Supporting Information

For

Per- and Polyfluoroalkyl Substances in Canadian Fast Food Packaging

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Section S1. Product Sampling.

Tables S3 and S4 describe the food packaging items sampled. All samples collected during sampling round 1 (February to March 2020) were measured using only particle-induced gamma ray emission (PIGE) spectroscopy. All samples collected during sampling round 2 (August 2020) were measured using PIGE, targeted LC-MS/MS, targeted GC-MS, and non-target LC-MS.

At each retailer, clean packaging un-soiled by food was collected with minimal touching and promptly sealed in an individual zip-lock plastic bag to avoid cross-sample contamination. For quality control, duplicate contact materials were collected for every 10th item - yielding 10 field duplicates total.

In the lab, each sample was prepared donning clean disposable gloves on a clean PFAS-free aluminum foil work surface, after wiping down all used surfaces and tools with isopropyl alcohol. Materials that contained food residue were gently rinsed first with reverse osmosis (RO) water. The packaging was measured for thickness using an electric caliper before being cut into 2 x 2 cm^2 coupons using an aluminum size template to minimize touching the sample.

Lab blanks were prepared for every 10^{th} sample from a 2 x 2 cm^2 sample of clean Kimwipe. Field blanks were identical samples collected from retailers at the same time. For quality control, a lab duplicate sample was prepared for every 10^{th} sample prepared – yielding 5 in total. Each sample and lab blank was sealed into individual Ziplock bags and shipped to the University of

Notre Dame for analysis via PIGE, or Indiana University for analysis via targeted GC-MS and LC-MS/MS. Samples were prepared via the same method for non-targeted LC-MS analysis at the University of Toronto.

Section S2. PIGE Analysis.

For quality assurance/quality control (QA/QC) of sample variation, sample preparation, and analysis techniques, field duplicates and lab duplicates were analyzed blind to the analyst. In addition, reproducibility was tested by reanalyzing 5% of samples 2 to 5 times.

In general, PIGE involves irradiating each sample over 3 minutes using an ion beam of 10 nA of 3.4 MeV protons. This irradiation causes identifiable γ-rays to emit from the sample due to the de-excitation of 19F at 110 and 197 keV allowing for the counting of background-subtraction integrations. These summed γ-rays can then be converted to total F concentrations, as expressed in ppm F, by generating calibration curves using inorganic F standards, for which we can relate concentration of F to the PIGE counts. Because PIGE irradiated photons are limited to reaching 220 μm depths into solids, varying sample thickness can lead to varying fluorine signal response. To account for varying thickness in fast food packaging samples, quantification of total fluorine was performed using sample thickness to perform thickness-dependent quantification. This approach was previously detailed in Xia *et al.* (2022).[1] In this work textiles of various thickness were spiked with solutions of inorganic fluoride to generate calibration curves for each textile. The slope measured from each calibration curve was plotted against textile thickness to relate sample thickness to signal response, allowing for application to samples with known thickness. For samples above the penetration depth of 220 μm it is necessary to test both sides of thicker samples to measure fluorine content on each side of the material (taking the higher F side to represent the packaging subsampled from).

Four samples were chosen as field duplicates. Five samples were chosen as lab duplicates, and duplicate samples were collected from the same item. Nine samples were collected twice, once during each of the two sampling periods. Table S9 summarises the results of these tests. Five Kimwipe blank samples were collected and measured, all of which were below the detection limit for the method. These blanks were used in the formation of the calibration curve for the samples.

Section S3. Sample Preparation and Instrumental Analysis for Targeted Analysis.

Tables S5-S8 summarize all compounds analyzed during targeted analysis, including their molecular weights, formulas, retention times, precursor and product ions (LC-MS/MS), and qualifier and quantifier ions (GC-MS). Additional instrumental parameters can be found in previous studies.[2,3] In brief, each sample was spiked with 20 ng each of surrogate standard and extracted first with 3 mL of 4:1 hexane/isopropanol twice and then with 3 mL of 1:1 methanol/acetonitrile twice. Each extraction step was performed using sonication for 30 min followed by centrifugation at 3000 rpm for 5 min. The supernatants were combined and reduced

