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Perfluorocarboxylic Acid Content in 116 Articles of Commerce

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Abstract

Several recent studies have found elevated levels of perfluorocarboxylic acids (PFCAs) in house dust, suggesting strongly the presence of indoor sources of these compounds. The main goal of this study was to identify and rank potentially important indoor sources by determining the PFCA content in articles of commerce (AOCs). We analyzed 116 AOC samples purchased from retail outlets in the United States between March 2007 and May 2008 by using a newly developed extraction/analytical method. For these 116 samples, the content of perfluorooctanoic acid (PFOA-C8) ranged from non-detectable to 6750 ng/g, whereas the content of total PFCAs (the sum of C5 to C12 acids) ranged from non-detectable to 47100 ng/g. Given the quantities of articles found in typical homes, it is clear that professional carpet-care liquids, pre-treated carpeting, treated floor waxes and sealants, and treated home textile products and upholstery are likely the most important PFCA sources in non-occupational indoor environments. The perfluorochemical-containing AOC market has been in a transition period. Limited data suggest that the PFCA content in AOCs has shown a downward trend overall. However, definitive confirmation of such a trend will require long-term monitoring. More studies are needed to better understand PFCA transport, exposure routes, and ways to reduce exposures in indoor environments.

Acronyms and Abbreviations

| | |
|----------|--|
| AOC | articles of commerce |
| CAS# | chemical abstract service registration number |
| HPLC | high-performance liquid chromatography |
| IAP | internal audit program |
| LC/MS/MS | liquid chromatography / tandem mass spectrometry |
| PFC | perfluorochemical |
| PFCA | perfluorocarboxylic acid |
| PFOA | perfluorooctanoic acid |
| PTFE | polytetrafluoroethylene |
| RCS | recovery check standard |
| RSD | relative standard deviation |
| TPFCA | total perfluorocarbonyl acids (the sum of C5 to C12 PFCAs) |
| WD | wavelength dispersive |
| XRF | X-ray fluorescence |

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1. Introduction

Although man-made perfluorochemicals (PFCs) have been widely used for several decades, their potential impacts on human health and the global environment did not draw much attention until the turn of the century when evidence of their widespread presence in various environmental media, wildlife, and human tissue became clear^[1-3]. Toxicological studies indicate that perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) – the two most extensively studied perfluorinated compounds so far – can cause developmental and systemic toxicity in laboratory animals. Reviews of the existing toxicological data can be found in references 4-6. The potential health risks associated with perfluorocarboxylic acids (PFCAs) have promoted intensive research on the sources, transport, transformation, and distribution of these chemicals and their precursors in environmental media, as well as research related to ways to reduce the health risks. Despite the significant progress that has been made so far, researchers are yet to reach a consensus on what are the most important routes by which the general population is exposed to these chemicals. In particular, there are different opinions on whether PFCA-containing AOCs are significant contributors to the total exposure. For instance, a study conducted by Washburn, et al. in 2005 concluded that exposures to PFOA during consumer use of the articles evaluated in their study were not expected to cause adverse health effects in infants, children, adolescents, and adults, nor result in quantifiable levels of PFOA in human serum^[7]. A more recent study by Fromme, et al. used the data from indoor measurements in Canada and Norway and estimated that, for the general population in Western countries, the inhalation of house dust contributed only 0.6% of the mean PFOA daily intake and 8.2% of the high PFOA daily intake^[8]. By contrast, Tittlemier, et al. identified treated carpeting as the second most important source of exposure for PFOA after ingestion of food^[9]. A study by Trudel, et al. found that the consumption of contaminated food is the most important pathway causing exposure to PFOA, followed by ingestion of dust and inhalation of air in low- and intermediate-exposure scenarios. Their study also found that direct, product-related exposure is dominant in high-exposure scenarios, in which consumers regularly use PFC-containing products, such as impregnation sprays, or have treated carpets in their homes^[10]. Trudel and his co-workers also observed that product-related exposure tends to be more important for PFOA than for PFOS, most likely because PFOS is no longer used in consumer products. It is, therefore, apparent that the paucity of indoor source and exposure data contributes to the significant uncertainty and differences of opinion about the most prevalent exposure routes for these compounds.

The fact that elevated levels of PFCAs have been detected in house dust in Japan^[11], Canada^[12], and the United States^[13] strongly suggests the presence of indoor sources. It is well known that fluorotelomer and fluoropolymer products are sources of PFCAs and that PFCAs may exist in fluorotelomer products as unwanted by-products and in fluoropolymer products as residuals^[14]. Because a broad range of AOCs either contain or are treated with fluorotelomer and fluoropolymer products^[15,16], they can be potential sources of PFCAs. Given that AOCs are often used in close proximity to humans, it is

hypothesized that they can contribute to human exposure to PFCAs either directly (e.g., by dermal contact and hand-to-mouth transfer) or indirectly (e.g., inhalation of suspended particles from treated carpet and other interior surfaces).

There have been several studies of the PFCA content in AOCs, but most of them report a single compound – PFOA. In 2005, Washburn and his colleagues reported the PFOA content in 14 article groups based on theoretical calculations and analytical measurements. Of these groups, pre-treated carpeting and carpeting treated with carpet-care solution had the highest PFOA loadings: 0.2 to 0.6 and 0.2 to 2 mg of PFOA per kg of article, respectively ^[7]. Studies by other researchers reported PFOA content in non-stick cookware, food contact paper, thread sealant tape, and dental floss ^[17-20]. Data for other PFCAs in AOCs are rather scarce. One study by Sinclair, et al. reported the C5 to C12 PFCA content in three brands of popcorn packaging paper ^[20].

The main goal of this study was to identify the major PFCA sources in non-occupational, indoor environments by determining the content of these chemicals in a variety of AOCs and rank them in terms of source strengths. Policy-makers and manufacturers can use the data for risk management purposes. The results also provide a snapshot of the current uses of PFCAs on the market and may serve as baseline data for future, long-term monitoring.

2. Conclusions

We analyzed 116 AOC samples purchased from retail outlets in the United States between March 2008 and May 2008 to determine the extractable content of C5 to C12 PFCAs using a newly developed extraction/analytical method. To the authors' best knowledge, this is the first time that the C5 to C12 PFCA contents in a wide variety of AOCs are been reported. The PFCA contents in these samples cover a broad range, from nondetectable to as high as 6750 ng/g for PFOA and from non-detectable to 47100 ng/g for total PFCAs (i.e., the sum of C5 to C12). In typical American homes with carpeted floors, pre-treated carpet and commercial carpet-care liquids are likely the most significant PFCA sources among the 13 article categories studied. For homes without carpeting, floor waxes and stone/tile/wood sealants that contain fluorotelomers products are important sources of PFCAs. Other potentially important indoor sources include treated home textile, upholstery and apparel, and household carpet/fabric care liquids and foams. The data presented in this report may help explain why PFCAs are frequently detected in house dust. While the exact mechanisms by which PFCAs are transferred from sources to dust are not well characterized, existing data strongly suggest that AOCs may contribute to indoor human exposures to PFCAs either directly (dermal contact and hand-to-mouth activities) or indirectly (inhalation of dust).

3. Recommendations

Further research is needed in the following areas to better understand the effect of PFCA-containing AOCs on human exposure: (1) PFCA transfer from sources to indoor air and surfaces; (2) the relationship between AOCs and inhalation exposure; (3) the significance of dermal exposure; (4) risk management measures for reducing PFCA levels in polluted homes; and (5) monitoring of the market transition on a global scale over an extended period of time.

4. Materials and Methods

4.1 Sample Collection

AOC samples were purchased from retail outlets in the United States. Before collecting samples, a survey was conducted to determine the availability of AOCs that contained or were treated with fluorinated chemicals. Sample candidates were identified based on one of the following claims by the vendors: (a) the article contains fluorinated chemicals identifiable by their trade names (e.g., Scotchgard, GoreTex, and Teflon); (b) the article contains fluorinated chemicals identifiable by the chemical names (e.g., polytetrafluoroethylene or PTFE); or (c) the article was identified as having certain properties that are common for articles treated with fluorinated chemicals (e.g., stain resistant, water repellent, and anti-grease). Sample candidates were purchased from local stores in the Raleigh and Durham areas of North Carolina, in Atlanta and New York City, and from on-line stores. While it was not the goal of this study to obtain statistically representative samples for the entire U.S. market, efforts were made to maximize the representativeness of the samples by considering the following factors whenever applicable: article category, trade name of the fluoropolymer or fluorotelomer product, brand name of the article, price range (high, medium, and low), store type (chain stores, high-end stores, low-end stores, and specialty stores), and country of origin.

4.2 Verifying the Presence of Fluorine

Sample articles obtained from the market were first screened for the presence of fluorine to exclude those with false claims and those that achieved certain surface properties (e.g., anti-grease) without using fluorinated chemicals. Sample articles containing less than 0.01% fluorine by weight were discarded. Typically, 0.05 to 0.5% of the fluorochemical by weight of the article is used to ensure durable repellency^[15]. For articles made of polytetrafluoroethylene (PTFE), such as thread sealant tape and some dental floss, the fluorine content can be greater than 70%. Thus, the 0.01% cut-off provided an adequate safe margin to ensure that all sample articles treated with fluorinated chemicals were included for further analysis. The fluorine content was determined by wavelength dispersive (WD) X-ray fluorescence (XRF) spectrometry. Instrument and operating parameters are provided in Table 4-1.

Table 4-1. Analytical instrument and operating parameters for screen-testing AOC samples for the presence of fluorine by the XRF method ^a

| | |
|------------------------|---|
| Instrument | Panalytical PW2404 Wavelength Dispersive (WD) X-ray Fluorescence (XRF) Spectrometer equipped with the PW2540 Sample Changer |
| Software | SuperQ (Panalytical) for instrument control IQ+ (Panalytical) for calibration and quantification |
| Power of X-ray tube | 4000 watts |
| Measurement atmosphere | Vacuum (<10 mb) or under helium atmosphere |
| Scan method | A continuous scan mode followed by fluorine-specific data collection at the peak fluorine wavelength for an additional 10 second measurement. |

^a Liquid samples were tested on filters by wetting a 47-mm paper filter (Whatman) with approximately 0.5 mL sample, and then air drying the filter under an aluminum foil cover.

4.3 Sample Preparation, Handling and Storage

Upon receipt, solid articles (except cookware) were cut into smaller subsections for storage or extraction. Each AOC was divided into at least three subsections with a 60-mm, tungsten carbide steel, rotary cutter and stainless steel scissors. These subsections were used to cut specific-sized coupons (e.g., 4 ×4, 6×6, and 10×10 cm, depending on sample weight per unit area) as needed for extractions. Subsections for extraction were placed in a desiccator for a minimum of eight hours and then weighed prior to extraction. Non-stick cookware remained in its original package until extraction.

Liquid samples in bottles were subdivided into at least three 30-mL polypropylene vials. For aerosol cans, the liquids were shaken and then collected in a 50-mL beaker by gently pressing the release button. The liquids were then divided into aliquots and stored in polypropylene vials. Samples for extraction were stored in the refrigerator until needed for analysis.

All archived samples were individually wrapped in three layers of aluminum foil, placed in a sealed plastic bag, and stored in a climate-controlled room.

4.4 Sample Extraction and Analysis

Coupons of solid samples (approximately 1 g) were weighed and placed in a 50-mL, polyethylene, centrifuge vial with 45 mL of HPLC-grade methanol spiked with 100 µL of recovery check standard {RCS, 2 ng/µL perfluoro-*n*-[1, 2-¹³C₂] decanoic acid} and then extracted for 24 hours with a Nutating Mixer (Model VSN-5, PRO Scientific, Inc., CT, USA). The extract was transferred to a 170-mL borosilicate glass tube and concentrated to approximately 1 mL under a nitrogen atmosphere by using a RapidVap

N₂ Evaporation System (Model 791000, LabConco, MO, USA), which was modified at the factory to remove all Teflon parts and coatings. The blow-down sample was transferred from the tube to a 10-mL volumetric flask through a 0.1 µm Anotop syringe filter (Whatman International, Madestone, England); the tube was rinsed five times with a solution consisting of 60% (v/v) methanol and 40% (v/v) 2 mN ammonium acetate aqueous solution (hereafter referred to as the 60:40 solution). The rinse liquids were filtered and combined with the blow-down sample. After adding 100 µL of the internal standard {0.5 ng/µL perfluoro-*n*-[1, 2, 3, 4-¹³C₄] octanoic acid}, the sample was brought to 10-mL with the 60:40 solution and sonicated for 10 minutes before LC/MS/MS analysis.

