Supporting Information

How Do We Measure Poly- and Perfluoroalkyl Substances (PFASs) at the Surface of Consumer Products?

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Methods

Materials. A Thermo Scientific™ Barnstead™ GenPure™ xCAD Plus (Lake Balboa, CA, U.S.A.) UV-TOC provided ultrapure deionized water (DI) with a resistivity of >18 MΩ · cm. LC-MS grade methanol (J.T. Baker, Center Valley, PA, U.S.A.), reagent grade formic acid and BioUltra ammonium acetate were obtained from Sigma Aldrich (St. Louis, MO, U.S.A.).

LC-MS/MS Extraction. The methanol extraction was adapted from previous studies.[1, 2] For this study, 1 ± 0.03 g subsamples (when possible) were cut with methanol-cleaned scissors and added to 15 mL polypropylene centrifuge tubes. Then, 12 mL of methanol was added to each centrifuge tube. Each sample was placed on a rotating shaker table for 1 hour and bath sonicated at 40 ^{0}C for 2 hours. Samples were then centrifuged at 4,000 rcf for 10 minutes and the supernatant was decanted into a new 15 mL polypropylene centrifuge tube and kept at 4 ^{0}C until analysis. One blank centrifuge tube was extracted within every set of 12 samples. We also assessed the sample area:weight ratio to convert concentrations into units of nmol m^{-2} (Table S4).

LC-MS/MS Analysis. Before LC-MS/MS analysis, the methanol extracts were warmed to room temperature and inverted before subsampling. An aliquot of methanol extract (705 µL) was combined with DI Water (750 µL) and 10 ng mL^{-1} internal standard solution (45 µL) in a polypropylene microcentrifuge tube. Samples were then briefly vortexed, centrifuged at 13,000 rpm, and then the supernatant was transferred to a polypropylene autosampler vial for analysis with an Agilent (Santa Clara, CA) 6460 triple quadrupole LC-MS/MS with electrospray ionization in negative ion mode. LC-MS/MS analysis followed the procedure outlined in Weber et al.[3] The injection volume was 300 µL. All calibration samples were prepared in a 50:50 methanol:water solution to match the sample composition. In this study, the linear calibration ranged from ~2 ng L^{-1} - 1200 ng L^{-1}, contained up to 8 points, and was forced through zero and 1/x weighted. All compounds were required to have $r^2 > 0.99$ for the calibration. N-EtFOSAA and N-MeFOSAA were added to the compounds measured by Weber et al.[3] (see Table S3 for parameters). If samples exceeded the calibration range by over 15% they were diluted and re-analyzed.

The limit of quantification was defined as the average sample concentration at which the signal-to-noise ratio was 10, plus the concentration of any LC-MS/MS blank (50:50 DI water:methanol with internal standards) above this value. The method quantification limit (MQL) was corrected for dilution. All blanks included in the extraction procedure were below the MQL.

Quality Assurance/Quality Control. Duplicate injections of ~20% of the samples were completed and the results were averaged (average relative percent difference <25% except for N-EtFOSAA, which was 74% for a single sample). Recovery and precision experiments were performed on 3 samples (1 food contact material: plate, 1 textile: carpet, 1 domestic product: curtain) that had initial PFAS concentrations below the MQL to evaluate the effectiveness of the extraction method. Briefly, four 1 ± 0.03 g samples were cut from the chosen samples and added to individual 15 mL polypropylene centrifuge tubes. All 12 sample was spiked with 7.2 ng of a native stock solution in methanol. The stock solution was added directly onto the sample and left to equilibrate for 30 minutes at room temperature. The extraction and analysis followed the methods outlined in the LC-MS/MS Extraction section. See Table S7 for results. Percent recoveries were within ±30% of expected values except for PFBS (66.3% recovery, plate), FOSA (69% recovery, plate and 69.7% recovery, carpet) and 8:2 FtS, which was removed from further consideration due to recoveries of only 22.6% for the carpet sample. Recoveries of 8:2 FtS may have been low due to matrix effects. PFDS was removed from further consideration due to poor reproducibility of continuing calibration checks.

XPS Analysis. The peak deconvolution and atomic ratio were calculated using the Thermo Scientific Avantage software. The accuracy of the C1s peak fitting is ±2%. The pass energy was 50 eV for the high-resolution scans and 200 eV for the survey. At least 2 scans were performed for each data point for both the survey and the high-resolution scan. For food contact materials the food contact side was measured, whereas for textiles, the outer surface was measured (outer lining of jackets, the surface that humans would come into contact with for upholstery, etc.). For domestic products, the surface that was presumed to come into contact with humans most frequently was measured. Depth profiles were completed by etching with a monatomic argon ion beam (ion gun energy = 250 eV) with 10 seconds intervals. The material etching rate was 0.15 nm sec^{-1}, based on a tantalum pentoxide reference material. Therefore, 1.5 nm of material was etched between each XPS data point. However, because consumer products likely are not etched at the same rate as tantalum pentoxide, we estimated an etching rate of 15 nm sec^{-1} (see below section on Sputtering Yield for Consumer Products).

Methanol Extraction for XPS. To reduce variability, a small section (<1 cm^2) of material was removed for pre-extraction XPS, and then a second subsample neighboring the first subsample was removed for post-extraction XPS. Samples were extracted following the LC-MS/MS extraction procedure outlined above, and then air-dried before analysis with XPS. Four samples with no fluorine pre-extraction were also included as controls. One of these controls (sample 9) had detectable fluorine post-extraction. The extraction was repeated, but sample 9 still displayed a small fluorine peak (<1.5% F) in the post-extraction sample. In both experiments, sample 9 had no fluorine detected via XPS pre-extraction. The other samples included as blank controls had no fluorine pre- or post-extraction. Sample 9 has a polylactic acid lining (Table S1). Methanol extraction may have removed this lining, exposing an underlying fluorine-containing surface. This is supported by the LC-MS/MS results, which show PFHxA and PFOA were present in sample 9.

LC-QTOF-MS Analysis. A Shimadzu high performance reverse-phase liquid chromatography (HPLC) system coupled with a Sciex 5600 Triple Quadrupole Time-of-Flight (QTOF) MS was used to analyze methanol extracts from the 11 samples that had detectable fluorine with XPS. Samples were extracted as outlined in the LC-MS/MS Extraction section, but with ACS-grade methanol. This analysis was completed to determine whether there were structures not captured with LC-MS/MS analysis prevalent in the samples. The HPLC was fitted with a Kinetex EVO C18 (2x.1 x 100 mm, 5 µm, 100 Å) column kept at 40 ºC and equipped with a Phenomenex AF0-8497 filter. The injection volume was 50 µL for each sample and mobile phases were water with 0.15% acetic acid (A) and 20 mM ammonium acetate in methanol (B). Samples were scanned in both positive and negative ESI scan modes, and data acquisition and processing were performed using Analyst TF 1.6 and PeakView 2.2 software, respectively.