to ~5 mL under nitrogen. 100 mg of Envi-Carb was added to the extract for clean-up, and the mixture was vortexed for 1 min and centrifuged for 5 min. The resulting sample was reduced to 500 μL under nitrogen, and then filtered using a centrifuge filter. The filtrate was transferred into a 1-mL polypropylene vial and spiked with 50 ng each of the internal standards used for quantitation. LC-MS/MS analysis was done using an ultrahigh performance LC coupled with a triple-quadrupole MS (Agilent 1290 Infinity II UPLC−6470 QQQ-MS) in negative electro- spray ionization mode. GC-MS analysis was performed on an Agilent 7890 GC−5977B PCI-MS operated in the positive chemical ionization mode. For extracts of the five samples from the hydrolysis assay, only neutral PFAS were measured.

A procedural blank and a matrix spike sample were processed along with each batch sample to evaluate possible contamination from laboratory operations and the performance of the method. The recoveries of surrogate standards were in the range of 60−130%. Samples were corrected for recovery using the appropriate surrogate standards. After this correction, matrix spike recoveries of individual analytes were all within 80%−115%. Additionally, the data were blank corrected by subtracting the corresponding average solvent blank on a mass basis. The LODs were defined as the average solvent blank + 3 × standard deviation (n = 5) or the amount of chemical generating a signal-to-noise of 5 if the compound was not detected in the solvent blanks. In addition, five Kimwipe field blanks were analyzed, and all the results were below the corresponding LODs. In the present study, one sample, a plastic-lined paper popcorn bag, was chosen as a field duplicate (see Table S8). Only compounds which were measured above the LOD are reported.

Section S4. Sample Preparation and Instrumental Analysis for Hydrolysis.

In brief, 1 mL 1 mol/L NaOH solution in methanol/water (90:10) was added to a 15 ml glass vial containing ~30 mg small pieces of samples, and then spiked with 40 ng each of the surrogate standards. The vial was vortexed for 1 min and placed in an oven at 60 ºC for 16h. After the vial was cooled to room temperature, the remaining solution was transferred to a clean polypropylene (PP) vial, and 0.6 mL of a mixture of methyl tert-butyl ether/n-hexane (1:1, v/v) and 2 mL of LC/MS grade water were added. Samples were shaken for 30 minutes, and the organic (bottom) layer was transferred to a new clean PP vial with a glass pipette. Anhydrous Na_2SO_4 was added to remove the water in the sample until the organic layer became clear. The extracts were transferred to a plastic Eppendorf vial with 50 mg anhydrous $NaSO_4$, and left to dry for 1 h with occasional shaking. Finally, the extracts were transferred into a 1-mL PP vial, spiked with 100 ng of internal standards and analyzed using GC/MS for targeted analysis of FTOHs and fluorotelomer (meth)acrylates (FT(M)Acs) as described in Section S3.

Section S5. Sample Preparation and Instrumental Analysis for Non-targeted Analysis.

Each sample was placed in a 15 mL PP tube with 10 mL HPLC-grade methanol. A procedure blank was prepared by using an empty PP tube without sample. The mixtures were shaken vigorously for 1 h, sonicated at 40 °C for 2 h, and centrifuged at 4000 g for 10 min. The extracts were decanted into 15 mL PP tubes, evaporated under N_2, and reconstituted in 200 μL of HPLC-grade methanol. Finally, the extract was filtered through an Acrodisc GHP Syringe Filter (pore

size 0.2 μm) before LC-MS analysis. For recovery calculations, three representative material types were chosen: a paper bag, a plastic-lined paper bag, and a molded fiber bowl. These samples were placed in a 15 mL PP tube and spiked with 20 ng each of PFCA compounds ranging from PFBA-PFTeDA. The tubes were then sealed and stored at room temperature in the dark for 24 hours, before being extracted as described above, for a final theoretical concentration of 100 μg/L. Each spike was performed three times, and the average value is reported. Procedure blanks were obtained by running solvent through the entire extraction and analysis process.