A different extraction procedure was used for cookware, because the interior coatings were difficult to remove from the metal base. Cookware was extracted by covering the entire cooking surface with 100 to 150 mL of methanol spiked with 100 µL of the recovery check standard to a depth of approximately 0.3 mm. To minimize solvent evaporation during extraction, the opening of the cookware was tightly sealed with aluminum foil by compressing the foil to the inside and outside walls of the cookware edge to a depth of approximately 0.5 cm. The static extraction was conducted at room temperature. After 24 hours, the extract was collected from the cookware and concentrated to 1 mL by the procedure described above.

All solid AOC samples were extracted by a single-step, 24-hour extraction, and the extraction efficiency, as determined by consecutive extractions, ranged from 70% to 100% except for non-stick cookware, which had an extraction efficiency of 46% for PFOA. The analytical results were reported as “extractable PFCAs.”

For liquid samples, approximately 1 mL of sample were weighed, spiked with 100 µL of 2 ng/µL of the recovery check standard, and diluted to 25 mL with the 60:40 solution. The diluted samples were sonicated for 10 minutes and then filtered with a 50-mL tube-top filter (Corning, Inc., NY, USA; 0.22-µm pore size). Ten milliliters (10 mL) of the filtrate were transferred into a 10-mL volumetric flask and spiked with 100 µL of the internal standard. The final solution was sonicated for 10 minutes before LC/MS/MS analysis. For samples with high levels of PFCAs, a second dilution was needed before adding the recovery check standard.

Sample quantification was conducted using an Agilent 1100 HPLC equipped with an Applied Biosystem API 3200 Triple Quadrupole Mass Spectrometer with a Turbo V ion-spray interface. The instrument was calibrated for eight PFCA homologues (Table 4-2) plus the recovery check standard at eight concentration levels in the concentration range of 0.3 to 100 ng/mL with triplicate injections. The instrument detection limits for individual PFCAs in the injection sample were ≤0.05 ng/mL. The method detection limits were 1.0 to 3.9 ng/g for solid AOCs and 1.1 to 6.8 ng/g for liquid AOCs. The practical quantification limit for the injection sample was 0.3 ng/mL, which is equivalent to 3 ng/g for solid AOCs and 7.5 ng/g for liquid AOCs.

The extraction and analytical methods described above have been evaluated and are reported elsewhere ^[21].

Table 4-2. Analyte names, chemicals formulas, and chemical abstracts service registration numbers (CAS#)

| Analyte name | Chemical formula | CAS# |
|--------------------------|--------------------------------------|-----------|
| perfluoropentanoic acid | C ₅ F ₁₁ COOH | 2706-90-3 |
| perfluorohexanoic acid | C ₆ F ₁₃ COOH | 307-24-4 |
| perfluoroheptanoic acid | C ₇ F ₁₅ COOH | 375-85-9 |
| perfluorooctanoic acid | C ₈ F ₁₇ COOH | 335-67-1 |
| perfluorononanoic acid | C ₉ F ₁₉ COOH | 375-95-1 |
| perfluorodecanoic acid | C ₁₀ F ₂₁ COOH | 335-76-2 |
| perfluoroundecanoic acid | C ₁₁ F ₂₃ COOH | 2058-94-8 |
| perfluorododecanoic acid | C ₁₂ F ₂₅ COOH | 307-55-1 |

4.5 Quality Assurance and Quality Control

A quality assurance project plan (QAPP) was developed before the start of the project. The acceptance criterion for the calibration curve was that the coefficient of determination (r^2) be no less than 0.99. The internal audit program (IAP) standard, which contains at least four of the calibrated PFCAs using a different chemical source, was prepared by someone other than the person who prepared the calibration standards. The analyst who conducted the calibration received the IAP standard without knowing the concentrations. IAP standards were analyzed after each calibration as a measurement of calibration verification. The criterion for acceptance was that the calculated concentration and measured IAP standard using the calibration had to be within 15% of each other. Daily calibration check (DCC) standards, approximately 5 ng/mL for each analyte, were analyzed to evaluate the LC/MS/MS performance. Analytical results of a sample batch were considered acceptable only if the percent recovery of the DCC was within $100 \pm 15\%$ and the percent relative standard deviation (%RSD) of DCCs was within $\pm 15\%$. All samples and standards were injected in triplicate.

Solvents, glassware, gloves, and the HPLC system were routinely checked for PFCA contamination. A solvent blank was prepared with each set of standards and samples to assess the solvent and the HPLC system.

Each AOC sample was extracted in duplicate for LC/MS/MS analysis. The analytical results were considered acceptable when the measured concentrations were in the calibration range, the RSD for duplicates was within 20%, and the recovery of the recovery check standard was within $100 \pm 20\%$.

5. Results

5.1 Statistics of AOC Samples

A total of 130 AOC samples containing at least 0.01% (w/w) fluorine were obtained between March 2007 and May 2008. The dates the samples were manufactured were unknown, because the product labels did not include that information. Breakdowns of these samples by article category are given in Table 5-1. Sample descriptions and conversion factors are presented in the Appendix (Table A-1).

Table 5-1. Sample breakdowns by article category

| Category ID | Category name | Samples |
|-------------|--|---------|
| A | Pre-treated carpeting | 9 |
| B | Commercial carpet-care liquids | 9 |
| C | Household carpet/fabric-care liquids and foams | 12 |
| D | Treated apparel | 16 |
| E | Treated home textile and upholstery | 14 |
| F | Treated non-woven medical garments | 5 |
| G | Treated floor waxes and stone/wood sealants | 11 |
| H | Treated food contact paper | 5 |
| I | Membranes for apparel | 10 |
| J | Thread sealant tapes and pastes | 10 |
| K | Non-stick cookware | 14 |
| L | Dental floss and plaque removers | 8 |
| M | Miscellaneous ^a | 7 |

^a Includes four car-care products, two boat-care products, one deck cleaner, and one dry sack for outdoor use.

The samples were divided approximately equally between domestic and imported products: United States – 58, China – 35, Thailand – 5, Dominican Republic – 3, France – 3, Malaysia – 3, Mexico – 3, Bangladesh – 2, Canada – 2, Indonesia – 2, Ireland – 2, Sri Lanka – 2, Vietnam – 2, Brazil – 1, Colombia – 1, England – 1, Italy – 1, Nicaragua – 1, and Pakistan – 1. The origins of two samples could not be identified. According to the product labels, six imported articles used materials or contain components made in the United States, and one domestic product used imported material.

5.2 Data Quality

Samples were analyzed in batches. Each batch included four to seven AOC samples in duplicate, one solvent blank, one field blank, and two daily calibration check standards. For individual analytes, the relative standard deviation for duplicate samples had to be

within 20% to be accepted. The estimated precision of the results was $100 \pm 20\%$ based on duplicate samples, and the accuracy was $100 \pm 20\%$ based on the recovery of RCS. The data presented were not adjusted for recovery of RCS. The RCS recoveries for individual samples are available in the last column in Tables 5-2 through 5-14. Of the 130 AOC samples, 116 were analyzed successfully. For the 116 sets of valid data, the average percent recovery of RCS was 97.9%. The results of the remaining 14 samples failed to meet the data quality requirements after three or more trials, and the data were discarded. Low recovery (i.e., <80%) for the recovery check standard was the common cause of the failures. These samples are identified in the footnotes of Tables 5-2 through 5-14. They belong to eight article categories, and no obvious trends could be identified.

5.3 Extractable PFCA Content in AOC Samples

Complete data for PFCA content in individual samples is presented in Tables 5-2 through 5-14, in which the following abbreviations and fonts are used:

- BDL = result below instrument detection limit,
- NR = not reported (i.e., the result does not meet data quality requirements),
- RCS = recovery check standard; RCS percent recovery is the average of duplicate or triplicate samples, and
- Italics* = result below practical quantification limit.

Figure 5-1 shows the distribution of the total amount of PFCAs (TPFCA, the sum of C5 to C12 PFCAs) for the 116 samples. Note that, for data comparability, the results for non-stick cookware have been converted from (ng/cm^2) to (ng/g) by assuming an average coating thickness of $50 \mu\text{m}$ and a PTFE density of $2.2 \text{ g}/\text{cm}^3$ ^[17].

Table 5-2. Extractable PFCAs in pre-treated carpeting (ng/g fiber) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|-----------------------|----------------------|------------------|
| A-1 | NR | 3.98×10 ¹ | 1.41×10 ¹ | 1.04×10 ¹ | 6.31×10 ⁰ | 5.29×10 ⁰ | 2.34×10 ⁰ | BDL | 81.7 |
| A-2 | BDL | BDL | NR | BDL | BDL | BDL | 6.17×10 ⁻¹ | BDL | 91.4 |
| A-3 | BDL | BDL | NR | 8.39×10 ⁰ | NR | 7.26×10 ⁰ | NR | 5.19×10 ⁰ | 84.3 |
| A-4 | BDL | BDL | BDL | BDL | BDL | BDL | 6.59×10 ⁻¹ | BDL | 90.8 |
| A-5 ^b | NR | NR | 7.46×10 ¹ | 4.62×10 ² | 7.26×10 ¹ | 8.84×10 ¹ | 2.01×10 ¹ | NR | 81.9 |
| A-6 ^c | BDL | NR | NR | 5.77×10 ⁰ | 6.56×10 ⁰ | BDL | 3.64×10 ⁰ | 2.10×10 ⁰ | 84.6 |
| A-8 ^b | 2.16×10 ² | 2.42×10 ² | 5.16×10 ² | 2.98×10 ² | 2.92×10 ² | 1.46×10 ² | 5.22×10 ¹ | 6.35×10 ¹ | 92.4 |
| A-9 | 1.15×10 ¹ | 1.92×10 ¹ | 4.30×10 ¹ | 1.99×10 ¹ | 2.07×10 ¹ | 1.84×10 ¹ | 1.23×10 ¹ | 4.20×10 ¹ | 84.7 |

^a Results for sample A-7 failed to meet data quality requirements;

^b This sample was extracted in triplicate;

^c According to the vendor, sample A-6 is a replacement of A-5.

Table 5-3. Extractable PFCAs in commercial carpet-care liquids (ng/g)

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|------------------|
| B-1 | 1.73×10^3 | 5.20×10^3 | 1.41×10^4 | 6.75×10^3 | 8.86×10^3 | 4.38×10^3 | 4.00×10^3 | 2.15×10^3 | 100 |
| B-2 | 1.61×10^1 | 1.75×10^2 | 5.18×10^1 | 5.96×10^2 | NR | 1.67×10^2 | 2.80×10^1 | NR | 84.1 |
| B-3 | 2.19×10^1 | 4.48×10^1 | 4.95×10^1 | 5.01×10^1 | 5.61×10^1 | NR | NR | NR | 100 |
| B-4 ^a | 1.23×10^2 | 1.00×10^3 | 7.47×10^1 | 5.99×10^2 | NR | NR | NR | NR | 106 |
| B-5 | 1.14×10^1 | 3.09×10^1 | 1.98×10^1 | 1.91×10^1 | 6.38×10^0 | BDL | NR | NR | 99.9 |
| B-6 | 1.94×10^3 | 5.25×10^3 | 1.3×10^4 | 5.01×10^3 | 8.46×10^3 | 2.93×10^3 | 3.05×10^3 | 9.57×10^2 | 90.8 |
| B-7 ^b | 3.63×10^2 | 9.28×10^2 | 2.56×10^3 | 1.84×10^3 | NR | 1.33×10^3 | 8.44×10^2 | NR | 101 |
| B-8 ^a | 3.77×10^2 | 1.84×10^3 | 1.48×10^3 | 1.72×10^3 | 1.30×10^3 | 8.41×10^2 | 5.07×10^2 | 4.43×10^2 | 96.3 |
| B-9 ^b | NR | 2.20×10^1 | 1.42×10^1 | 2.55×10^1 | 1.75×10^1 | 2.10×10^1 | 1.62×10^1 | 8.52×10^0 | 101 |

^a Samples B-4 and B-8 are of same brand, but were purchased 1 year apart;

^b Samples B-7 and B-9 are of same brand, but were purchased 1 year apart;

Table 5-4. Extractable PFCAs in household carpet/fabric care liquids and foams (ng/g)