Calculations

Sputtering Yield for Consumer Products. We determined the depth of etching achieved in each step based on the sputtering yield (atoms/argon ion) of the material.[4-6] The XPS etching rate is based on a tantalum pentoxide reference material (0.15 nm s^{-1}, with 10 seconds of etching between each data point), which is likely not similar to the consumer products measured here due to differences between organic and inorganic materials.[5] A formula derived in previous work[5] was applied here, and the etching rate of the consumer products was estimated to be less than 2 orders of magnitude larger than the etching rate for tantalum pentoxide, assuming the samples are similar to organics like polystyrene or polycarbonate. We consider 2 orders of magnitude an upper bound, since fluoropolymers like PTFE and PVDF were found to have sputtering yields less than polystyrene or polycarbonate,[7] and these samples are known to contain carbon-fluorine bonds. This results in a maximum estimated etching depth of ~10 µm for the depth profiles shown here (Figures S1) and in the manuscript.

XPS and LC-MS/MS Weight Percent. XPS results in atomic percent fluorine were converted to weight percent fluorine as follows:

$$wt\%\ F = \frac{at\%\ F * A_r(F)}{at\%\ F * A_r(F) + \sum_{i=element} at\ \%\ i * A_r(i)} * 100$$

Where A_r(element) is the atomic weight of the element and i cycles through all elements detected in the sample in addition to fluorine.

Assuming all fluorine originated from the top 10 nm of materials, LC-MS/MS results were converted to weight percent. The following equation uses PFOA as an example:

$$wt.\%\ F = \frac{nmol_{PFOA\ extracted}}{g_{mat}} * \frac{mol}{10^9\ nmol} * \frac{15\ mol_F}{1\ mol_{\ PFOA}} * \frac{19\ g_F}{1\ mol_F} * \frac{Sample\ Thickness\ (nm)}{10nm}$$

Where g_{mat} is the total weight of the material extracted.

Tables

Table S1. Sample inventory. FCM = Food contact material.

Sample #	Weight of Material Extracted (g)	Weight/Area Ratio (g m^{-2})	Category	Sub-Category	New/ Used	Country of Origin	Compostable	Material
1	1.00	339.0	FCM	Bowl	New		yes	Molded Fiber
2	1.01	234.0	FCM	Plate	New		yes	Molded Fiber
3	1.01	338.7	FCM	Plate	New		yes	Plant Fiber
4	1.00	458.9	FCM	Takeout Box	New		yes	
5	0.99	391.4	FCM	Takeout Box	New		yes	Plant Fiber
6	1.00	359.0	FCM	Takeout Box	New		yes	Plant Fiber
7	1.00	365.3	FCM	Takeout Box	New		yes	Plant-Based
8	1.01	402.8	FCM	Takeout Box	New		yes	
9	1.00	414.1	FCM	Takeout Box	New		yes	Plant-Based, Polylactic Acid Lining
10	1.01	396.3	FCM	Takeout Box	New		yes	
11	1.00	373.0	FCM	Takeout Box	New		Unknown	
12	1.00	415.6	FCM	Bowl	New		Unknown	
13	1.01	90.97	FCM	Popcorn Bag	New	USA	Unknown	
14	1.02	29.71	FCM	Food Bag	New		Unknown	
15	0.99	1545	FCM	Utensil	New		yes	
16	0.99	402.5	FCM	Cup	New		yes	Plant-Based Lining
17	1.01	398.8	FCM	Takeout Box	New		yes	Plant Fiber, Post Consumer Material
18	1.01	306.3	FCM	Cup	New		yes	Paper, Plant-Based Lining
19	1.01	895.6	FCM	Plate	New		yes	Palm Leaves
20	1.01	605.5	FCM	Tray	New		yes	Palm Leaves
21	1.00	346.6	FCM	Takeout Box	New		yes	Polylactic Acid
22	1.00	336.1	FCM	Bowl	New		yes	Paper, Plant-Based Lining
23	0.99	347.6	FCM	Bowl	New		yes	Plant-Based
24	0.99	352.0	FCM	Cup	New	USA	yes	Polylactic Acid
25	1.00	287.5	FCM	Cup	New	USA	yes	Polylactic Acid
26	0.98	354.4	FCM	Lid	New	USA	yes	Polylactic Acid
27	1.01	302.4	FCM	Cup	New		yes	Paper, Plant-Based Lining
28	1.01	377.7	FCM	Lid	New		yes	Polylactic Acid
29	1.00	304.9	FCM	Takeout Box	New		yes	Polylactic Acid
30	0.99	296.3	FCM	Takeout Box	New		yes	
31	1.00	333.2	FCM	Takeout Box	New		Unknown	

Sample #	Weight of Material Extracted (g)	Weight/Area Ratio (g m^{-2})	Category	Sub-Category	New/ Used	Country of Origin	Compostable	Material
32	1.01	372.6	FCM	Cup	New		Unknown	
33	0.99	249.2	FCM	Cup	New		Unknown	
34	1.01	38.92	FCM	Food Bag	New		Unknown	
35	1.01	340.6	FCM	Bowl	New		Unknown	
36	0.99	276.1	FCM	Cup	New		Unknown	
37	1.02	266.5	FCM	Cup	New		Unknown	Polypropylene
38	0.99	344.8	FCM	Lid	New		Unknown	Polyethylene Terephthalate
39	1.01	355.1	FCM	Lid	New		Unknown	Polyethylene Terephthalate
40	0.99	386.1	FCM	Cup	New		Unknown	Polystyrene
41	0.99	49.78	FCM	Food Bag	New		Unknown	
42	1.01	755.1	FCM	Food Stick	New		Unknown	
43	1.00	268.2	FCM	Plate	New		Unknown	Paper
44	1.02	218.2	FCM	Cup	New	USA	Unknown	
45	1.01	39.06	FCM	Food Paper	New	USA	Unknown	
46	1.02	3017	Textile	Carpet	New			Nylon Fiber
47	1.01	394.7	Textile	Upholstery	New			Polyester, Dyed Nylon, Acrylic
48	1.02	2721	Textile	Carpet	New			Nylon Fiber
49	1.01	2303	Textile	Carpet	New			Polyester Fiber
50	1.00	2655	Textile	Carpet	New			Nylon Fiber
51	1.00	383.5	Textile	Upholstery	New	USA		Nylon, Polyester, Acrylic
52	0.99	2348	Textile	Carpet	New			Nylon Fiber
53	0.98	2513	Textile	Carpet	New			Polyester, Polytrimethylene Terephthalate Fiber
54	1.00	522.7	Textile	Upholstery	New	USA		Cotton, Nylon, Polyester, Acrylic
55	0.98	2375	Textile	Carpet	New			Polytrimethylene Terephthalate Fiber
56	1.01	2500	Textile	Carpet	New			Nylon Fiber
57	0.99	226.6	Textile	Backpack	New	China		Nylon, Polyester
58	0.99	259.6	Textile	Backpack	New	Vietnam		Nylon, Polyester
59	1.02	154.0	Textile	Mattress Pad	New			
60	1.01	3500	Textile	Carpet	Used			
61	1.00	245.0	Textile	Upholstery	Used	Taiwan		
62	0.57	555.6	Textile	Upholstery	Used			Leather
63	1.02	280.7	Textile	Jacket	Used	China		Polyurethane Shell
64	0.60	844.2	Textile	Upholstery	Used	USA		Leather
65	0.87	416.6	Textile	Upholstery	Used	USA		