PFAS analysis followed the protocol described in a previous study.[4] All food packaging samples were analyzed using a Q Exactive mass spectrometer equipped with a Vanquish UHPLC system (Thermo Fisher Scientific, USA). Mobile phase A was 2 mM ammonium acetate in ultrapure water and B was HPLC grade methanol. The injection volume was 2 μL, and the PFAS were separated by a Hypersil Gold C18 column (100 × 2.1 mm, 3 μm, Thermo Scientific). The temperature of the column chamber was set to 40 °C, and the sampler chamber was 4 °C. The initial HPLC gradient was 10% B which was increased to 100% over 7 min, and was held static for 4.5 min. Then the gradient of B was decreased to 10% and held for 1 min to equilibrate. The flow rate of mobile phases was 0.3 mL/min. Data were acquired in full MS and data-independent acquisition (DIA) modes. The parameters of mass spectrometry were one full MS^1 scan (150-1000 *m/z*) switch mode recorded at resolution R = 70,000 (at *m/z* 200) with a maximum of 3×10^6 ions, which were collected within 100 ms. The full MS^1 scan was followed by DIA MS^2 scans (150-450, 450-750 *m/z*) recorded at resolution R = 35,000 (at *m/z* 200), with a maximum of 1×10^5 ions, which were collected within 60 ms. DIA data were acquired by using 10 *m/z* isolation windows per MS^2 scan. Therefore, in total, there were thirty 10- *m/z* wide windows between 150-450 and 450-750 *m/z*. The mass spectrometry was conducted in negative electrospray ionization mode, with spray voltage of 3kV, sheath gas flow rate of 30 L/h, auxiliary gas flow rate of 6 L/h, and capillary temperature of 300 °C.

All nontarget data analyses were accomplished with an in-house R script. Raw MS files were converted to mzXML format using MSconvert (ProteoWizard). The peaks were detected with the 'XCMS' R package[5] at a mass tolerance of 2.5 ppm. The peak features were matched across samples with a mass tolerance of 2.5 ppm and retention time window of 20 seconds after retention time adjustment. Only the features detected with intensities 3 times higher than those in procedure plan were kept for subsequent data analysis. The differentiated features were searched against 1,763 PFAS curated by Liu et al.[6] with a mass tolerance of 3 ppm. 89 PFAS were initially detected by exact mass matching. To further exclude false identifications, we manually interpreted the MS^2 spectra of each PFAS. Only 10 PFAS with characteristic fragments detected from MS^2 spectra were considered as reliable identifications, and their confidence levels were assigned according to the Schymanski et al. scale.[7] For confidence level 1, the identity was confirmed via authentic standard. For level 2, the structures were supported by diagnostic fragments, e.g., $[M\text{-}4HF\text{-}CO_2\text{-}H]^-$ and $[M\text{-}3HF\text{-}CO_2\text{-}H]^-$ for fluorotelomer carboxylic acids (FTCAs). For level 3, the structures were supported by MS^2 spectra with less informative fragments, e.g., $[C_nF_{2n+1}]^-$ for H-substituted polyfluoroalkyl (linear) carboxylic acids, or by literature references in which the compound was assigned a confidence level of 3.

Potential homologue series of PFAS were further searched in MS^1 within a mass tolerance of 3 ppm by differentiating $(CF_2)_n$ for those PFAS beyond the current database. Retention times of compounds within these homologue series were used as part of the confirmation of their

identities. For example, shorter-chain compounds within a series should have correspondingly shorter retention times. The identities of all PFAS and their confidence levels are provided in Table S14.

For NTA data analysis, chromatograms for each of the PFCA standards used for spike and recovery analysis and method optimization are displayed in Figure S1, alongside the relative intensity of each compound at 100 ppb.

Figure S2 shows spike and recovery values for PFCA standards ranging from PFBA - PFTeDA. The recoveries for each compound ranged from 82% - 176% after blank subtraction. Recoveries appeared to be material specific, with the molded fiber bowl having the lowest average recovery values and the plastic-lined paper bag having the highest. As the primary aim of nontargeted analysis is to detect unknown PFAS beyond targeted analysis, rather than quantitative measurement, sample cleanup was not performed to reduce the loss of compounds. The current recoveries without significant matrix suppression effects are acceptable. A spiked solvent sample which was put through the extraction process showed that no loss of compounds occurred during the extraction procedure.

Section S6. PIGE Comparison Summary.