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|------------------|
| C-1 | BDL | BDL | 1.14×10^1 | 6.97×10^0 | 3.09×10^0 | 1.14×10^0 | BDL | NR | 120 |
| C-2 | BDL | BDL | BDL | BDL | 1.63×10^1 | 3.25×10^1 | 4.29×10^1 | BDL | 105 |
| C-3 | 1.40×10^2 | 1.09×10^3 | 2.50×10^3 | 1.18×10^3 | 1.71×10^3 | 6.76×10^2 | 8.01×10^2 | 3.28×10^2 | 103 |
| C-4 | NR | 7.55×10^1 | NR | 6.66×10^2 | NR | 1.04×10^2 | BDL | BDL | 98.3 |
| C-5 | NR | 1.95×10^2 | BDL | BDL | BDL | BDL | BDL | BDL | 93.4 |
| C-6 | NR | 3.90×10^1 | BDL | 1.09×10^1 | BDL | BDL | BDL | BDL | 103 |
| C-7 | NR | BDL | BDL | BDL | BDL | BDL | BDL | BDL | 97.6 |
| C-8 | NR | BDL | BDL | BDL | BDL | BDL | BDL | BDL | 81.0 |
| C-9 | BDL | 1.73×10^2 | BDL | 7.07×10^2 | 1.99×10^1 | 2.89×10^2 | 4.51×10^1 | BDL | 94.6 |
| C-10 | BDL | 4.18×10^1 | BDL | 8.79×10^1 | 2.15×10^1 | 3.36×10^1 | 1.21×10^1 | 1.69×10^1 | 105 |
| C-11 | 1.84×10^1 | 5.61×10^1 | 1.21×10^1 | 1.37×10^2 | 5.70×10^0 | 4.00×10^1 | BDL | 7.91×10^0 | 86.3 |
| C-12 | NR | BDL | BDL | 6.64×10^1 | NR | 1.86×10^1 | BDL | BDL | 108 |

Table 5-5. Extractable PFCAs in treated apparel (ng/g)^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|-----------------------|----------------------|----------------------|----------------------|-----------------------|----------------------|-----------------------|-----------------------|------------------|
| D-1 ^b | 7.26×10 ¹ | 1.53×10 ² | 2.21×10 ² | 1.14×10 ² | 7.72×10 ¹ | 5.37×10 ¹ | 3.94×10 ¹ | 4.10×10 ¹ | 102 |
| D-2 | 5.41×10 ⁻² | 1.43×10 ⁰ | 1.08×10 ⁰ | NR | 2.86×10 ⁻¹ | NR | 5.04×10 ⁻¹ | 4.27×10 ⁻¹ | 101 |
| D-3 | 1.64×10 ¹ | 4.32×10 ¹ | 6.49×10 ¹ | 1.61×10 ² | 2.35×10 ² | 6.92×10 ¹ | 6.15×10 ¹ | 2.12×10 ¹ | 88.9 |
| D-4 | NR | 2.70×10 ¹ | 8.96×10 ⁰ | 3.80×10 ¹ | 3.85×10 ⁰ | 2.20×10 ¹ | 1.39×10 ⁰ | 1.45×10 ¹ | 99.9 |
| D-5 | NR | 2.72×10 ¹ | NR | 3.20×10 ¹ | 5.97×10 ⁰ | 1.35×10 ¹ | 3.60×10 ⁰ | 8.04×10 ⁰ | 99.3 |
| D-6 | 1.08×10 ⁰ | 2.71×10 ⁰ | 3.11×10 ⁰ | 5.44×10 ⁰ | 2.59×10 ⁰ | 3.26×10 ⁰ | NR | NR | 108 |
| D-7 | 3.94×10 ⁰ | 3.83×10 ¹ | 8.05×10 ⁰ | 5.55×10 ¹ | 4.13×10 ⁰ | 2.83×10 ¹ | 1.94×10 ⁰ | 1.30×10 ¹ | 103 |
| D-8 | 6.77×10 ⁰ | 2.91×10 ¹ | 1.27×10 ¹ | 8.55×10 ¹ | 8.73×10 ⁰ | 3.93×10 ¹ | 3.61×10 ⁰ | 6.47×10 ⁰ | 103 |
| D-10 | NR | 6.37×10 ¹ | 1.87×10 ¹ | 1.09×10 ² | 1.36×10 ¹ | 4.68×10 ¹ | 5.17×10 ⁰ | NR | 96.0 |
| D-11 | NR | 2.83×10 ¹ | 4.92×10 ⁰ | 4.43×10 ¹ | 3.55×10 ⁰ | 2.39×10 ¹ | 1.81×10 ⁰ | 9.42×10 ⁰ | 80.0 |
| D-12 | BDL | 4.21×10 ⁰ | 4.73×10 ⁰ | 2.76×10 ¹ | 6.54×10 ¹ | 1.66×10 ¹ | 2.14×10 ¹ | NR | 113 |
| D-13 | 1.75×10 ⁰ | 1.40×10 ¹ | 4.66×10 ⁰ | 6.93×10 ¹ | 6.28×10 ⁰ | 2.32×10 ¹ | 2.00×10 ⁰ | 5.36×10 ⁰ | 89.3 |
| D-14 | 4.26×10 ⁰ | 2.61×10 ¹ | 7.12×10 ⁰ | NR | NR | 2.30×10 ¹ | 2.03×10 ⁰ | NR | 96.0 |
| D-15 | 3.55×10 ⁰ | 4.61×10 ⁰ | 8.58×10 ⁰ | 1.31×10 ¹ | 5.86×10 ⁰ | 8.06×10 ⁰ | 1.38×10 ⁰ | 3.67×10 ⁰ | 86.7 |

^a Results for samples D-9 and D-16 failed to meet data quality requirements;

^b This sample was extracted in triplicate.

Table 5-6. Extractable PFCAs in treated home textile and upholstery (ng/g)

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|--------------------|--------------------|-----------------------|-----------------------|-----------------------|--------------------|-----------------------|-----------------------|------------------|
| E-1 | BDL | BDL | NR | 5.83×10^0 | BDL | 2.66×10^0 | NR | NR | 111 |
| E-2 | BDL | 1.19×10^0 | 1.22×10^0 | 3.26×10^0 | 1.36×10^0 | BDL | BDL | NR | 108 |
| E-3 | 3.09×10^1 | 6.22×10^1 | 9.46×10^1 | 2.93×10^2 | 3.18×10^2 | 1.80×10^2 | 9.80×10^1 | NR | 97.3 |
| E-4 | BDL | BDL | 1.80×10^0 | 3.49×10^0 | 2.01×10^0 | BDL | BDL | NR | 91.1 |
| E-5 | BDL | BDL | 4.09×10^{-1} | 6.12×10^{-1} | 3.96×10^{-1} | BDL | BDL | BDL | 89.2 |
| E-6 | BDL | 2.96×10^0 | 1.76×10^0 | 2.18×10^0 | NR | 2.20×10^0 | 6.80×10^{-1} | 7.78×10^{-1} | 102 |
| E-7 | 2.16×10^1 | 6.80×10^1 | 9.66×10^1 | 3.30×10^2 | 2.13×10^2 | 1.25×10^2 | 4.57×10^1 | 4.30×10^1 | 100 |
| E-8 | BDL | 1.05×10^1 | NR | 1.88×10^1 | 7.15×10^0 | 8.99×10^0 | NR | NR | 106 |
| E-9 ^a | 9.47×10^1 | 2.38×10^2 | 5.15×10^2 | 4.38×10^2 | 4.37×10^2 | 2.47×10^2 | 1.86×10^2 | 1.10×10^2 | 110 |
| E-10 | BDL | BDL | 1.11×10^0 | NR | NR | 2.42×10^0 | BDL | 9.34×10^{-1} | 99.0 |
| E-11 | BDL | 8.56×10^0 | 3.14×10^0 | 2.75×10^1 | 3.95×10^0 | 8.37×10^0 | BDL | BDL | 97.2 |
| E-12 | BDL | BDL | 7.96×10^1 | 8.50×10^1 | 3.41×10^1 | BDL | BDL | 3.73×10^1 | 97.9 |
| E-13 | 1.43×10^0 | 3.87×10^0 | 6.03×10^0 | 1.17×10^1 | 4.37×10^0 | 6.55×10^0 | 5.63×10^{-1} | NR | 103 |
| E-14 | BDL | 1.59×10^0 | 1.23×10^0 | 3.83×10^0 | 1.78×10^0 | 2.29×10^0 | 1.09×10^0 | 1.13×10^0 | 104 |

^a This sample was extracted in triplicate.

Table 5-7. Extractable PFCAs in treated non-woven medical garments (ng/g)

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|------------------|
| F-1 | 4.31×10^0 | 1.45×10^1 | 2.05×10^1 | 4.62×10^1 | 7.82×10^1 | 2.90×10^1 | 2.74×10^1 | 1.05×10^1 | 85.8 |
| F-2 | 4.26×10^0 | NR | 1.84×10^1 | 4.71×10^1 | 8.21×10^1 | 2.00×10^1 | 2.48×10^1 | 8.72×10^0 | 86.9 |
| F-3 | BDL | NR | 9.03×10^0 | 6.07×10^1 | 6.33×10^0 | 1.74×10^1 | NR | 5.30×10^0 | 80.4 |
| F-4 | 7.86×10^2 | 5.98×10^2 | 5.06×10^2 | 3.69×10^2 | 3.34×10^2 | 2.18×10^2 | 1.73×10^2 | 8.89×10^1 | 101 |
| F-5 | 6.02×10^0 | 1.43×10^1 | 2.24×10^1 | 8.42×10^1 | 1.08×10^2 | 6.42×10^1 | 4.17×10^1 | 2.69×10^1 | 101 |

Table 5-8. Extractable PFCAs in floor waxes and stone/tile/wood sealants (ng/g) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|------------------|
| G-1 | NR | 4.83×10 ¹ | 6.39×10 ¹ | 4.48×10 ¹ | 5.04×10 ¹ | 1.91×10 ¹ | 2.21×10 ¹ | 1.41×10 ¹ | 98.2 |
| G-2 ^b | 7.19×10 ⁰ | 1.53×10 ¹ | 2.14×10 ¹ | 7.50×10 ⁰ | 4.19×10 ⁰ | 3.56×10 ⁰ | 1.65×10 ⁰ | 2.97×10 ⁰ | 95.4 |
| G-3 ^b | 8.19×10 ⁰ | 1.41×10 ¹ | 2.75×10 ¹ | 1.32×10 ¹ | 1.23×10 ¹ | 5.32×10 ⁰ | 2.12×10 ⁰ | 3.33×10 ⁰ | 109 |
| G-4 ^b | 8.62×10 ⁰ | 1.91×10 ¹ | 2.73×10 ¹ | 1.56×10 ¹ | 1.17×10 ¹ | NR | 2.10×10 ⁰ | NR | 116 |
| G-6 | BDL | 4.07×10 ¹ | 6.25×10 ¹ | 3.69×10 ¹ | 4.76×10 ¹ | 2.38×10 ¹ | 2.22×10 ¹ | 2.37×10 ¹ | 101 |
| G-7 | NR | 1.17×10 ³ | 1.05×10 ² | 8.05×10 ² | 3.31×10 ¹ | 3.30×10 ² | BDL | BDL | 90.2 |
| G-8 | 1.12×10 ² | 7.72×10 ² | 3.13×10 ² | 4.77×10 ² | 1.55×10 ² | 2.65×10 ² | BDL | NR | 94.4 |
| G-9 | BDL | BDL | BDL | 4.35×10 ¹ | 3.30×10 ¹ | 3.44×10 ¹ | BDL | BDL | 99.8 |
| G-10 | 3.15×10 ² | 5.38×10 ³ | 5.46×10 ² | 3.72×10 ³ | 2.60×10 ² | 1.58×10 ³ | 4.15×10 ¹ | 4.70×10 ² | 92.3 |
| G-11 | 3.73×10 ² | 1.16×10 ³ | 1.56×10 ³ | 1.21×10 ³ | 9.39×10 ² | 6.32×10 ² | 3.75×10 ² | 2.84×10 ² | 110 |

^a Results for sample G-5 failed to meet data quality requirements;

^b Samples G-2, G-3, and G-4 could not be filtered after dilution. They were applied on aluminum foil as a thin layer and dried under cover. The dry films were then extracted as solid samples. The potential PFCA loss during the curing period was not evaluated but is expected to be insignificant.

Table 5-9. Extractable PFCAs in food contact paper (ng/g)^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|--------------------|--------------------|--------------------|--------------------|--------------------|--------------------|-----|--------------------|------------------|
| H-2 | BDL | BDL | BDL | BDL | 2.25×10^0 | BDL | BDL | BDL | 106 |
| H-3 | BDL | 1.17×10^1 | BDL | BDL | BDL | BDL | BDL | BDL | 86.9 |
| H-4 | BDL | NR | BDL | 1.04×10^2 | BDL | 7.02×10^1 | BDL | 5.40×10^1 | 105 |
| H-5 | 2.21×10^2 | 4.43×10^3 | 2.85×10^3 | 4.64×10^3 | 1.53×10^1 | BDL | BDL | BDL | 103 |

^a Results for sample H-1 failed to meet data quality requirements.