Sample #	Weight of Material Extracted (g)	Weight/Area Ratio (g m^{-2})	Category	Sub-Category	New/ Used	Country of Origin	Compostable	Material
66	1.02	329.1	Textile	Jacket	Used	Pakistan		Cotton, Polyester
67	1.01	65.16	Textile	Jacket	Used	Taiwan		Nylon
68	0.99	81.00	Textile	Jacket	Used			
69	0.99	3802	Textile	Carpet	New			Nylon Fibers
70	0.98	3677	Textile	Carpet	New			
71	1.00	27.48	Textile	Jacket	New	China		Ethylene-Vinyl Acetate
72	0.99	2336	Textile	Carpet	New			Nylon Fiber
73	1.00	2194	Textile	Carpet	New			Nylon Fiber
74	1.01	2692	Textile	Carpet	New			Nylon Fiber
75	1.01	2314	Textile	Carpet	New			Wool, Olefin, Polypropylene
76	0.98	2367	Textile	Carpet	New			Wool, Olefin, Polypropylene
77	1.01	2222	Textile	Carpet	New			Polyester, Polypropylene
78	1.00	2200	Textile	Carpet	New			Polyester, Polypropylene
79	1.02	2967	Textile	Carpet	New			Polytrimethylene Terephthalate, Polypropylene
80	1.01	476.0	Textile	Backpack	New	China		Cotton
81	1.00	476.7	Textile	Backpack	New	China		Nylon, Polyester
82	1.00	350.1	Textile	Jacket	Used			Polyurethane, Viscose, Polyester
83	1.01	298.2	Domestic Prod.	Bandage	New	China		
84	1.03	46.52	Domestic Prod.	Lens Wipes	New			Microfiber
85	1.00	813.7	Domestic Prod.	Boot	Used	China		
86	1.00	66.67	Domestic Prod.	Mask	New	China		Polypropylene
87	0.99	770.4	Domestic Prod.	Bandage	New	Hungary		
88	1.00	166.1	Domestic Prod.	Folder	New	USA		
89	1.02	92.12	Domestic Prod.	Label	New	Mexico		
90	1.00	438.7	Domestic Prod.	Notebook Cover	New	Vietnam		
91	1.02	265.9	Domestic Prod.	Dry Erase Tape	New	USA		
92	1.01	259.0	Domestic Prod.	Folder	New	China		
93	1.01	220.7	Domestic Prod.	Shower Curtain	New			
94	1.01	293.0	Domestic Prod.	Mask	New	USA		Polyurethane Foam, Polypropylene, Polyester

Table S2. Compound names, abbreviations, internal standards, molecular formulas and molecular weights. Modified from Weber et al.[3]

Compound Name	Abbreviation	Internal Standard	Mol. Formula	Mol. Weight
Perfluorinated Carboxylates				
Perfluorobutanoate	PFBA	$[^{13}C_4]$ PFBA	$C_3F_7COO^-$	213
Perfluoropentanoate	PFPeA	$[^{13}C_2]$ PFHxA	$C_4F_9COO^-$	263
Perfluorohexanoate	PFHxA	$[^{13}C_2]$ PFHxA	$C_5F_{11}COO^-$	313
Perfluoroheptanoate	PFHpA	$[^{13}C_2]$ PFHxA	$C_6F_{13}COO^-$	363
Perfluorooctanoate	PFOA	$[^{13}C_4]$ PFOA	$C_7F_{15}COO^-$	413
Perfluorononanoate	PFNA	$[^{13}C_5]$ PFNA	$C_8F_{17}COO^-$	463
Perfluorodecanoate	PFDA	$[^{13}C_2]$ PFDA	$C_9F_{19}COO^-$	513
Perfluoroundecanoate	PFUnDA	$[^{13}C_2]$ PFUnDA	$C_{10}F_{21}COO^-$	563
Perfluorododecanoate	PFDoDA	$[^{13}C_2]$ PFDoDA	$C_{11}F_{23}COO^-$	613
Perfluorinated Sulfonates				
Perfluorobutane sulfonate	PFBS	$[^{18}O_2]$ PFHxS	$C_4F_9SO_3^-$	299
Perfluorohexane sulfonate	PFHxS	$[^{18}O_2]$ PFHxS	$C_6F_{13}SO_3^-$	399
Perfluorooctane sulfonate	PFOS	$[^{13}C_4]$ PFOS	$C_8F_{17}SO_3^-$	499
Perfluorodecane sulfonate	PFDS	$[^{13}C_4]$ PFOS	$C_{10}F_{21}SO_3^-$	599
Perfluoroalkyl Sulfonamides				
Perfluorooctane sulfonamide	FOSA	$[^{13}C_8]$ FOSA	$C_8F_{17}SO_2NH_2$	498
Perfluoroalkyl Sulfonamidoacetic Acids				
N-Methyl perfluorooctane sulfonamidoacetic acid	N-MeFOSAA	d5-N-MeFOSAA	$C_8F_{17}SO_2N(CH_3)CH_2COO^-$	570
N-Ethyl perfluorooctane sulfonamidoacetic acid	N-EtFOSAA	d3-N-EtFOSAA	$C_8F_{17}SO_2N(C_2H_5)CH_2COO^-$	584
Fluorotelomer Sulfonates				
6:2 fluorotelomer sulfonate	6:2 FtS	$[^{13}C_2]$ 6:2 FtS	$C_6F_{13}CH_2CH_2SO_3^-$	427
8:2 fluorotelomer sulfonate	8:2 FtS	$[^{13}C_2]$ 6:2 FtS	$C_8F_{17}CH_2CH_2SO_3^-$	527

