russell et al., 2008; Russell et al., 2010). The variable concentrations of PFCAs and PFCAs and FTIHs most likely reflect the degree of treatment with side-chain fluorinated polymers to provide surface protection (Dinglasan-Panililo and Mabury, 2006; Buck et al., 2011). The low trace-level concentrations of PFCAs in the majority of samples may, however, also be due to a low level addition of PFCAs to products as act as leveling agents in dyes and detergents (Kissa, 2001). It is also possible that products that have not been intentionally treated with PFASs become contaminated throughout the retail chain as it has been shown that furniture and outdoor textile shops have highly elevated concentrations of FTIHs compared to other indoor environments (Schlumber et al., 2013). The presence of low molecular weight PFASs in food contact materials has previously been attributed to residuals present in polyfluoralkyl phosphoric acid esters (PAPs) which are used for the treatment of paper and board (Regley et al., 2005; Trier et al., 2011).

### 3.2. Comparison of PFAS concentrations with previous studies and regulatory guidelines

The present study demonstrates that PFASs can be detected in a wide range of imported consumer products using sensitive analytical techniques. However, among the 45 products analyzed, the majority of samples contained low molecular weight, non-polymeric PFASs at trace levels. The presence of PFASs was not due to intentional treatment of textiles with these substances, but rather due to residual impurities of low molecular weight PFASs in polymeric surface treatments that are based on PFAS chemistry (Paul et al., 2009; Wang et al., 2013). One out of 45 samples contained concentrations of PFOS (1.7 μg m⁻²) above the EU and Norwegian limit of 1 μg m⁻² (ECHA, 2013). To put these measurements into perspective a comparison with previous studies on PFAS residuals in various consumer products is presented in Table 3.

The literature on PFASs in consumer products consists of industry studies conducted on fluorocatalytic raw materials or treated materials with a known composition (Fraunhofer, 2004;
Concentrations above MDL are presented in bold.

several commitments to replace long-chain PFAS with short-chain (Fraunhofer, 2004). Since the of PFASs display considerable variability, ranging over (Filipovic et al., 2013, 2014) and the total off-gassing of neutral residuals in several consumer product categories in the US between 2007 and 2011. It may also be noted in this context that the products imported from China (which has ongoing production of PFOS and related chemicals) do not have notably different composition of PFASs in products sampled randomly from the market in recent years. The high detection frequency of 6:2 FTOH in this study rather implies that the textile treatment solutions used in Chinese textiles are primarily based on short-chain PFASs.

Although there is indicative evidence that PFAS concentrations in consumer products have decreased due to industry actions, differences in sampling and analysis may also explain some of the variability between different studies (Table 1). To begin with, the lack of a stringent sample definition for treated consumer products makes it difficult to know if the analyzed samples are representative of the product categories present on the market. In contrast to this, most performance by industry (Fraunhofer, 2004; Washburn et al., 2005; Mawn et al., 2005; Larsen et al., 2006; Russell et al., 2008, 2010), market screening studies have limited information about how the samples were treated and handled prior to the date of purchase. Differences in sub-sampling may also affect the quantitative results. For instance, some studies report the presence of PFASs in the treated carpet fibers (Washburn et al., 2005), whereas this and other studies (Hertzke et al., 2012; Liu et al., 2014) also analyzed the carpet backing. Repeated measurements of PFAS residuals in well-defined fluorochrome raw materials and finished products may also display some variability due to the heterogeneity of the samples (Fraunhofer, 2004; Larsen et al., 2006; Liu et al., 2014). For measurements of FTOHs using GC–MS it has also been hypothesized that fragmentation of co-extracted polymeric PFASs in the GC inlet may lead to an overestimation of the extractable amount of residuals (Larsen et al., 2006). However, we did not test for this since no information about the applied fluoropolymers in the investigated products was available to us. Given the efforts to regulate PFOS (UNEP, 2009) and more recently PFOA in finished products there’s clearly a need for robust and standardized methods for sampling and analysis. Inter-laboratory comparison studies (Weiss et al., 2013) would also be valuable to evaluate and improve the reproducibility of analytical data for PFASs in various consumer products.