Table 5-10. Extractable PFCAs in membranes for apparel (ng/g) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|------------------|
| I-1 | BDL | 1.72×10 ¹ | 1.15×10 ¹ | 7.70×10 ¹ | 5.95×10 ⁰ | 2.43×10 ¹ | 3.24×10 ⁰ | NR | 109 |
| I-2 | BDL | NR | 3.23×10 ⁰ | 9.15×10 ⁰ | 1.84×10 ⁰ | 4.33×10 ⁰ | 1.10×10 ⁰ | 3.26×10 ⁰ | 108 |
| I-4 | BDL | NR | NR | 2.96×10 ¹ | 1.06×10 ¹ | 7.59×10 ⁰ | 6.01×10 ⁰ | NR | 108 |
| I-5 | 8.23×10 ⁰ | 5.09×10 ¹ | 1.21×10 ¹ | 1.63×10 ² | 1.28×10 ¹ | 7.22×10 ¹ | NR | 2.36×10 ¹ | 113 |
| I-7 | BDL | NR | 1.09×10 ¹ | 4.35×10 ¹ | 8.08×10 ⁰ | 1.35×10 ¹ | NR | 6.29×10 ⁰ | 82.4 |
| I-8 | 3.33×10 ⁰ | 3.03×10 ¹ | 5.46×10 ⁰ | 8.26×10 ¹ | 5.97×10 ⁰ | 2.77×10 ¹ | 2.02×10 ⁰ | NR | 87.7 |
| I-9 ^b | BDL | NR | 4.66×10 ⁰ | 1.04×10 ² | 5.37×10 ⁰ | 4.19×10 ¹ | 3.41×10 ⁰ | 8.59×10 ⁰ | 111 |
| I-10 | BDL | 2.05×10 ¹ | 5.02×10 ⁰ | 7.30×10 ¹ | 5.95×10 ⁰ | 2.92×10 ¹ | 3.03×10 ⁰ | 9.82×10 ⁰ | 112 |

^a Results for sample I-3 and I-6 failed to meet data quality requirements.

^b This sample was extracted in triplicate.

Table 5-11. Extractable PFCAs in thread seal tapes and pastes (ng/g) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|--------------------|--------------------|-----------------------|--------------------|--------------------|--------------------|--------------------|--------------------|------------------|
| J-1 | BDL | BDL | 2.85×10^0 | 4.43×10^0 | 2.82×10^0 | BDL | 1.69×10^0 | NR | 99.2 |
| J-2 | BDL | BDL | BDL | 5.35×10^0 | 2.31×10^0 | BDL | 1.42×10^0 | NR | 102 |
| J-3 | BDL | BDL | 7.32×10^{-1} | NR | NR | NR | BDL | NR | 82.9 |
| J-4 | BDL | BDL | NR | NR | NR | NR | BDL | NR | 92.0 |
| J-5 | BDL | BDL | NR | 2.30×10^1 | 2.28×10^0 | BDL | 1.38×10^0 | 2.14×10^0 | 96.1 |
| J-6 | BDL | NR | NR | 1.44×10^3 | 2.80×10^0 | BDL | BDL | BDL | 105 |
| J-7 | BDL | 1.97×10^1 | 2.87×10^1 | 3.49×10^3 | 4.56×10^0 | BDL | BDL | BDL | 109 |
| J-9 | BDL | BDL | BDL | BDL | 1.67×10^1 | BDL | 9.86×10^0 | 1.25×10^1 | 93.0 |
| J-10 | 9.50×10^1 | 3.26×10^1 | 3.24×10^1 | 3.89×10^1 | 4.06×10^1 | 3.62×10^1 | 4.06×10^1 | 3.75×10^1 | 87.0 |

^a Results for sample J-8 failed to meet data quality requirements.

Table 5-12. Extractable PFCAs in non-stick cookware (ng/cm² coated surface)^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|------------------|
| K-1 | BDL | BDL | BDL | BDL | 4.89×10^{-3} | BDL | 2.89×10^{-3} | 3.70×10^{-3} | 103 |
| K-3 | 1.30×10^{-2} | BDL | 1.55×10^{-3} | NR | 2.47×10^{-3} | BDL | 1.48×10^{-3} | 1.88×10^{-3} | 96.9 |
| K-4 | NR | NR | NR | 2.09×10^{-2} | NR | NR | NR | NR | 92.0 |
| K-5 | NR | NR | NR | 4.74×10^{-2} | NR | NR | NR | NR | 99.4 |
| K-6 | BDL | BDL | BDL | BDL | BDL | BDL | BDL | 1.37×10^{-2} | 98.2 |
| K-7 | BDL | BDL | BDL | BDL | BDL | BDL | BDL | 1.07×10^{-2} | 94.5 |
| K-8 | BDL | 3.71×10^{-3} | BDL | BDL | BDL | BDL | BDL | 3.63×10^{-3} | 96.4 |
| K-9 | BDL | BDL | 2.55×10^{-3} | 4.02×10^{-3} | 4.31×10^{-3} | BDL | 2.55×10^{-3} | 3.23×10^{-3} | 98.3 |
| K-10 | NR | BDL | 4.01×10^{-3} | BDL | 8.50×10^{-3} | BDL | 3.89×10^{-3} | 4.81×10^{-3} | 99.5 |
| K-11 | BDL | BDL | NR | 7.57×10^{-3} | 5.66×10^{-3} | BDL | 3.12×10^{-3} | 3.96×10^{-3} | 95.6 |
| K-12 | BDL | BDL | 8.69×10^{-3} | BDL | BDL | BDL | BDL | 8.57×10^{-3} | 92.2 |
| K-13 | 1.87×10^{-2} | 1.31×10^{-2} | 1.72×10^{-2} | 1.21×10^{-2} | 9.85×10^{-3} | 9.56×10^{-3} | 9.95×10^{-3} | 1.85×10^{-2} | 95.5 |
| K-14 | NR | 7.02×10^{-3} | 9.00×10^{-3} | 1.25×10^{-2} | 2.67×10^{-3} | BDL | 2.60×10^{-3} | 9.06×10^{-3} | 114 |

^a Results for sample K-2 failed to meet data quality requirements.

Table 5-13. Extractable PFCAs in dental floss and plaque removers (ng/g) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|--------------------|-----|-----------------------|--------------------|-----------------------|-----|--------------------|--------------------|------------------|
| L-1 | BDL | BDL | BDL | NR | 9.39×10^{-1} | BDL | BDL | NR | 102 |
| L-2 | 2.50×10^0 | NR | 6.30×10^0 | 5.48×10^0 | NR | BDL | 2.87×10^0 | NR | 81.4 |
| L-4 | BDL | BDL | 6.20×10^{-1} | NR | 7.02×10^{-1} | BDL | BDL | 1.43×10^0 | 118 |
| L-5 | BDL | BDL | BDL | 9.67×10^1 | BDL | BDL | BDL | BDL | 112 |
| L-7 | BDL | BDL | 3.99×10^0 | BDL | 5.81×10^0 | BDL | 3.43×10^0 | 4.40×10^0 | 102 |
| L-8 | BDL | BDL | 1.71×10^0 | 4.58×10^1 | 2.25×10^0 | BDL | 1.33×10^0 | 1.69×10^0 | 106 |

^a Results for sample L-3 and L-6 failed to meet data quality requirements.

Table 5-14. Extractable PFCAs in miscellaneous AOC samples (ng/g) ^a

| Sample ID | C5 | C6 | C7 | C8 | C9 | C10 | C11 | C12 | RCS Recovery (%) |
|-----------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|------------------|
| M-1 | 1.08×10 ¹ | 2.09×10 ¹ | 6.01×10 ¹ | 2.49×10 ¹ | 3.28×10 ¹ | 1.44×10 ¹ | 1.47×10 ¹ | 1.09×10 ¹ | 84.9 |
| M-2 | BDL | BDL | BDL | BDL | 8.26×10 ¹ | BDL | BDL | BDL | 93.0 |
| M-3 | BDL | 1.73×10 ² | 2.05×10 ² | 1.25×10 ² | 6.95×10 ¹ | BDL | BDL | BDL | 89.7 |
| M-5 | BDL | BDL | 2.98×10 ⁰ | BDL | BDL | BDL | BDL | 2.97×10 ⁰ | 80.7 |

^a Results for sample M-4, M-6 and L-7 failed to meet data quality requirements.

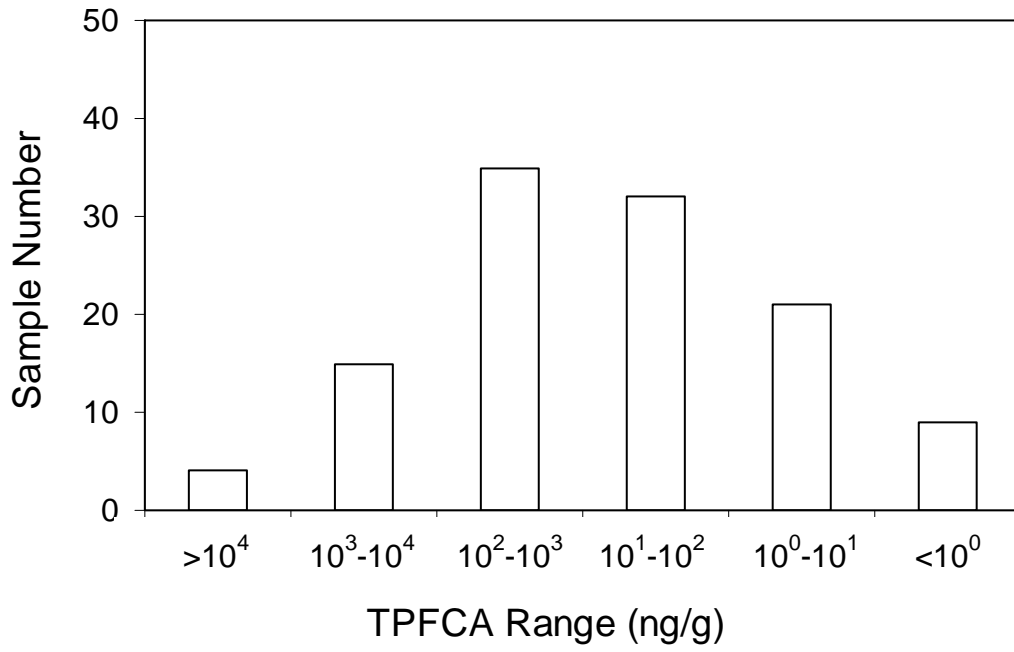


Figure 5-1. Distribution of total extractable PFCAs for 116 AOC samples.

5.4 Statistics by sample category

The ranges, arithmetic means, and medians of PFCA content for individual article categories are presented in Figures 5-2 through 5-14. The overall TPFCA range is from non-detectable to 47100 ng/g; the range for PFOA is from non-detectable to 6750 ng/g. For most article categories, the means were greater than the medians in most cases, indicating that the PFCA content in a small number of samples was significantly higher than in the rest of samples.

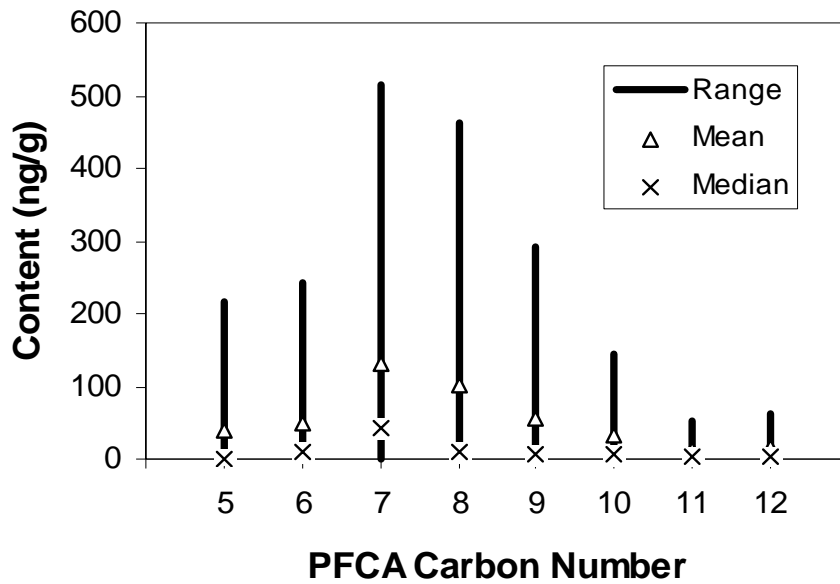


Figure 5-2. Ranges, arithmetic means, and medians for PFCAs in the fiber of pre-treated carpeting (N = 8).