Table S3. LC-MS/MS parameters for N-EtFOSAA and N-MeFOSAA. All other parameters were as defined in Weber et al.[3]

Analyte	Type	Internal Standard	Precursor Ion Mass	Product Ion Mass	Fragmentor Voltage (V)	Collision Energy (V)
Perfluoroalkyl Sulfonamidoacetic Acids						
N-EtFOSAA	Target	d3-N-EtFOSAA	584	419	110	15
N-EtFOSAA (Q1)				526		20
N-MeFOSAA	Target	d5-N-MeFOSAA	570	419	120	20
N-MeFOSAA (Q1)				512		20
Internal Standards						
d3-N-EtFOSAA	ISTD		589	419	120	15
d5-N-MeFOSAA	ISTD		573	419	120	20

Table S4. LC-MS/MS results for all PFASs quantified (nmol m^{-2}). Samples above the MQL, but with qualifiers out of range are labeled QH (qualifier above ±30% range) or QL (qualifier below ±30% range). PFCAs = perfluorinated carboxylates, PFSAs = perfluorinated sulfonates, precursors = perfluoroalkyl acid precursors.

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
1	958	76.6	837	28.5	6.98	<MQL	0.719	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.203	<MQL
2	645	<MQL	538	16.9	4.01	0.199	0.606	<MQL	0.184	<MQL	<MQL	<MQL	0.247	0.323	0.279	<MQL
3	2.56	2.31	10.8	4.77	31.4	4.54	37.9	3.19	25.0	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
4	17.6	11.1	59.9	3.44	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
5	<MQL	<MQL	4.90	3.78	34.9	3.28	25.3	2.00	9.86	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
6	<MQL	1.85	8.24	3.49	26.3	2.57	19.6	1.34	7.88	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
7	<MQL	<MQL	5.75	1.73	6.35	0.682	4.08	QH	2.31	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
8	<MQL	<MQL	9.28	1.70	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
9	<MQL	<MQL	1.96	<MQL	0.281	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
10	<MQL	<MQL	QL	QL	0.429	0.354	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.161	<MQL
11	<MQL	<MQL	38.1	QL	0.408	0.321	0.479	QL	0.542	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL
12	<MQL	<MQL	32.4	0.965	0.465	0.452	0.698	0.823	0.749	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
13	<MQL	<MQL	1.73	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
14	<MQL	<MQL	0.226	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
15	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
16	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
17	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
18	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
19	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
20	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
21	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
22	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
23	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
24	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
25	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
26	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
27	<MQL	<MQL	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
28	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
29	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
30	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
31	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
32	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
33	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
34	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
35	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
36	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
37	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
38	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
39	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
40	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
41	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
42	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
43	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
44	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
45	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
46	32.1	26.8	137	30.0	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	1.05	<MQL	<MQL	<MQL
47	5.15	5.42	47.3	12.1	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	0.17	<MQL	<MQL	<MQL
48	9.22	<MQL	37.0	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
49	35.0	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	QL	<MQL	<MQL	<MQL	<MQL	<MQL
50	<MQL	<MQL	QH	<MQL	3.12	<MQL	<MQL	<MQL	<MQL	21.3	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
51	6.51	2.77	10.2	0.465	0.524	<MQL	0.248	<MQL	<MQL	<MQL	<MQL	<MQL	QH	<MQL	<MQL	<MQL
52	<MQL	<MQL	9.82	<MQL	QL	<MQL	<MQL	<MQL	<MQL	6.00	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
53	9.04	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	5.74	QL	<MQL	<MQL	<MQL	<MQL	<MQL
54	5.43	<MQL	7.08	0.569	0.364	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QH	<MQL	<MQL	<MQL
55	6.25	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	4.83	QL	<MQL	<MQL	<MQL	<MQL	<MQL
56	<MQL	<MQL	6.46	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
57	<MQL	<MQL	0.951	<MQL	4.37	<MQL	1.01	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
58	<MQL	<MQL	<MQL	<MQL	1.09	<MQL	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
59	<MQL	<MQL	<MQL	<MQL	0.101	<MQL	<MQL	<MQL	0.120	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
60	<MQL	15.3	157	67.6	3200	61.6	1230	23.4	306	<MQL	<MQL	6.97	12.4	1.64	10.1	0.990
61	281	<MQL	0.872	<MQL	3.66	19.9	QH	5.33	QH	9.17	<MQL	QL	QH	<MQL	QL	0.0859
62	<MQL	<MQL	4.97	4.05	6.91	2.99	QH	QL	1.36	QH	<MQL	12.4	2.89	QH	QL	<MQL
63	<MQL	<MQL	1.55	0.730	3.72	0.435	QH	<MQL	0.188	QH	0.505	2.90	<MQL	0.169	1.37	<MQL
64	<MQL	<MQL	<MQL	QL	2.33	QL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	<MQL	<MQL	4.27	<MQL
65	<MQL	<MQL	QL	<MQL	2.50	QH	QH	QH	0.869	<MQL	<MQL	1.80	1.08	<MQL	QH	<MQL
66	<MQL	<MQL	0.482	<MQL	1.74	<MQL	1.63	<MQL	0.811	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
67	<MQL	<MQL	<MQL	<MQL	0.378	0.0405	0.175	<MQL	0.0518	QL	<MQL	<MQL	<MQL	<MQL	0.0397	<MQL
68	<MQL	<MQL	<MQL	<MQL	0.0915	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.09	<MQL	QL	<MQL
69	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
70	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
71	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
72	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
73	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
74	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
75	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
76	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
77	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
78	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
79	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	QL	<MQL	<MQL	<MQL	<MQL	<MQL
80	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
81	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
82	<MQL	<MQL	QL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
83	<MQL	<MQL	0.816	0.397	1.68	QH	0.679	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
84	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.0199	<MQL	<MQL	<MQL	<MQL
85	<MQL	<MQL	QL	<MQL	2.40	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.979	<MQL	<MQL	1.47	0.242
86	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
87	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
88	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
89	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL

Sample #	PFCAs									PFSAs			Precursors			
	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
90	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
91	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
92	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
93	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
94	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

Table S5. LC-MS/MS results for all PFASs quantified (µg kg^{-1}). Samples above the MQL, but with qualifiers out of range are labeled QH (qualifier above ±30% range) or QL (qualifier below ±30% range). PFCAs = perfluorinated carboxylates, PFSAs = perfluorinated sulfonates, precursors = perfluoroalkyl acid precursors.