### Table 2
Extractable concentrations of neutral PFASs ($\mu g \text{ m}^{-2}$) in consumer products imported from China.

| Sample nr. | Sample type | Concentration ($\mu g \text{ m}^{-2}$) |
|------------|-------------|--------------------------------------|
| T1         | Furniture textiles Table cloth          | <0.5 22.3 0.9 <0.5                  |
| T2         | Furniture textiles Curtain              | <0.5 8.49 163 60.17                |
| T3         | Furniture textiles Pillow case          | <0.5 <0.5 1.0 <0.5                  |
| T4         | Furniture textiles Pillow case          | <0.5 241.1 1.4 <0.5                |
| T5         | Furniture textiles Towel                | <0.5 69.99 1.3 0.87               |
| T6         | Furniture textiles Bed sheet            | <0.5 <0.5 0.7 <0.5                   |
| T7         | Furniture textiles baby blanket         | <0.5 <0.5 <0.5 <0.5                  |
| T8         | Furniture textiles chair cover          | <0.5 0.55 <0.5 <0.5                  |
| T9         | Furniture textiles Kitchen towel        | <0.5 <0.5 <0.5 <0.5                  |
| T10        | Furniture textiles Baby blanket         | <0.5 <0.5 <0.5 <0.5                  |
| T11        | Furniture textiles chair cover          | <0.5 26.2 1.0 0.55                 |
| T12        | Furniture textiles fleece blanket       | <0.5 18.2 2.4 0.98                  |
| T13        | Furniture textiles Sofa cover           | <0.5 <0.5 <0.5 <0.5                  |
| T23        | Furniture textiles Sofa pillow fabric   | <0.5 <0.5 <0.5 <0.5                  |
| T24        | Furniture textiles Roll packed quilts   | <0.5 <0.5 <0.5 <0.5                  |
| T30        | Furniture textiles Baby bed cover       | <0.5 <0.5 <0.5 <0.5                  |
| T31        | Furniture textiles Baby mattress        | <0.5 <0.5 <0.5 <0.5                  |
| T32        | Furniture textiles Down quilt           | <0.5 <0.5 <0.5 <0.5                  |
| T33        | Furniture textiles Down quilt           | <0.5 <0.5 <0.5 <0.5                  |
| J1         | Furniture textiles Pillow               | <0.5 373.7 <0.5 <0.5                |
| J3         | Furniture textiles Curtain              | <0.5 54.0 <0.5 <0.5                  |
| J4         | Furniture textiles Seat pillow          | <0.5 43.4 <0.5 <0.5                  |
| J6         | Furniture textiles Fleece blanket       | <0.5 29.6 <0.5 <0.5                  |
| J7         | Furniture textiles Bed linen            | <0.5 <0.5 <0.5 <0.5                  |
| J8         | Furniture textiles Towel                | <0.5 36.1 <0.5 <0.5                  |
| J10        | Furniture textiles Quilt               | <0.5 16.8 <0.5 <0.5                  |
| J12        | Furniture textiles Quilt                | <0.5 24.6 <0.5 <0.5                  |
| T13        | Carpets/mats Sheep skin                | <0.5 <0.5 <0.5 <0.5                  |
| T17        | Carpets/mats Carpet                    | <0.5 <0.5 <0.5 <0.5                  |
| T18        | Carpets/mats Carpet                    | <0.5 <0.5 <0.5 <0.5                  |
| T19        | Carpets/mats Carpet                    | <0.5 65.1 1.7 <0.5                  |
| T20        | Carpets/mats Sheep skin                | 0.71 25.2 0.9 <0.5                  |
| T21        | Carpets/mats Rug                       | <0.5 22.3 1.4 <0.5                  |
| J5         | Carpets/mats Lamb skin                 | <0.5 57.0 <0.5 <0.5                  |
| J9         | Carpets/mats Bath mat                  | <0.5 74.4 <0.5 <0.5                  |
| J11        | Carpets/mats Bath mat                  | <0.5 59.1 <0.5 <0.5                  |
| T14        | Cotton/leather clothes Jacket          | <0.5 <0.5 <0.5 <0.5                  |
| T15        | Cotton/leather clothes Jacket          | <0.5 0.71 <0.5 <0.5                  |
| T16        | Cotton/leather clothes Jacket          | <0.5 <0.5 <0.5 <0.5                  |
| J2         | Cotton/leather clothes Bath robe       | <0.5 373 <0.5 <0.5                  |
| T25        | Food contact material Baking mold      | <0.5 <0.5 <0.5 <0.5                  |
| T26        | Food contact material Paper plates      | <0.5 12.1 3.29 <0.5                  |
| T27        | Food contact material Baking cover      | <0.5 <0.5 <0.5 <0.5                  |
| T28        | Food contact material Paper plates      | <0.5 <0.5 <0.5 <0.5                  |
| T29        | Food contact material Paper cup        | <0.5 <0.5 <0.5 <0.5                  |

Concentrations above MDL are presented in bold.
municipal wastewater influents after correcting for background contamination in tap water (Filipovic and Berger, 2014). For 8:2 FTOH the intermediate population normalized emission rates estimated in this study were a factor 300 lower than the estimated total diffuse emissions in the city of Zurich (Wang et al., 2012). Unfortunately, a comparison with total diffuse emissions was not possible for 6:2 FTOH, since these were not calculated by Wang et al. (2012).