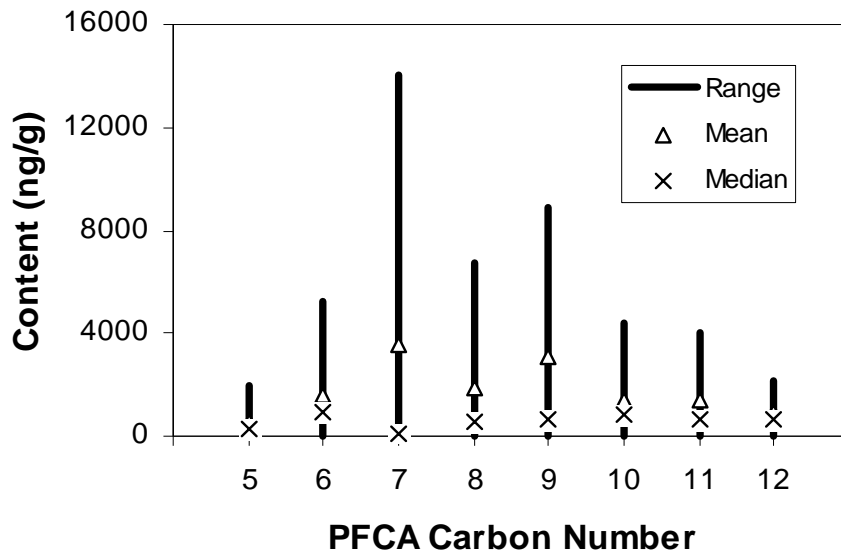


Figure 5-3. Ranges, arithmetic means, and medians for PFCAs in commercial carpet-care liquids (N = 9).

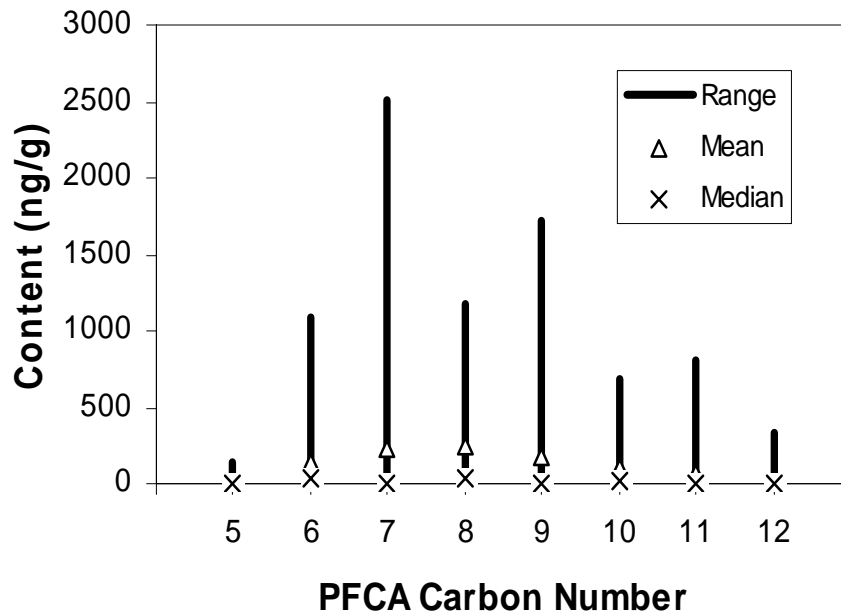


Figure 5-4. Ranges, arithmetic means, and medians for PFCAs in household carpet / fabric care liquids and foams (N = 12)

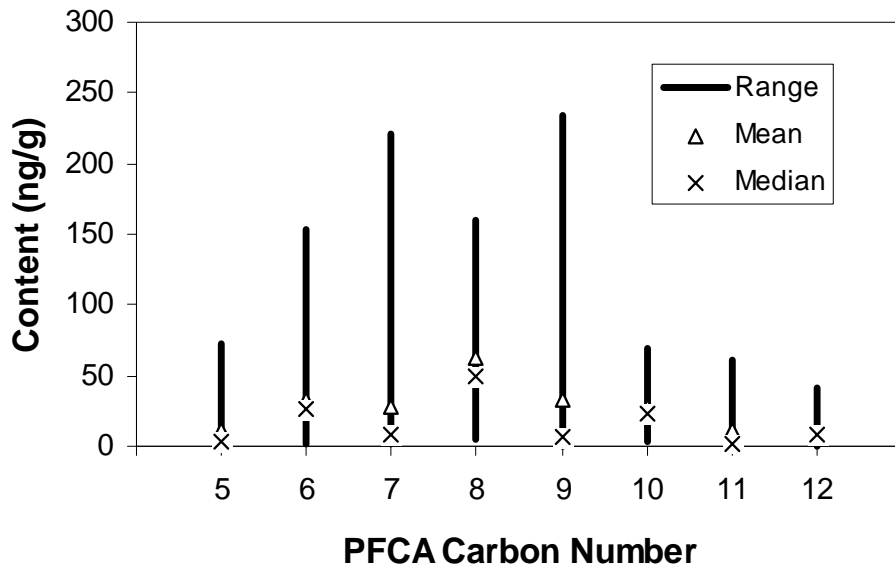


Figure 5-5. Ranges, arithmetic means, and medians for PFCAs in treated apparel (N = 14)

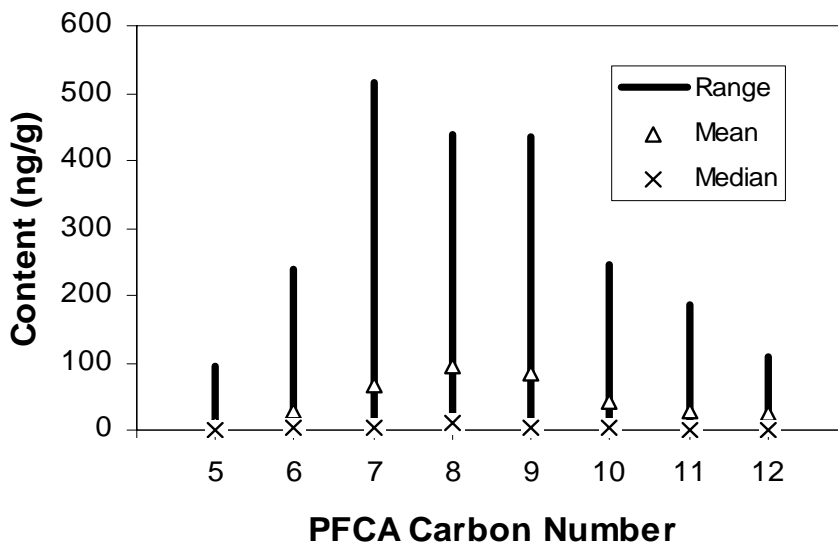


Figure 5-6. Ranges, arithmetic means, and medians for PFCAs in treated home textile products and upholstery (N = 14).

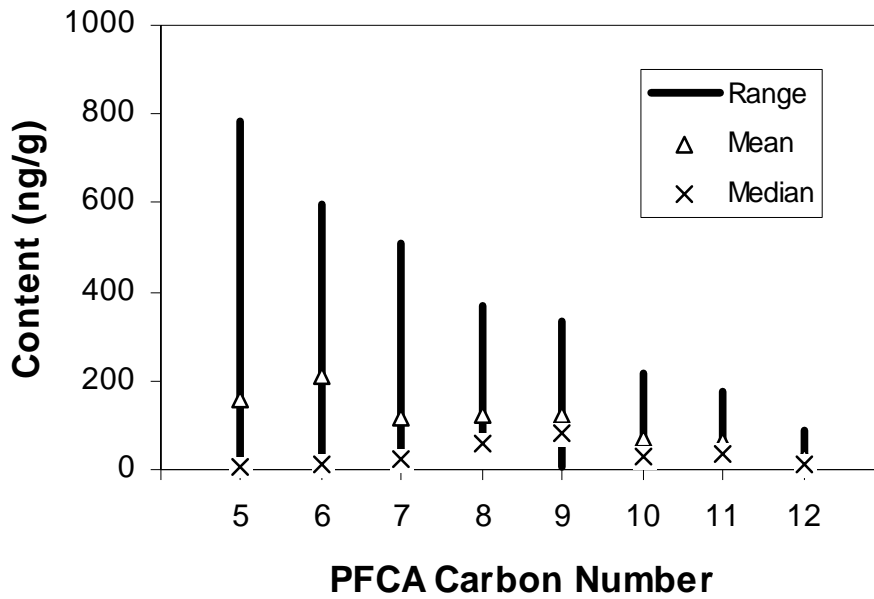


Figure 5-7. Ranges, arithmetic means, and medians for PFCAs in non-woven medical garments (N = 5)

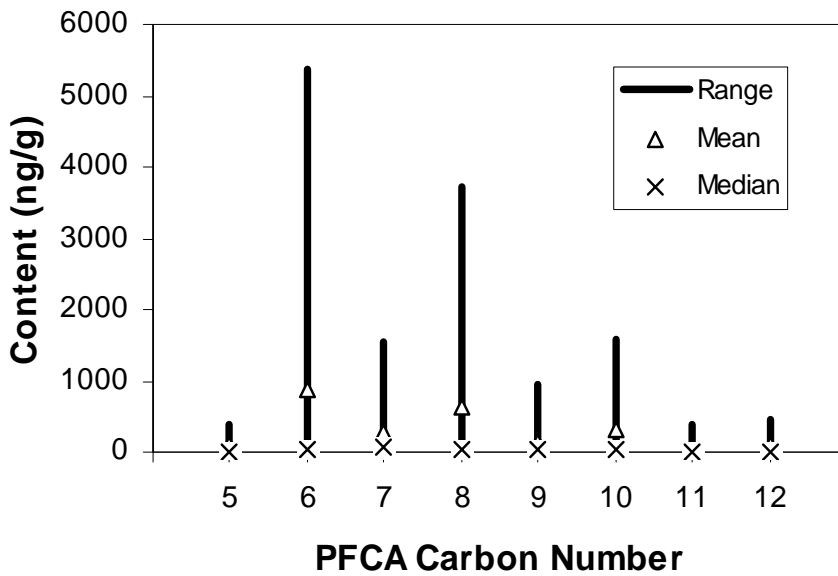


Figure 5-8. Ranges, arithmetic means, and medians for PFCAs in floor waxes and stone/tile/wood sealants (N = 10).

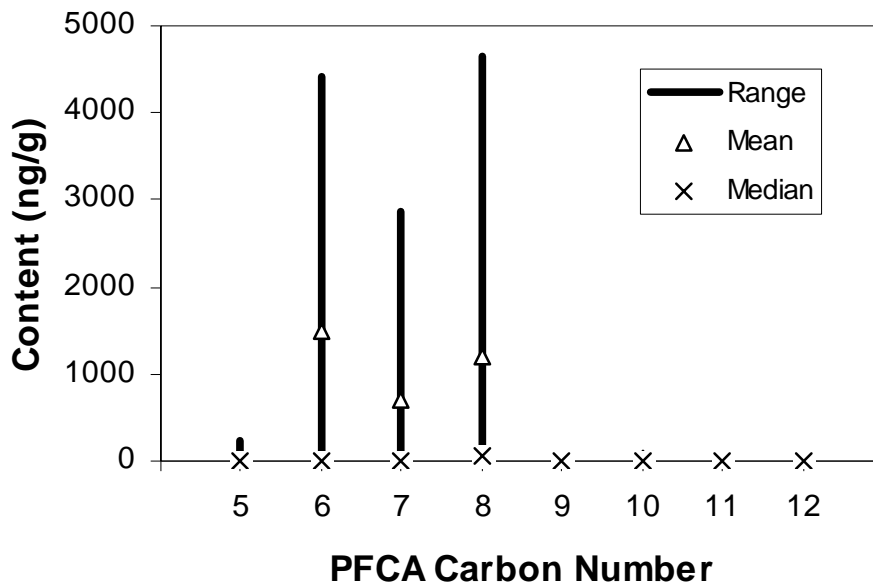


Figure 5-9. Ranges, arithmetic means, and medians for PFCAs in food contact paper (N = 4)

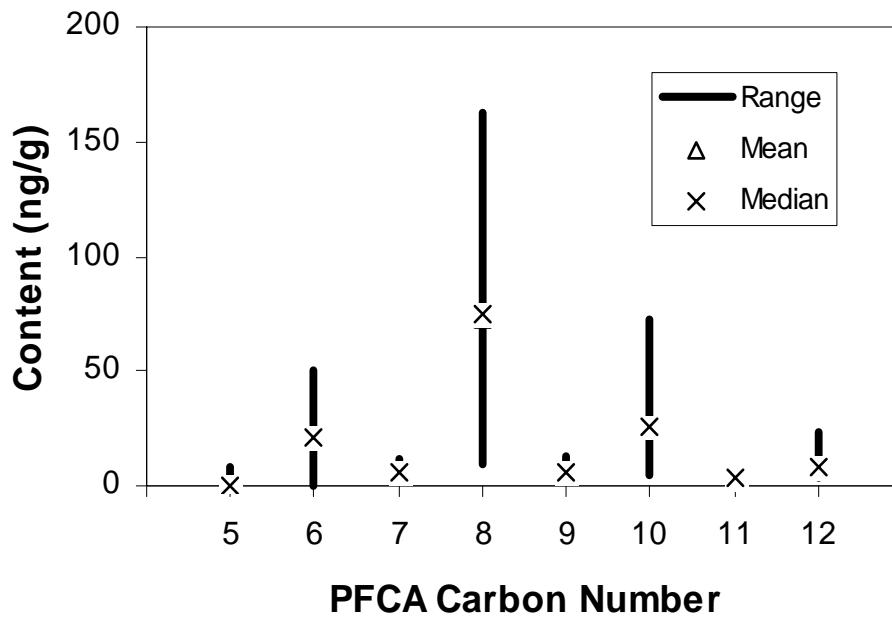


Figure 5-10. Ranges, arithmetic means, and medians for PFCAs in the membranes for apparel (N = 9). Note that the means are invisible because they are superimposed by the medians.