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
1	602	59.4	773	30.5	8.51	<MQL	1.09	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.350	<MQL
2	587	<MQL	720	26.2	7.07	0.394	1.33	<MQL	0.481	<MQL	<MQL	<MQL	0.451	0.786	0.697	<MQL
3	1.61	1.80	10.0	5.11	38.3	6.20	57.4	5.31	45.3	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
4	8.16	6.38	40.9	2.72	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
5	<MQL	<MQL	3.92	3.50	36.9	3.88	33.1	2.88	15.4	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
6	<MQL	1.35	7.19	3.53	30.3	3.32	28.0	2.10	13.5	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
7	<MQL	<MQL	4.93	1.72	7.18	0.864	5.74	QH	3.88	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
8	<MQL	<MQL	7.22	1.53	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
9	<MQL	<MQL	1.48	<MQL	0.281	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
10	<MQL	<MQL	QL	QL	0.447	0.414	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.238	<MQL
11	<MQL	<MQL	32.0	QL	0.452	0.399	0.659	QL	0.891	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL
12	<MQL	<MQL	24.4	0.843	0.463	0.503	0.861	1.12	1.10	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
13	<MQL	<MQL	5.94	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
14	<MQL	<MQL	2.38	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
15	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
16	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
17	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
18	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
19	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
20	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
21	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
22	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
23	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
24	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
25	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
26	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
27	<MQL	<MQL	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
28	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
29	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
30	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
31	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
32	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
33	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
34	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
35	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
36	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
37	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
38	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
39	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
40	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
41	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
42	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
43	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
44	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
45	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
46	2.27	2.33	14.2	3.61	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	0.149	<MQL	<MQL	<MQL
47	2.78	3.61	37.5	11.1	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	0.188	<MQL	<MQL	<MQL
48	0.722	<MQL	4.25	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

	PFCAs									PFSAs			Precursors			
Sample #	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
49	3.24	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	QL	<MQL	<MQL	<MQL	<MQL	<MQL
50	<MQL	<MQL	QH	<MQL	0.485	<MQL	<MQL	<MQL	<MQL	2.40	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
51	3.62	1.90	8.31	0.440	0.564	<MQL	0.332	<MQL	<MQL	<MQL	<MQL	<MQL	QH	<MQL	<MQL	<MQL
52	<MQL	<MQL	1.31	<MQL	QL	<MQL	<MQL	<MQL	<MQL	0.764	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
53	0.766	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.683	QL	<MQL	<MQL	<MQL	<MQL	<MQL
54	2.21	<MQL	4.24	0.395	0.288	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QH	<MQL	<MQL	<MQL
55	0.561	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.608	QL	<MQL	<MQL	<MQL	<MQL	<MQL
56	<MQL	<MQL	0.809	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
57	<MQL	<MQL	1.31	<MQL	7.96	<MQL	2.29	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
58	<MQL	<MQL	<MQL	<MQL	1.74	<MQL	QH	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
59	<MQL	<MQL	<MQL	<MQL	0.270	<MQL	<MQL	<MQL	0.478	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
60	<MQL	1.15	14.1	7.01	378	8.15	180.220	3.76	53.6	<MQL	<MQL	0.994	1.51	0.267	1.68	0.141
61	244	<MQL	1.11	<MQL	6.17	37.5	QH	12.3	QH	11.2	<MQL	QL	QH	<MQL	QL	0.175
62	<MQL	<MQL	2.80	2.65	5.14	2.49	QH	QL	1.50	QH	<MQL	11.1	2.22	QH	QL	<MQL
63	<MQL	<MQL	1.72	0.944	5.48	0.717	QH	<MQL	0.410	QH	0.718	5.16	<MQL	0.343	2.85	<MQL
64	<MQL	<MQL	<MQL	QL	1.14	QL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	<MQL	<MQL	2.95	<MQL
65	<MQL	<MQL	QL	<MQL	2.48	QH	QH	QH	1.28	<MQL	<MQL	2.15	1.11	<MQL	QH	<MQL
66	<MQL	<MQL	0.458	<MQL	2.18	<MQL	2.54	<MQL	1.51	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
67	<MQL	<MQL	<MQL	<MQL	2.40	0.288	1.38	<MQL	0.487	QL	<MQL	<MQL	<MQL	<MQL	0.356	<MQL
68	<MQL	<MQL	<MQL	<MQL	0.466	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.500	<MQL	QL	<MQL
69	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
70	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
71	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
72	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
73	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
74	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

Sample #	PFCAs									PFSAs			Precursors			
	PFBA	PFPeA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFBS	PFHxS	PFOS	6:2 FtS	N-MeFOSAA	N-EtFOSAA	FOSA
75	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
76	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
77	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
78	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
79	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	QL	<MQL	<MQL	<MQL	<MQL	<MQL
80	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
81	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
82	<MQL	<MQL	QL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
83	<MQL	<MQL	0.857	0.484	2.33	QH	1.17	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
84	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.214	<MQL	<MQL	<MQL	<MQL
85	<MQL	<MQL	QL	<MQL	1.22	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	0.600	<MQL	<MQL	1.06	0.148
86	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
87	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
88	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
89	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	QL	<MQL	<MQL	<MQL	<MQL	<MQL
90	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
91	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
92	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
93	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL
94	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL	<MQL

Table S6. LC-MS/MS method quantification limits (MQL).The range of MQLs is presented below. MQL was separately determined for each batch of analyses to account for any variation in LC-MS/MS conditions.

Compound	MQL Range (ng g^{-1})
PFBA	0.32 - 1.1
PFPeA	0.080 - 1.5
PFHxA	0.43 - 1.8
PFHpA	0.11 - 1.9
PFOA	0.22 - 0.58
PFNA	0.15 - 1.6
PFDA	0.29 - 2.0
PFUnDA	0.40 - 2.6
PFDoDA	0.41 - 3.7
PFBS	0.063 - 0.31
PFHxS	0.078 - 0.40
PFOS	0.20 - 2.9
6:2 FtS	0.082 - 0.14
N-MeFOSAA	0.24 - 1.3
N-EtFOSAA	0.22 - 1.6
FOSA	0.063 - 0.45

Table S7. LC-MS/MS recovery and precision results. Average percent recovery, and relative standard deviation (RSD) of native PFAS spikes (7.2 ng) added to a plate (n = 4), curtain (n = 4), and carpet (n = 4) before extraction. PFAS concentrations were <MQL in the plate, curtain, and carpet before spiking.