Schultes et al. measured between 391 and 4088 ppm F in three French fry packages.[8] Robel et al. measured total F ranging from below the LOD to 430 nmol F/cm^2 for 5 fast food packaging.[9] The total F was above the LOD for 3/5 samples.[9] Ritter et al. measured total F in 25 fast food packaging.[10] 13 of the samples had total F below the LOD, while the remaining 12 contained between 57-331 nmol F/cm^2.[10] In a widescale study of fast-food paper and paperboard food contact packaging collected from multiple brands across the United States, Schaider et al. detected F > 16 nmol of F/cm^2 in 40% of 328 samples.[11] This slightly lower rate of detection versus our finding of 45% is in line with differences in sampling methods between the Schaider et al. study and ours. Schaider et al. aimed to represent a broad distribution of packaging types and geographic locations, whereas our study aimed to conduct in depth analyses on a smaller number of high-F samples.

Section S7. Discussion of Non-Targeted Analysis.

Group A. Several diagnostic fragments of 6:2 FTUCA were observed, including [M-HF-CO_2-H]$^-$ and [M-CH_2-HF-CO_2-H]$^-$, consistent with previous studies on MS^2 fragmentation of 6:2 FTUCA.[12] This identification was further confirmed by comparing retention time and MS^2 spectra with those obtained using a commercially available standard (Figure S7, Table S14). The compound was detected in all molded fiber bowl samples, despite coming from different retailers.

Group B. Both 5:3 FTCA and 6:3 FTCA were supported by the presence of multiple diagnostic fragments (Figure S8, Table S14). The fragmentation pattern for this group was consistent with previous studies (e.g., [M-C_2H_4-CO_2-H]$^-$ at *m/z* 318.9797), which allowed for localization of the H-substitutions.[9,11,13]

Group C. The MS^2 spectra clearly demonstrated the presence of ether bond ($[M-2HF-OCH_2-CO_2-H]^-$) and hydrocarbons ($[M-4HF-CO_2-H]^-$) (Figure S9, Table S14).[14] It was not possible to determine the exact location of the hydrogen substitutions.

Group D. The fragments available in the MS^2 spectra for the compounds were $[M-CO_2-H]^-$, and $[C_nF_{2n+1}]^-$ (Figure S10, Table S14). These match fragments found in the literature but are not characteristic for identification of the location of the double bond. [14,15]

Group E. The fragments available in the MS^2 for the n=9, 11, 12, and 13 compounds were $[M-HF-CO_2-H]^-$ and $[C_nF_{2n+1}]^-$ (Figure S11, Table S14). They matched the fragments listed in the literature, but it was not possible to locate the H-substitution beyond determining that it was not on the terminal three carbons.[12,14,16,17] The n=6, 8, and 10 compounds only had the $[M-HF-CO_2-H]^-$ in the MS^2 spectra, meaning that it was also not possible to confirm the location of the H-substitution. The n=5 and 7 compounds did not have any identifying fragments due to their low abundances, but their identities were supported by their retention time order relative to other homologues.

Group F. In-source fragments for this compound were detected in the MS^1 spectrum (Figure S12, Table S14). In particular, we found $[M-HF-2CO_2-H]^-$ in the MS^1 of all compounds in this group. The presence of two CO_2 neutral loss supported two carboxylates contained in these PFAS. The $[M-HF-2CO_2-H]^-$ in source fragment was also detected for all compounds in this group except n=7. The n=10 compound had a C_7F_{13} fragment in the MS^2, which matched that observed in the MS^2 of a standard in a previous publication.[14]

Section S8. Discussion of Canadian Food Packaging Regulations and Example Probable Daily Intake Calculation.

The onus for ensuring the safety of food packaging in Canada lies with manufacturers, who can opt to seek a pre-market assessment by the Health Products and Food Branch of Health Canada, as described in Section B.23.001 of the Food and Drugs Act and Regulations (SI Section S7).[18] If a pre-market assessment for a new additive or single constituent used in food packaging is requested, Health Canada asks for information on the product identity, proposed usage, migration data (e.g., using a 10 or 95% ethanol extraction), and toxicological data.