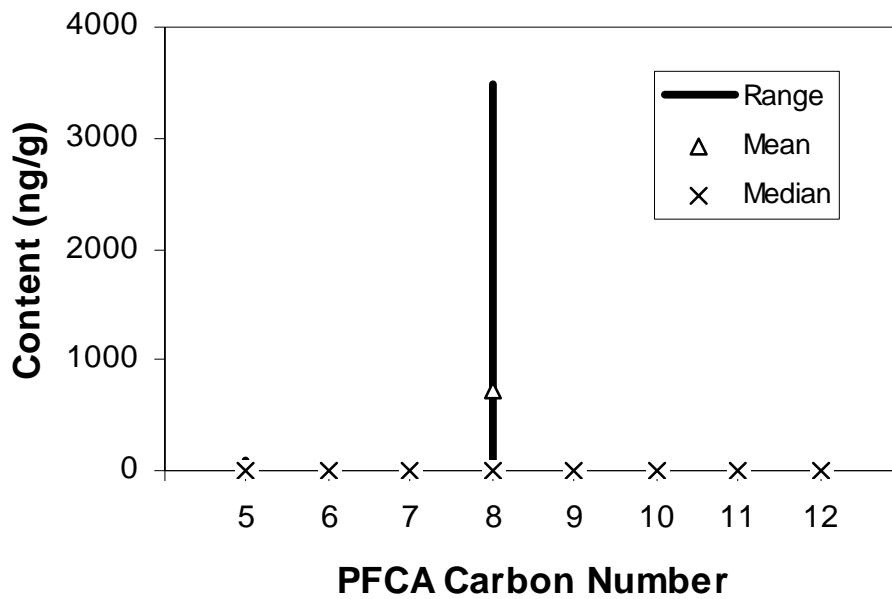


Figure 5-11. Ranges, arithmetic means, and medians for PFCAs in thread seal tape and pastes (N = 9)

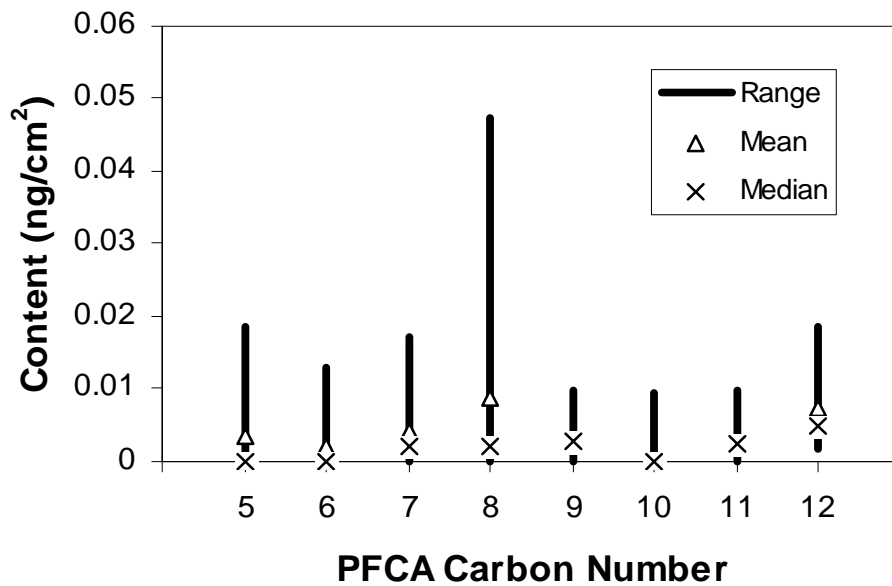


Figure 5-12. Ranges, arithmetic means, and medians for PFCAs in non-stick cookware (N = 13)

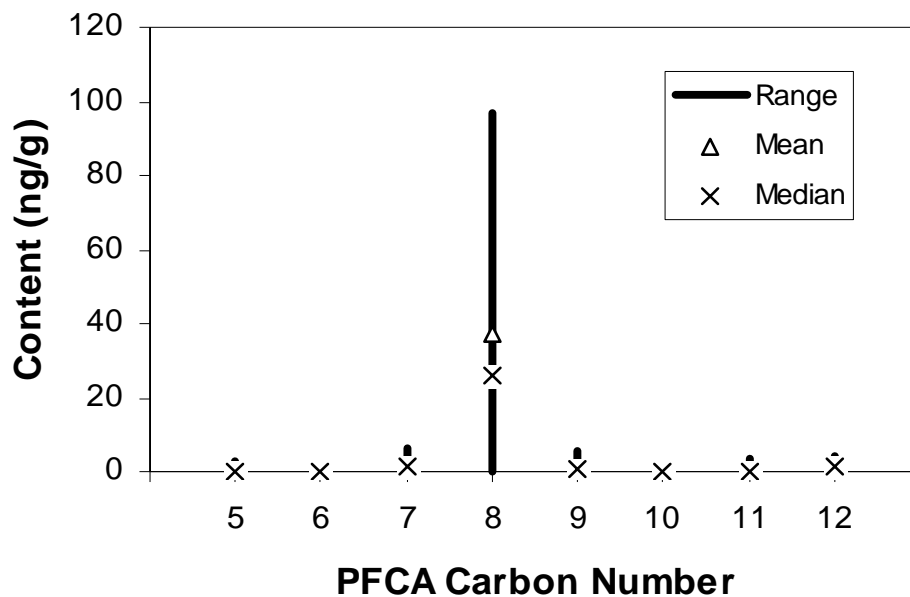


Figure 5-13. Ranges, arithmetic means, and medians for PFCAs in dental floss and plaque removers made from PTFE (N = 6)

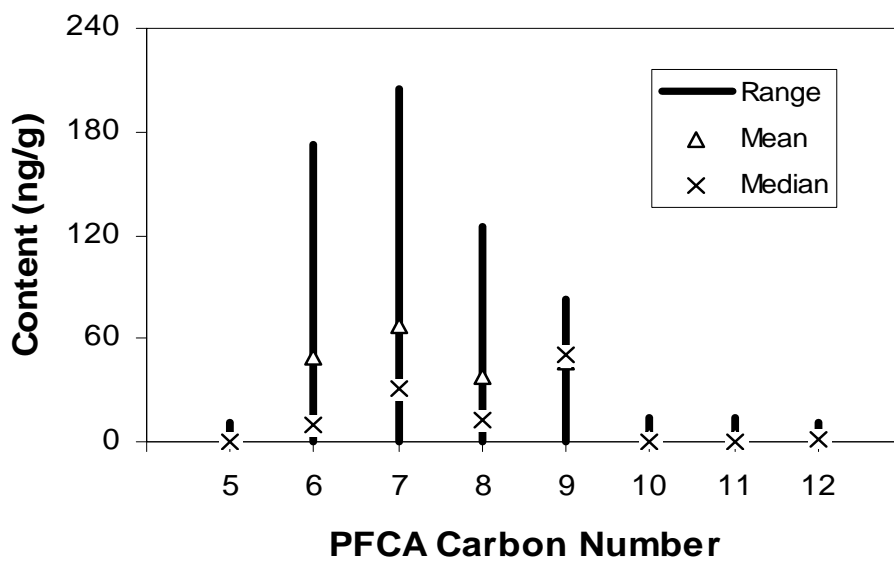


Figure 5-14. Ranges, arithmetic means, and medians for PFCAs in miscellaneous articles (N = 4)

6. Discussion

6.1 Source Strengths

A recent study by Strynar, et al. ^[13] reported that the house dust samples collected from 102 home and 10 daycare centers in the United States showed higher PFOA content and greater prevalence compared to samples collected from Japan ^[11] and Canada ^[12]. These authors also find that 96.4% of their samples have quantifiable levels of PFOA, suggesting widespread PFCA contamination in American homes. In simplified terms, the strength of a PFCA source can be expressed as the product of the PFCA content in unit weight (or area) and the quantity of the article in a given microenvironment. Table 6-1 compares the source strengths by using the arithmetic means for different AOC categories and the estimated quantities of articles in a hypothetical “typical” American home. Among the 13 article categories, professional carpet-care liquids, pre-treated carpeting, floor waxes and stone/tile/wood sealers and household textiles and upholstery are likely the largest PFCA sources in American homes.

Table 6-1. Comparison of source strengths for total amount of PFCA (TPFCA) in a hypothetical, “typical” American home ^a

| Group ID | Article category | TPFCA in article | Article quantity ^b | TPFCA in home (mg) |
|----------|--|--------------------------|-------------------------------|--------------------|
| A | Pre-treated carpeting ^c | 48.4 ng/cm ² | 150 m ² | 72.6 |
| B | Commercial carpet-care liquids | 12000 ng/g | 6 kg ^d | 71.8 |
| C | Household carpet/fabric-care liquids and foams | 953 ng/g | 1 kg | 0.95 |
| D | Treated apparel | 198 ng/g | 2 kg | 0.40 |
| E | Treated home textile and upholstery | 336 ng/g | 5 kg | 1.68 |
| F | Treated non-woven medical garments | 795 ng/g | 0 kg | 0 |
| G | Treated floor waxes and stone/tile/wood sealants | 2430 ng/g | 1 kg | 2.42 |
| H | Treated food contact paper | 3100 ng/g | 0.01 kg | 0.03 |
| I | Membranes for apparel | 124 ng/g | 1 kg | 0.12 |
| J | Thread seal tapes and pastes | 603 ng/g | 0.02 kg | 0.01 |
| K | Non-stick cookware | 0.028 ng/cm ² | 1 m ² | 0.0003 |
| L | Dental floss and plaque removers | 31.3 ng/g | 0.005 kg | 0.0002 |
| M | Miscellaneous | 69.5 ng/g | 0 | 0 |

^aThe average, single-family home size in the U.S. in 2004 was 2330 ft² (<http://www.nahb.org/>). ^b The quantities of articles are rough estimates. ^c Assuming 70% of floor area is carpet; conversion factors for total PFCA are given in supporting information. ^d For one application; dilution factor is considered.

6.2 Comparison with literature values

Comparison between the results of this study and those of other reported studies can be made only for PFOA, because data are unavailable in the literature for other PFCAs. As shown in Table 6-2, which compares the ranges of PFOA content in different article categories, the results from this study appear significantly higher than the literature values for treated non-woven medical garments, stone/tile/wood sealants, membranes for apparel, food contact paper, and dental floss/tape. The opposite appears true for treated apparel and treated home textiles. Washburn, et al. did not detect PFOA in any treated medical garments samples ^[7], but PFOA was detected at relatively high levels in all five samples in this study (Table 5-7 and Figure 5-7). Further statistical comparisons are impossible due to the lack of details about the literature values.