	Plate (n = 4)		Curtain (n = 4)		Carpet (n = 4)	
	Average % Recovery	**% RSD**	**Average % Recovery**	**% RSD**	**Average % Recovery**	**% RSD**
PFBA	109	2.55	103	3.55	102	2.07
PFPeA	104	1.75	92.7	2.93	96	1.80
PFHxA	111	2.12	103	3.17	103	1.56
PFHpA	119	2.88	104	3.29	109	2.07
PFOA	109	1.51	103	2.53	105	1.59
PFNA	111	4.13	104	4.34	107	6.12
PFDA	123	5.47	113	3.74	114	4.38
PFUnDA	112	7.40	104	6.02	108	8.08
PFDoDA	110	8.22	105	7.26	108	3.64
PFBS	66.3	4.78	80.3	1.54	78.6	5.58
PFHxS	73.4	5.37	84.5	2.47	97.0	5.43
PFOS	85.8	4.33	93.9	4.77	88.3	6.12
6:2 FtS	99.3	28.4	80.2	4.09	84.9	5.53
8:2 FtS	104	5.42	139	8.52	22.6	6.75
N-MeFOSAA	106	5.82	101	8.52	97.9	7.54
N-EtFOSAA	126	8.97	109	4.99	116	7.85
FOSA	69.0	5.51	78.5	7.54	69.7	4.58

Table S8. XPS results. The limit of detection (LOD) is 1 atomic percent.

Sample	Average Atomic % F Pre-Extraction	Minimum/ Maximum Atomic % F	Average Weight % F Pre-Extraction	Minimum/ Maximum Weight % F	Average Atomic % F Post-Extraction	Minimum/ Maximum Atomic % F
1	9.74	8.76/10.7	13.3	12.0/14.6	12.8	12.0/14.2
2	8.45	7.09/9.80	11.6	9.76/13.4	14.2	13.7/14.7
4	17.7	16.4/19.1	23.2	21.5/24.9	18.8	17.6/20.0
7	<LOD		<LOD		N/A	
8	14.2	13.7/14.8	19.2	18.5/20.0	24.3	23.3/25.2
9	<LOD		<LOD		1.32	1.21/1.42
10	<LOD		<LOD		N/A	
11	<LOD		<LOD		N/A	
12	<LOD		<LOD		N/A	
14	28.3	27.0/29.6	36.0	34.5/37.4	18.5	18.2/18.8
15	<LOD		<LOD		N/A	
16	<LOD		<LOD		<LOD	
17	<LOD		<LOD		N/A	
18	<LOD		<LOD		N/A	
19	<LOD		<LOD		N/A	
20	<LOD		<LOD		N/A	
32	<LOD		<LOD		N/A	
33	<LOD		<LOD		N/A	
47	34.5	32.7/36.3	42.5	40.7/44.3	31.9	30.6/35.1
51	44.9	43.6/46.2	54.6	54.0/55.2	31.8	31.6/31.9
54	31.1	30.8/31.5	38.5	38.1/39.0	20.4	18.2/21.6
59	<LOD		<LOD		N/A	
61	6.38	5.64/7.12	8.60	7.63/9.48	3.91	3.25/4.56
62	<LOD		<LOD		N/A	
64	<LOD		<LOD		N/A	
65	<LOD		<LOD		N/A	
66	<LOD		<LOD		N/A	
67	7.39	6.83/7.94	9.96	9.25/10.7	5.71	5.25/6.17
68	3.70	3.61/3.79	4.99	4.81/5.16	2.32	2.17/2.46
71	<LOD		<LOD		N/A	
79	<LOD		<LOD		N/A	
81	<LOD		<LOD		<LOD	
82	<LOD		<LOD		<LOD	
83	<LOD		<LOD		N/A	
84	<LOD		<LOD		N/A	
85	<LOD		<LOD		N/A	
86	<LOD		<LOD		<LOD	
87	<LOD		<LOD		N/A	
88	<LOD		<LOD		N/A	
89	<LOD		<LOD		N/A	
90	<LOD		<LOD		N/A	
91	<LOD		<LOD		N/A	
92	<LOD		<LOD		N/A	
93	<LOD		<LOD		N/A	
94	<LOD		<LOD		N/A	

Table S9. LC-QTOF-MS results. Name, family, molecular formula, theoretical m/z, and peak area for sample methanol extracts. Data analyzed at Purdue University, Department of Agronomy, Interdisciplinary Ecological Science and Engineering.

Negative Mode					
Sample	Name	Family	Molecular Formula	Theoretical m/z	Area
1	2-perfluorohexyl ethanoic acid (FHEA)	Fluorotelomer carboxylic acids (FTCAs)	C8H3F13O2	376.98472	2880
	Perfluoro-n-pentanoic acid (PFPeA)	Perfluoroalkyl carboxylic acids (PFCAs)	C5HF9O2	262.97546	2186
	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	44303
	3-Perfluoropentyl propanoic acid (FPePA)	Fluorotelomer carboxylic acids (FTCAs)	C8H5F11O2	341.00356	1822
	2H-perfluoro-2-octenoic acid (FHUEA)	Fluorotelomer unsaturated acids (FTUA)	C8H2F12O2	356.97849	15413
	10:2 fluorotelomer sulfonate (10:2 FTS)	Fluorotelomer sulfonic acids (FTSs)	C12H5F21O3S	626.95458	596
	6:2 fluorotelomer sulfonate (6:2 FTS)	Fluorotelomer sulfonic acids (FTSs)	C8H5F13O3S	426.96736	294
	Perfluoro-n-octanoic acid (PFOA)	Perfluoroalkyl carboxylic acids (PFCAs)	C8HF15O2	412.96588	1575
2	2-perfluorohexyl ethanoic acid (FHEA)	Fluorotelomer carboxylic acids (FTCAs)	C8H3F13O2	376.98472	1948
	Perfluoro-n-pentanoic acid (PFPeA)	Perfluoroalkyl carboxylic acids (PFCAs)	C5HF9O2	262.97546	1820
	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	45527
	2H-perfluoro-2-octenoic acid (FHUEA)	Fluorotelomer unsaturated acids (FTUA)	C8H2F12O2	356.97849	9946
	Perfluoro-n-butanoic acid (PFBA)	Perfluoroalkyl carboxylic acids (PFCAs)	C4HF7O2	212.97865	4261
	10:2 fluorotelomer sulfonate (10:2 FTS)	Fluorotelomer sulfonic acids (FTSs)	C12H5F21O3S	626.95458	439
	6:2 fluorotelomer sulfonate (6:2 FTS)	Fluorotelomer sulfonic acids (FTSs)	C8H5F13O3S	426.96736	321
	Perfluoro-n-octanoic acid (PFOA)	Perfluoroalkyl carboxylic acids (PFCAs)	C8HF15O2	412.96588	706
	3-Perfluoropentyl propanoic acid (FPePA)	Fluorotelomer carboxylic acids (FTCAs)	C8H5F11O2	341.00356	1216
4	2H-perfluoro-2-octenoic acid (FHUEA)	Fluorotelomer unsaturated acids (FTUA)	C8H2F12O2	356.97849	2635
	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	6597
	Perfluoro-n-butanoic acid (PFBA)	Perfluoroalkyl carboxylic acids (PFCAs)	C4HF7O2	212.97865	286
	Perfluoro-n-pentanoic acid (PFPeA)	Perfluoroalkyl carboxylic acids (PFCAs)	C5HF9O2	262.97546	512
8	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	327
14	Not Detected				