The lack of agreement between different emission estimation approaches (Fig. 1) suggests that emissions from imported consumer products, as calculated in this study, would have a negligible impact on the total diffuse emissions. This result is surprising since previous studies have suggested that treated textiles and carpets are the major diffuse sources of PFOA and 8:2 FTOH to water and air respectively (Buser and Morf, 2009). A comparison with the input data used by Buser and Morf (2009) reveal that total import numbers of treated textiles are slightly lower than this study (3.2 compared to 7.5 kg year⁻¹ capita⁻¹) whereas extractable concentrations of PFOA and 8:2 FTOH are 2–4 orders of magnitude higher than used in this study for the intermediate scenario. The discrepancy in emission estimates between this study and that of Buser and Morf (2009) can therefore be attributed to the relatively low concentrations of extractable PFASs observed in this study (Table 1). Combined with the preliminary evidence of a decreasing trend in PFAS concentrations in consumer products over time (Liu et al., 2014), the discrepancy in emission estimates may therefore suggest that urban diffuse releases to air and water (as estimated by Wang et al., 2012; Filipovic et al., 2013) reflect a legacy effect of old

![Figure 1](image-url)

**Fig. 1.** Comparison of population normalized emission rates of PFOA, 6:2 FTOH and 8:2 FTOH from imported consumer products (this study) with total diffuse emissions to water (Filipovic et al., 2013) and air (Wang et al., 2012). Total diffuse emission rates of PFOA from Filipovic et al. (2013) were based on domestic wastewater influent which was corrected for the background concentration in tap water. *Not calculated by Wang et al. (2012).*

| Concentration of extractable PFASs (ng g⁻¹) |
|---------------------------------------------|
| Sample                                      |
| n                                          |
| 6:2 FTOH                                    |
| 8:2 FTOH                                    |
| PFOA                                       |
| PFOS                                       |
| Sampling year                               |
| Ref.                                        |
| Treated fabric web                          |
| 2 NA                                        |
| NA                                          |
| NA                                          |
| 2001                                       |
| Fraunhofer 2004                             |
| Mill-treated carpet                         |
| >60 NA                                      |
| NA                                          |
| NA                                          |
| 2004                                       |
| Wang et al., 2005                           |
| Treated apparel                             |
| >100 NA                                     |
| NA                                          |
| NA                                          |
| 2004                                       |
| Washburn et al., 2005                       |
| Treated upholstery                         |
| 3 NA                                        |
| NA                                          |
| 2004                                       |
| Wang et al., 2005                           |
| Outdoor clothing                            |
| >14<110                                     |
| >14<1230000                                |
| 2006                                       |
| Berger and Herzke 2006                     |
| Pre-treated carpeting                       |
| 9 NA                                        |
| NA                                          |
| <6:2 MDL-226                               |
| 2007–2011                                   |
| Liu et al., 2014                           |
| Treated apparel                             |
| 13 NA                                       |
| NA                                          |
| <6:2 MDL-269                               |
| 2007–2010                                   |
| Liu et al., 2014                           |
| Treated home textiles and upholstery        |
| 6 NA                                        |
| NA                                          |
| 19–330                                     |
| 2007–2011                                   |
| Liu et al., 2014                           |
| Treated non-oven medical garments           |
| 9 NA                                        |
| NA                                          |
| <6:2 MDL-369                               |
| 2008–2011                                   |
| Liu et al., 2014                           |
| Food contact material                       |
| 9 NA                                        |
| NA                                          |
| 1.8–2500                                   |
| 2007–2011                                   |
| Liu et al., 2014                           |
| Membranes for apparel                       |
| 8 NA                                        |
| NA                                          |
| <6:2 MDL-163                               |
| 2007–2011                                   |
| Liu et al., 2014                           |
| Carpets                                    |
| 2 14–183                                    |
| 183–307                                    |
| <6:2 MDL-1.4                               |
| 0.6–0.9                                    |
| 2012                                       |
| Herzke et al., 2012                        |
| Textiles                                   |
| 2 36–128                                    |
| 509–840                                    |
| 2.7–25                                     |
| <6:2 MDL                                   |
| 2012                                       |
| Herzke et al., 2012                        |
| Leather                                    |
| 2 <6:2 MDL                                 |
| 22–385                                     |
| <6:2 MDL                                   |
| 2012                                       |
| Herzke et al., 2012                        |
| Furniture material                         |
| 2 <6:2 MDL                                 |
| <6:2 MDL                                   |
| 2012                                       |
| Herzke et al., 2012                        |
| Outdoor clothing                           |
| 7 <6:2 MDL-380                             |
| <6:2 MDL-5100                              |
| <6:2 MDL-7.4                               |
| 2012                                       |
| Brigden et al., 2013                       |
| Outdoor clothing                           |
| 17 <6:2 MDL-7200                           |
| <6:2 MDL-6600                              |
| <6:2 MDL-9.8                               |
| 2012                                       |
| Greenpeace 2013                            |
| Outdoor clothing                           |
| 15 <6:2 MDL-124                           |
| <6:2 MDL-3440                              |
| <6:2 MDL-30.6                              |
| 2011–2012                                  |
| Knepper et al., 2014                       |
| Furniture textiles                         |
| 27 <6:2 MDL-1210                           |
| <6:2 MDL-1000                              |
| <6:2 MDL-5.6                               |
| 2013                                       |
| This study                                 |
| Carpets, mats                              |
| 9 <6:2 MDL-62                             |
| <6:2 MDL-1.6                               |
| <6:2 MDL-0.17                              |
| 2013                                       |
| This study                                 |
| Cotton and leather clothes                 |
| 4 <6:2 MDL-2270                           |
| <6:2 MDL-1.42                              |
| <6:2 MDL-0.18                              |
| 2013                                       |
| This study                                 |
| Food contact material                       |
| 5 <6:2 MDL                                 |
| >6:2 MDL-1.8                               |
| <6:2 MDL                                   |
| 2013                                       |
| This study                                 |