Table 6-2. Comparison of PFOA content ranges between this study and literature values (in ng PFOA/g sample, unless indicated otherwise)

| Article category | Literature ^a | This study ^a |
|---------------------------------------|---|-------------------------------|
| Pre-treated carpeting | 200 to 600 ^b | ND (<1.5) to 462 |
| Carpet-care liquid treated carpeting | 200 to 2000 ^b | 0.6 to 224 ^c |
| Treated apparel | ND (<20) to 1400 ^b | 5.4 to 161 |
| Treated upholstery | ND (<34) ^b | 0.6 to 293 |
| Treated home textiles | ND (<20) to 1400 ^b | 3.8 to 438 |
| Treated non-woven medical garments | ND (<34) ^b | 46 to 369 |
| Industrial floor wax and wax removers | 0.5 to 60 ^b | 7.5 to 44.8 |
| Stone, tile, and wood sealants | ND (<100) ^b | 477 to 3720 |
| Membranes for apparel | 0.008 to 0.07 ng/cm ² ^b | 0.1 to 2.5 ng/cm ² |
| Food contact paper | 6 to 290 ^d | ND (<1.5) to 4640 |
| Dental floss/tape | 3 to 4 ^d | ND (<1.5) to 96.7 |
| Thread sealant tape | 1800 ^d | ND (<1.5) to 3490 |
| PTFE cookware | 4 to 75 ^d | ND (<1.5) to 4.3 |

^a ND: not detected (detection limit in parentheses). ^b Data source: ref 7 (based on theoretical calculations). ^c Calculated by using the recommended coverage and assuming the area density of the carpet is 0.25 g/cm². ^d Data source: ref 17.

6.3 Relative Abundance of PFCAs

For fluorotelomer-treated articles (categories A to H in Table 5-1), three patterns are recognizable: (a) the distribution of the relative abundances resembles, to a certain degree, a log-normal distribution (Figure 6-1); (b) PFCAs with even numbers of carbons are predominant (Figure 6-2); and (c) PFCAs with odd numbers of carbons are predominant (Figure 6-3). The frequencies of occurrence for the three patterns are (a) > (b) > (c). Case c is rare. Depending on the article categories, the relative abundance of PFOA (C8) ranges from 15% to 38% (Figure 6-4). The large value for category H (food

contact paper) was the result of a single sample that had the highest PFCA content. The previous study on popcorn packaging paper gave the PFOA abundance of 34% in total (C5 to C12) PFCAs^[20]. For articles containing or made from fluoropolymers (categories I to L), PFOA (C8) was the most abundant PFCA species. Its relative abundance ranged from 38% to 93%.

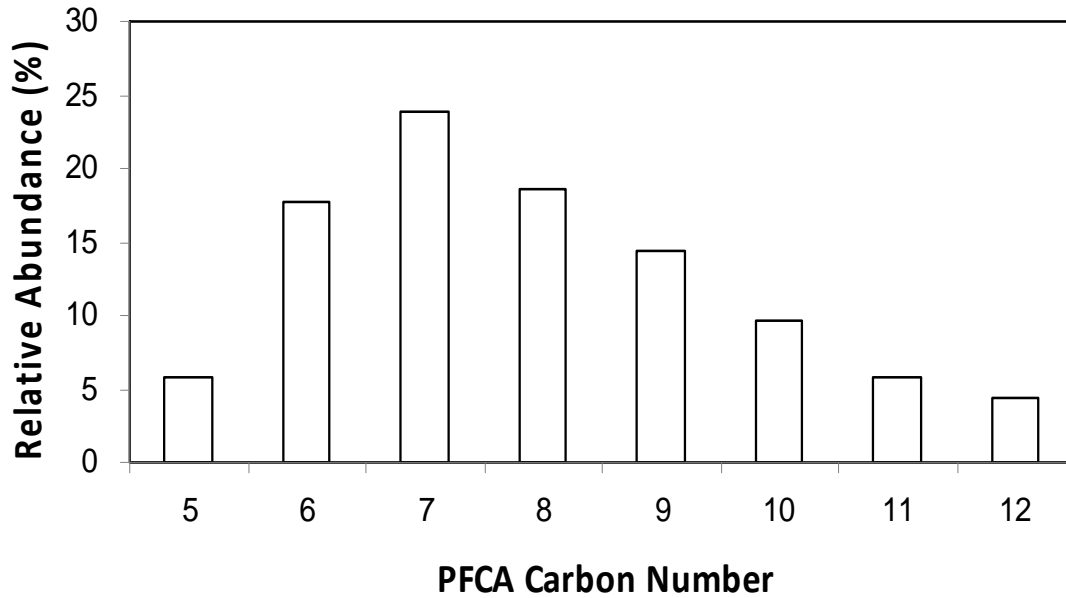


Figure 6-1. Relative abundance of PFCA in floor wax G-11.

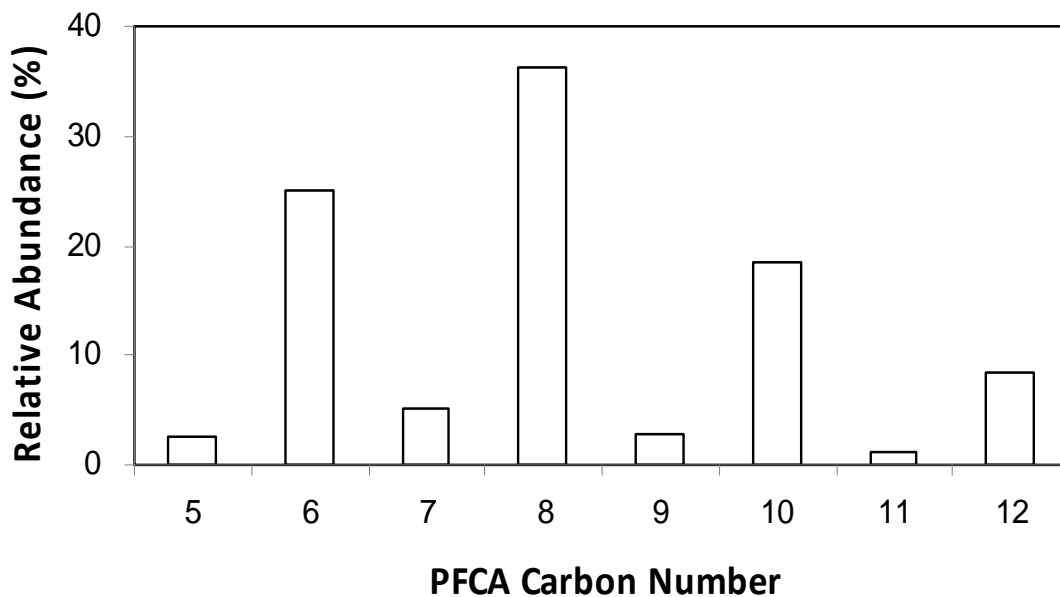


Figure 6-2. PFCA relative abundance in sample D-7 (treated apparel).

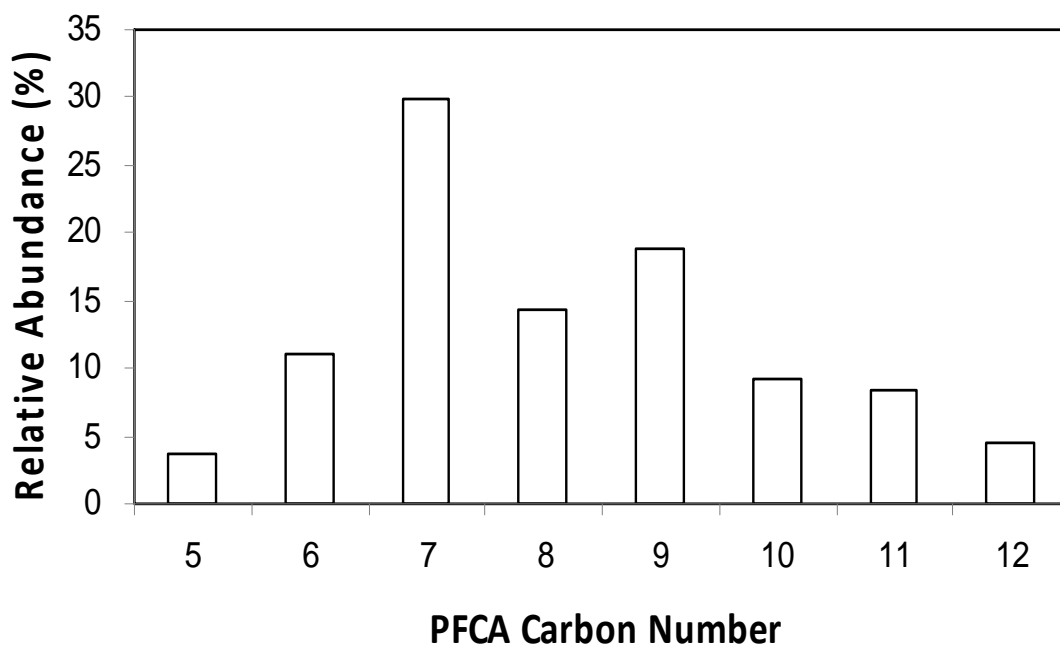


Figure 6-3. PFCA relative abundance in sample B-1 (commercial carpet-care liquid).

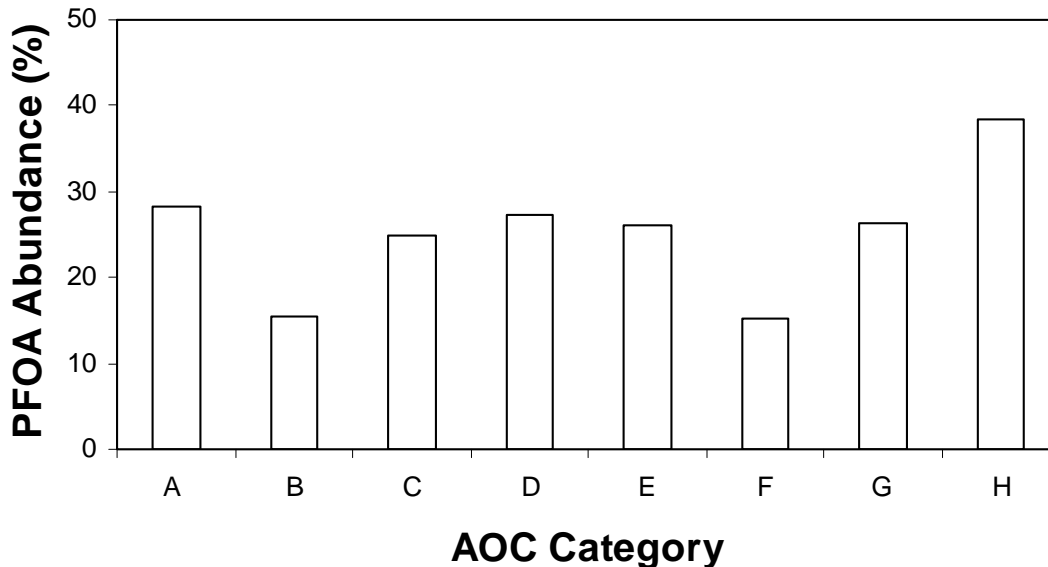


Figure 6-4. Relative abundance of PFOA (C8) in fluorotelomer-treated AOC samples (i.e., article categories A to H in Table 3-1).

6.4 Domestic versus Imported Articles

Side-by-side comparison between U.S. domestic products and imported products was difficult because they were unevenly distributed among the 13 article categories studied. For instance, pre-treated carpeting, carpet-care liquid, and floor wax categories are dominated by domestic products. On the other hand, treated apparel, treated home textile and upholstery, treated non-woven medical garments, and membranes for apparel categories are dominated by imported products. The articles with the highest PFCA content in each category were divided approximately evenly between domestic and imported products. Domestic products have the highest PFCA content in categories A, B, C, G, H, and M, while imports have the highest PFCA content in categories D, E, F, I, K, and L. The sample with the highest PFCA content treated non-woven medical garments (category F) was an imported product using materials made in the U.S. Because of the globalization of the world economy, it is obvious that international collaboration will be necessary to reduce further the PFCA content in consumer articles in the world market.

6.5 Market Trends

In recent years, government agencies, chemical companies, the research community, and environmental groups have been working to reduce the production of PFCAs and the use of PFCAs and their precursor chemicals in fluorotelomer and fluoropolymer products. For example, the U.S. Environmental Protection Agency initiated the PFOA Stewardship Program in 2006, in which the eight major companies in the industry committed voluntarily to reduce facility emissions and product content of

PFOA and related chemicals on a global basis by 95 percent no later than 2010 and to work toward eliminating emissions and product content of these chemicals by 2015 (<http://www.epa.gov/oppt/pfoa/pubs/pfoastewardship.htm>). During the process of collecting samples for this study, the application of fluorinated chemicals in AOCs had been undergoing a transition aimed at reducing the PFCA content. The availability of treated AOCs in certain categories, such as treated apparel and food contact paper, had been reduced. The limited data from this study seem to suggest that some fluorinated surface-modifying agents have been re-formulated to lower the PFCA content. The first two cases in Table 6-3 show such changes. The trends are uneven, however, and significant numbers of articles with high PFOA content are still on the market. In one case, the PFOA content even increased significantly (the third case in Table 6-3). The data in Table 3-3 are very limited in scope and the two sampling times are only one year apart. It takes a much longer period of time (e.g., several years) and a much wider range of sampling to confirm any trends.