47	Perfluoro-n-pentanoic acid (PFPeA)	Perfluoroalkyl carboxylic acids (PFCAs)	C5HF9O2	262.97546	267
51	6:2, 8:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C18H9F30O4P	888.96811	483
	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	607
	Perfluoro-n-octanoic acid (PFOA)	Perfluoroalkyl carboxylic acids (PFCAs)	C8HF15O2	412.96588	170
	6:2, 6:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C16H9F26O4P	788.97450	371
	8:2, 8:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C20H9F34O4P	988.96173	175
54	Perfluoro-n-hexanoic acid (PFHxA)	Perfluoroalkyl carboxylic acids (PFCAs)	C6HF11O2	312.97226	297
61	Perfluorononane sulfonate (PFNS)	Perfluoroalkyl sulfonic acids (PFSAs)	C9HF19O3S	548.92647	100681
	Perfluoro-n-butanoic acid (PFBA)	Perfluoroalkyl carboxylic acids (PFCAs)	C4HF7O2	212.97865	838
	Perfluoro-n-nonanoic acid (PFNA)	Perfluoroalkyl carboxylic acids (PFCAs)	C9HF17O2	462.96268	1179
	Perfluoro-n-undecanoic acid (PFUnDA)	Perfluoroalkyl carboxylic acids (PFCAs)	C11HF21O2	562.95629	845
	6:2, 6:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C16H9F26O4P	788.97450	39
67	8:2, 8:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C20H9F34O4P	988.96173	301
	Perfluoro-n-octanoic acid (PFOA)	Perfluoroalkyl carboxylic acids (PFCAs)	C8HF15O2	412.96588	348
	8:2, 10:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C22H9F38O4P	1088.95534	143
	6:2, 8:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C18H9F30O4P	888.96811	61
68	6:2, 6:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C16H9F26O4P	788.97450	780
	6:2, 8:2 fluorotelomer phosphate diester	Polyfluoroalkyl phosphoric acid diesters (diPAPs)	C18H9F30O4P	888.96811	236
		Positive Mode			
47	Perfluoropentane sulfonamido propyl dimethyl quaternary amine propanoate	Perfluoroalkyl sulfonamide amino carboxylates (PFSaAmA)	C13H17F11N2O4S	507.08116	2239
54	Perfluoropentane sulfonamido propyl dimethyl quaternary amine propanoate	Perfluoroalkyl sulfonamide amino carboxylates (PFSaAmA)	C13H17F11N2O4S	507.08116	445

Figures

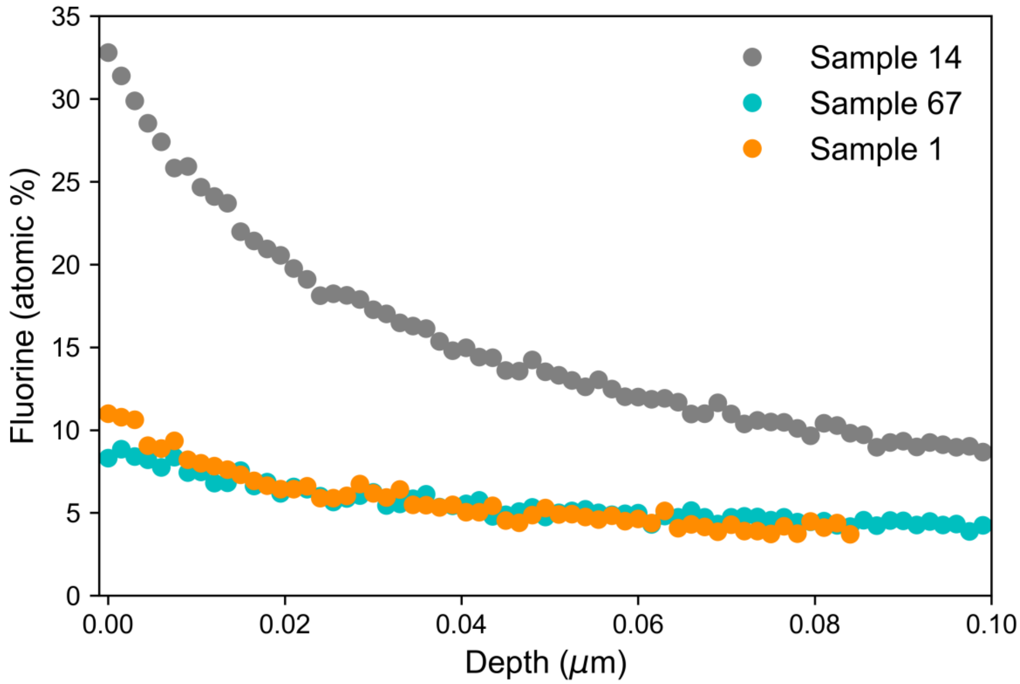

Figure S1. Depth profiles of sample 14 (food bag), sample 67 (used jacket), and sample 1 (bowl). Each point is a XPS measurement taken in between etching intervals (etching completed with an argon ion beam with 10 second intervals). The depth of etching for a tantalum pentoxide reference is plotted.