* Concentrations reported in μg m⁻² were transformed to ng g⁻¹ using an average weight of outdoor clothing textiles of 1.5 g dm⁻² derived from Berger and Herzke (2006).*

| Sample                                      |
|---------------------------------------------|
| Sample number (n)                           |
| Concentrations (ng g⁻¹)                     |
| 6:2 FTOH                                    |
| 8:2 FTOH                                    |
| PFOA                                        |
| PFOS                                        |
| Sampling year                               |
| Ref.                                        |
| Import products (Buser and Morf, 2009)       |
| Domestic wastewater influent (Filipovic et al., 2013) |
product emissions. For emissions of FTOHs to air, the releases occurring during disposal (e.g. landfill and incineration) (Weinberg et al., 2011a,b) should also be considered for a more representative comparison with Wang et al. (2012).

Another factor to consider is that textiles, carpets and food contact materials from other countries than China will add to the emissions estimated here. However, since the amount of textiles imported from China makes up approximately 40% of the total import of this product category (SSB, 2008) this could not explain the disagreement between emission estimates in Fig. 1. It is also possible that PFAS residues in consumer products not considered here, including personal care products (Fujii et al., 2013) and textile treatment solutions (Herzké et al., 2012; Knepper et al., 2014; Fiedler et al., 2011; Liu et al., 2014), make a significant contribution to the total diffuse emissions of PFASs.

4. Conclusions and future directions

This study demonstrates that extractable PFASs are present in a wide range of consumer products which are imported from China to Norway in large quantities. However, the concentrations measured here were lower than those reported in historically used products and the majority of consumer products imported from China comply with international regulations (UNEPE, 2009; ECHA, 2013). Based on the comparison with total per capita emission rates, we conclude that the emissions from consumer products imported from China account for 1.5 per cent of the discharges of PFOA to wastewater influents and 0.3 per cent of the emissions of 8:2 FTOH to air in the intermediate emission scenario using Norwegian import data. When using the data from the high emission scenario the relative importance of consumer product emissions increases to 9 and 3 per cent of the total per capita emissions for PFOA and FTOHs respectively. Thus, despite the uncertainty in extractable PFAS concentrations and emission factors we find no support for the hypothesis that diffuse emissions from imported consumer products would be a major ongoing source of PFASs to the environment in non-producing countries such as Norway.

A remaining caveat of constraining diffuse emissions from consumer products is the large variability in extractable PFAS residuals reported in different studies. Although some of this variability can be attributed to a decreasing temporal trend (Liu et al., 2014), biases in the collection of consumer product samples and differences in analytical methods cannot be ruled out. Further work should therefore be devoted to establish robust sampling and analysis protocols for PFASs in consumer product samples including inter-laboratory comparison studies.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2014.12.034.

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