Table 6-3. Changes in PFOA content for three AOC samples

| Product | PFOA content (ng/g) ^a | | p-Value ^b |
|-----------------------------|----------------------------------|--------------|----------------------|
| | Apr-May 2007 | May 2008 | |
| Pre-treated carpet | 462 ± 51.6 | 5.88 ± 0.444 | <0.001 |
| Commercial carpet protector | 1838 ± 92.7 | 24.5 ± 1.21 | <0.0001 |
| Commercial carpet protector | 599 ± 65.1 | 1722 ± 89.2 | <0.001 |

^a Mean and standard deviation for duplicate extractions; ^b For one-sided student test.

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Appendix Description of AOC Samples

Table A-1. Sample description and conversion factor ^a

| Sample ID | Description | Conversion Factor | |
|--|---|----------------------------|-----------------------|
| | | Units | Value |
| <i>A. Pre-treated carpeting</i> | | | |
| A-1 | Nylon Carpet 1 | g of fiber/cm ² | 1.41×10 ⁻¹ |
| A-2 | Renewably sourced polymer | g of fiber/cm ² | 8.57×10 ⁻² |
| A-3 | Polyester carpet | g of fiber/cm ² | 9.33×10 ⁻² |
| A-4 | Berber 94% Olefin/6%Nylon carpet | g of fiber/cm ² | 1.13×10 ⁻¹ |
| A-5 | Nylon Carpet 2 | g of fiber/cm ² | 1.31×10 ⁻¹ |
| A-6 | Frieze nylon carpet | g of fiber/cm ² | 7.15×10 ⁻² |
| A-7 | Nylon Carpet 3 | g of fiber/cm ² | 6.61×10 ⁻² |
| A-8 | Textured carpet | g of fiber/cm ² | 1.45×10 ⁻¹ |
| A-9 | Nylon Carpet 4 | g of fiber/cm ² | 7.44×10 ⁻² |
| <i>B. Commercial carpet/fabric care liquids (not applicable)</i> | | | |
| B-1 | Carpet and upholstery protector | N/A | N/A |
| B-2 | Carpet protector concentrate 1 | N/A | N/A |
| B-3 | Fabric protector solvent based | N/A | N/A |
| B-4 | Carpet protector concentrate 2 | N/A | N/A |
| B-5 | Carpet/upholstery protector concentrate | N/A | N/A |
| B-6 | Carpet protector concentrate 3 | N/A | N/A |
| B-7 | RTU Carpet protector | N/A | N/A |
| B-8 | Carpet protector | N/A | N/A |
| B-9 | RTU Carpet protector | N/A | N/A |
| <i>C. Carpet/fabric care liquids for spot treatment</i> | | | |
| C-1 | Household carpet shampoo | N/A | N/A |
| C-2 | Carpet spot and stain remover | N/A | N/A |
| C-3 | Spot removal kit | N/A | N/A |
| C-4 | Fabric protector 1 | N/A | N/A |
| C-5 | Fabric protector 2 | N/A | N/A |
| C-6 | Household carpet protector | N/A | N/A |
| C-7 | Household carpet/upholstery cleaner | N/A | N/A |
| C-8 | Household spot & stain remover | N/A | N/A |
| C-9 | Wash-in waterproofing | N/A | N/A |
| C-10 | Spray-on water repellent | N/A | N/A |
| C-11 | Wash-in waterproofing | N/A | N/A |
| C-12 | Spray-on water repellent | N/A | N/A |

^a N/A = not applicable.

Table A-1. Sample description and conversion factor (cont.)

| Sample ID | Description | Conversion Factor | |
|---|--|-------------------|-----------------------|
| | | Units | Value |
| <i>D. Treated apparel</i> | | | |
| D-1 | Man's pant | g/cm ² | 2.06×10 ⁻² |
| D-2 | Man's short pant | g/cm ² | 2.50×10 ⁻² |
| D-3 | Girl's uniform shirt | g/cm ² | 2.28×10 ⁻² |
| D-4 | Boy's dress shirt 1 | g/cm ² | 1.60×10 ⁻² |
| D-5 | Boy's dress pant 2 | g/cm ² | 2.61×10 ⁻² |
| D-6 | Lady's dress pant | g/cm ² | 2.35×10 ⁻² |
| D-7 | Girl's uniform pant | g/cm ² | 2.30×10 ⁻² |
| D-8 | Woman's hiking shoe | g/cm ² | 2.35×10 ⁻² |
| D-9 | Woman's knee pant | g/cm ² | 1.23×10 ⁻² |
| D-10 | Man's dress pant | g/cm ² | 2.93×10 ⁻² |
| D-11 | Man's polo shirt | g/cm ² | 2.18×10 ⁻² |
| D-12 | Man's nylon shirt | g/cm ² | 1.10×10 ⁻² |
| D-13 | Man's dress shirt 1 | g/cm ² | 1.14×10 ⁻² |
| D-14 | Man's dress pant 2 | g/cm ² | 2.78×10 ⁻² |
| D-15 | Man's dress pant 3 | g/cm ² | 2.65×10 ⁻² |
| D-16 | Lab coat | g/cm ² | 1.68×10 ⁻² |
| <i>E. Treated home textile and upholstery</i> | | | |
| E-1 | Microfiber fabric 1 | g/cm ² | 2.43×10 ⁻² |
| E-2 | Microfiber fabric 2 | g/cm ² | 2.45×10 ⁻² |
| E-3 | Loveseat slip cover 100% cotton | g/cm ² | 1.80×10 ⁻² |
| E-4 | Loveseat slip cover 100% polyester | g/cm ² | 2.49×10 ⁻² |
| E-5 | Loveseat slip cover 60% cotton/40% polyester | g/cm ² | 2.00×10 ⁻² |
| E-6 | Loveseat slip cover 100% polyester | g/cm ² | 1.40×10 ⁻² |
| E-7 | Mattress pad 1 | g/cm ² | 1.35×10 ⁻² |
| E-8 | Mattress pad 2 | g/cm ² | 1.95×10 ⁻² |
| E-9 | Mattress pad 3 | g/cm ² | 1.38×10 ⁻² |
| E-10 | Table cloth | g/cm ² | 1.60×10 ⁻² |
| E-11 | Ironing board cover | g/cm ² | 1.18×10 ⁻² |
| E-12 | Table cloth | g/cm ² | 2.19×10 ⁻² |
| E-13 | Upholstery fabric, 57% cotton/43% polyester | g/cm ² | 4.75×10 ⁻² |
| E-14 | 100% Cotton throw | g/cm ² | 2.20×10 ⁻² |
| <i>F. Treated non-woven medical garments</i> | | | |
| F-1 | Lab coat | g/cm ² | 6.00×10 ⁻³ |
| F-2 | Surgical gown 1 | g/cm ² | 6.00×10 ⁻³ |
| F-3 | Surgical gown 2 | g/cm ² | 5.00×10 ⁻³ |
| F-4 | Surgical gown 3 | g/cm ² | 7.50×10 ⁻³ |
| F-5 | Reuseable pillow | g/cm ² | 1.10×10 ⁻² |

Table A-1. Sample description and conversion factor (cont.)

| Sample ID | Description | Conversion Factor | |
|--|----------------------------|-------------------|-----------------------|
| | | Units | Value |
| <i>G. Treated floor waxes and stone/tile/wood sealants</i> | | | |
| G-1 | Household floor wax 1 | N/A | N/A |
| G-2 | Household floor wax 2 | N/A | N/A |
| G-3 | Commercial spray and buff | N/A | N/A |
| G-4 | Commercial floor sealer 1 | N/A | N/A |
| G-5 | Commercial floor sealer 2 | N/A | N/A |
| G-6 | Commercial floor sealer 3 | N/A | N/A |
| G-7 | Stone & tile sealer | N/A | N/A |
| G-8 | Granite sealer | N/A | N/A |
| G-9 | Household floor polish | N/A | N/A |
| G-10 | Marble & granite sealer | N/A | N/A |
| G-11 | Stone, tile & wood sealant | N/A | N/A |
| <i>H. Treated food contact paper</i> | | | |
| H-1 | French fry bag | g/cm ² | 3.25×10 ⁻³ |
| H-2 | French fry carton | g/cm ² | 2.41×10 ⁻² |
| H-3 | Popcorn bucket | g/cm ² | 1.99×10 ⁻² |
| H-4 | Popcorn bag | g/cm ² | 3.87×10 ⁻³ |
| H-5 | Microwave cooking bag | g/cm ² | 8.60×10 ⁻³ |
| <i>I. Membranes for apparel</i> | | | |
| I-1 | Women's rain jacket | g/cm ² | 9.83×10 ⁻³ |
| I-2 | Men's rain jacket | g/cm ² | 9.39×10 ⁻³ |
| I-3 | Men's jacket | g/cm ² | 8.64×10 ⁻³ |
| I-4 | Women's insulated pant | g/cm ² | 1.59×10 ⁻² |
| I-5 | Men's parka | g/cm ² | 1.82×10 ⁻² |
| I-6 | Woman's rain jacket | g/cm ² | 1.90×10 ⁻² |
| I-7 | Sombrero (hat) 1 | g/cm ² | 1.30×10 ⁻² |
| I-8 | Sombrero (hat) 2 | g/cm ² | 1.40×10 ⁻² |
| I-9 | Man's rain parka | g/cm ² | 9.00×10 ⁻³ |
| I-10 | Ball cap | g/cm ² | 1.83×10 ⁻² |
| <i>J. Thread seal tapes and pastes</i> | | | |
| J-1 | 1/2" Tape 1 | g/cm ² | 2.97×10 ⁻³ |
| J-2 | 1/2" Tape 2 | g/cm ² | 2.86×10 ⁻³ |
| J-3 | 1/2" Tape 3 | g/cm ² | 2.66×10 ⁻³ |
| J-4 | 1/2" Tape 4 | g/cm ² | 2.71×10 ⁻³ |
| J-5 | 1/2" Tape 5 | g/cm ² | 2.89×10 ⁻³ |
| J-6 | 1/2" Tape 6 | g/cm ² | 1.01×10 ⁻³ |
| J-7 | 1/2" Tape 7 | g/cm ² | 9.49×10 ⁻³ |
| J-8 | Pipe thread sealant 1 | N/A | N/A |
| J-9 | Pipe thread sealant 2 | N/A | N/A |

Table A-1. Sample description and conversion factor (cont.)

| Sample ID | Description | Conversion Factor | |
|--|-------------------------|-------------------|----------|
| | | Units | Value |
| <i>J. Thread seal tapes and pastes (cont.)</i> | | | |
| J-10 | Pipe thread sealant 3 | N/A | N/A |
| <i>K. Non-stick cookware</i> | | | |
| K-1 | 10" Fry pan | N/A | N/A |
| K-2 | 10" Fry pan w/lid | N/A | N/A |
| K-3 | 5 qt Sautesse w/lid | N/A | N/A |
| K-4 | 12" Fry pan w/lid | N/A | N/A |
| K-5 | 14" Utility pan w/lid | N/A | N/A |
| K-6 | 10" Camp fry pan | N/A | N/A |
| K-7 | Child's mess kit | N/A | N/A |
| K-8 | Cookie sheet | N/A | N/A |
| K-9 | 11" Skillet | N/A | N/A |
| K-10 | 2 qt Sauce pan | N/A | N/A |
| K-11 | 22-cm Fry pan | N/A | N/A |
| K-12 | 10" Skillet | N/A | N/A |
| K-13 | 10" Skillet | N/A | N/A |
| K-14 | 4 qt 10.5" Sauteuse pan | N/A | N/A |
| <i>L. Dental floss and plaque removers</i> | | | |
| L-1 | Unflavored floss | g/m | 1.75E-01 |
| L-2 | Mint floss | g/m | 1.30E-01 |
| L-3 | Satin floss | g/m | 9.91E-02 |
| L-4 | Satin tape | g/m | 1.21E-01 |
| L-5 | Flosser | g/m | 4.67E-02 |
| L-6 | Deep clean floss | g/m | 1.42E-01 |
| L-7 | Micro-mint flosser | g/m | 4.67E-02 |
| L-8 | Angle flosser | g/m | 1.53E-01 |
| <i>M. Miscellaneous</i> | | | |
| M-1 | Tire shine | N/A | N/A |
| M-2 | Car spray wax | N/A | N/A |
| M-3 | Car wheel cleaner | N/A | N/A |
| M-4 | Dry sack | g/cm ² | 1.00E-02 |
| M-5 | Deck cleaner | N/A | N/A |
| M-6 | Boat polish 1 | N/A | N/A |
| M-7 | Boat polish 2 | N/A | N/A |