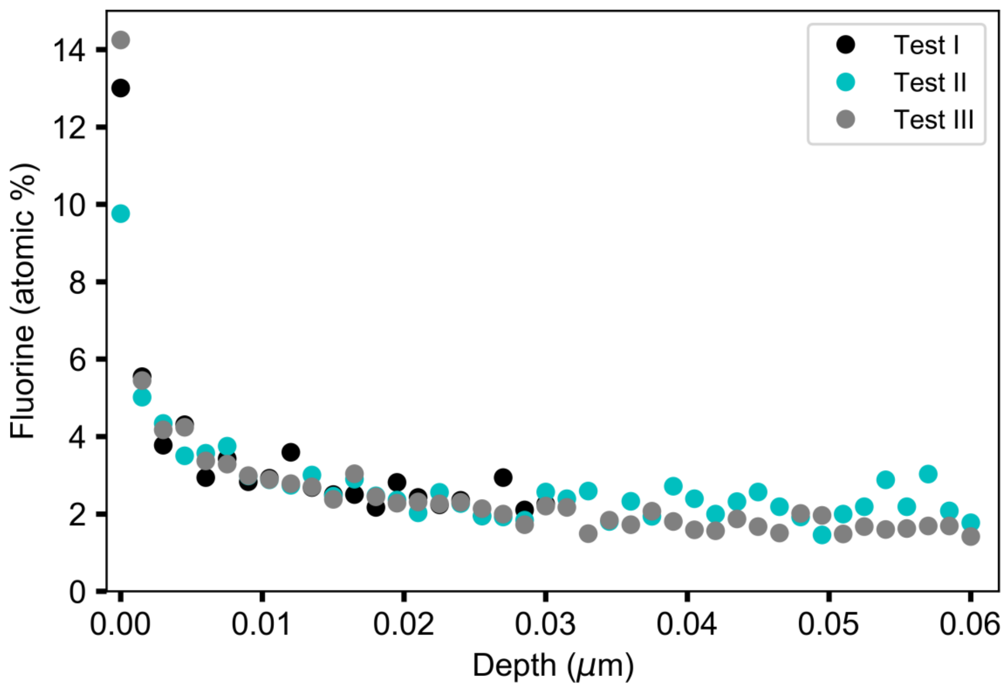

Figure S2. Repetitive depth profile analysis of sample 4 (a takeout box). Each point is a XPS measurement taken in between etching intervals (etching completed with an argon ion beam with 10 second intervals). The depth of etching for a tantalum pentoxide reference is plotted.

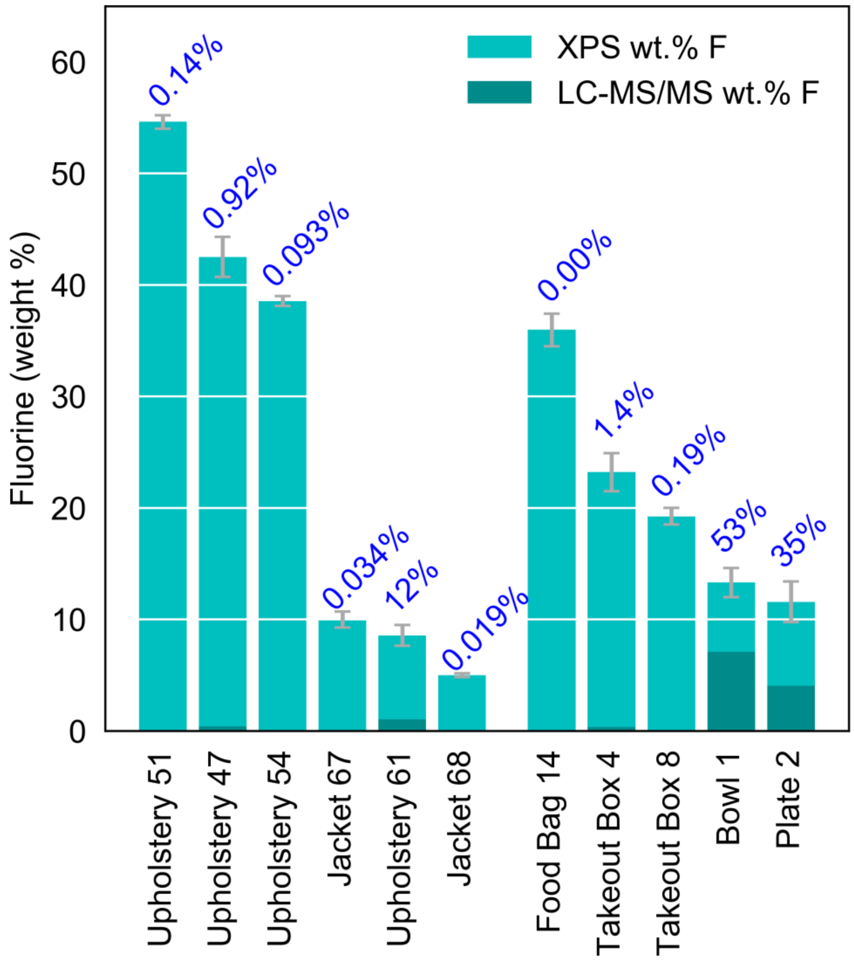

Figure S3. XPS wt.% F and LC-MS/MS wt.% F comparison. Whiskers are the minimum and maximum wt.% F from XPS, the bar is the average wt.% F. The percent of fluorine in the XPS explained by LC-MS/MS is labeled in blue at the top of each bar.

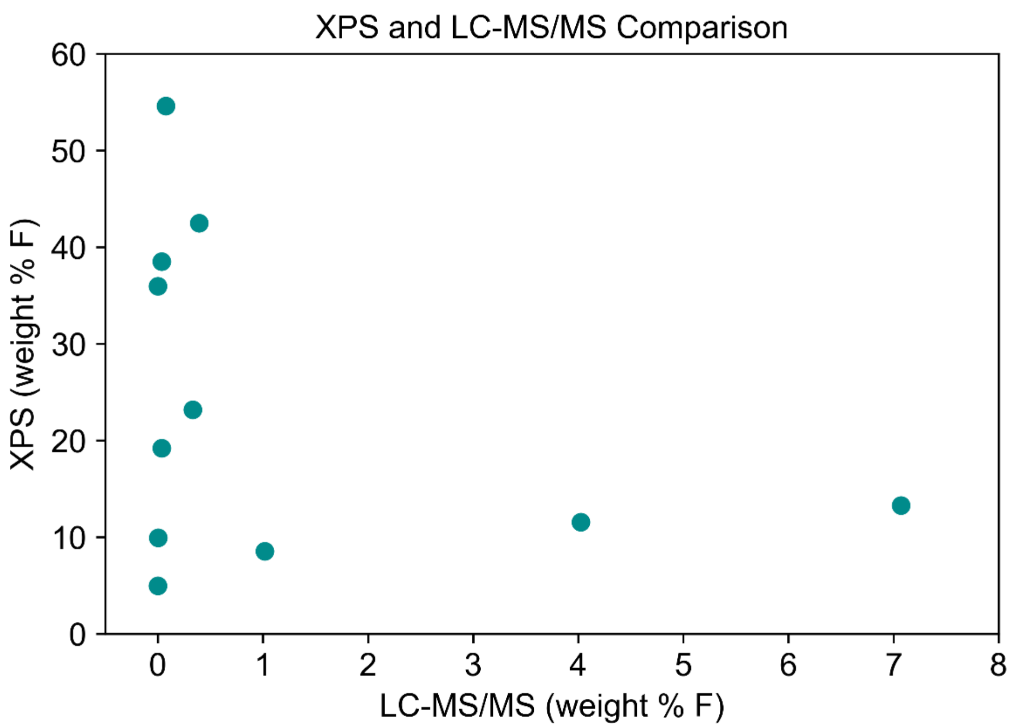

Figure S4. Comparison of LC-MS/MS and XPS data.

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