The guidance does not specify whether this threshold is for a single or total PFAS; if it pertains to a single PFAS, this toxicological information is not required to account for possible addition/synergistic effects of multiple PFAS.[19] This method also cannot account for unknown chemicals and unintentionally added chemicals, which would not be detected by targeted analysis and could be of concern.[19] Further, the analytical method is not prescribed, i.e., the method may not include compounds identified here using targeted and non-targeted analysis.

If toxicological information must be supplied, such information is very limited for most compounds listed here, including the ubiquitous 6:2 FTOH which is both toxic and bioaccumulative.[20–22]

The probable daily intake threshold is defined as 0.025 µg kg bw^{-1} d^{-1}.[18] Above, this, there are tiers of concern from "very low" at 0.025-0.1 to high at >25 µg kg bw^{-1} d^{-1}.

Targeted analysis detected between 1-1.6 µg/g of total PFAS per molded ("compostable") bowl, each of which weighed roughly 20 g. This means that each bowl contained roughly 20-32 µg of total PFAS. Migration rates of PFAS from food packaging vary according to the type of food packaging, temperature, contact time, extraction process used and PFAS chain length, with higher migration rates for short-chain compounds, e.g., PFHxA, PFHpA.[23] For example, Yuan et al. saw migration efficiencies ranging from 0.004% - 18.1% for different PFAS from molded fiber bowls using a 10% ethanol solution.[14] However, migration rates can be up to 100% for short chained compounds using an acetic acid extraction.[25]

Assuming a low migration rate of 1% for all compounds results in an estimated daily intake of 0.003-0.05 µg kg bw^{-1} d^{-1} for a molded fiber bowl, which ranges from below the threshold of concern to reach the very low concern level for the daily intake threshold set out by the Health Products and Food Branch of the Government of Canada. Assuming higher migration rates of 20-100% results in an estimated daily intake from 0.067-0.53 µg kg bw^{-1} d^{-1}. The highest estimate is within the range of low concern (0.1-2.5 µg kg bw^{-1} d^{-1}). For Probable Daily Intake values estimated according to regulations that exceed the very low and low threshold values, a submission to Health Canada must be accompanied by an estimate of toxicity from a QSAR (very low concern) to a short-term genotoxicity 28-day rodent feeding test (low concern).[18]

This type of reporting is likely for individual compounds (not the total PFAS), does not consider unexpected impurities and degradation products not traditionally measured by targeted analysis, and does not address the lack of toxicity data or vetted QSARs for many of the compounds reported in this study.

Section S9. Calculation of total F concentration (µg F/m^2) from PFAS concentration (ng/g) analyzed by targeted analysis in products

We used equation 1 to calculate the total F concentration in individual PFAS based on sample area for targeted analysis of extracts or after hydrolysis assay. We then summed all the total F concentrations of individual PFAS to get the total F concentration analyzed by targeted analysis of extracts or after hydrolysis assay in product. Equation 1 was used to convert between units of µg F/m^2 and ng/g.

$$C_i\ (\mu g\ F/m^2) = \frac{C_i\ (ng/g) \times m\ (g)}{A\ (m^2)} \times \frac{19 \times N_F}{M_W} \times 10^{-3} \quad (1)$$

where C_i (µg F/m^2) is the concentration of total F in individual PFAS compound *i* based on area, C_i (ng/g) is the concentration of individual PFAS compound *i*; m (g) is the mass of sample extracted for targeted analysis; A (m^2)) is the area of sample used for targeted analysis; M_W is the molecular weight of compound *i* (the atomic weight of F is 19 g/mol), g/mol; N_F is the number of F in compound *i*'s molecular formula; and 10^{-3} is the unit conversion from ng to µg.

Tables

Table S1. Summary of previous studies of fast food packaging using PIGE spectroscopy.

Year	Sample Type	Number of Samples	Location of Samples	Results (% samples with high, low, no F)
Schultes et al., 2019[8]	Paper bag, takeout cardboard container, microwave popcorn bags	N=9	Sweden	All 8 samples showed F hits, consistent across all 3 methods used to detect total fluorine.
Schaider et al., 2017[11]	Food contact paper, noncontact paper, food contact paperboard, paper cups, other beverages, miscellaneous	N=407	US (Washington, Massachusetts, Michigan, California, Washington DC)	Of the 407 samples, 33% had detectable total F concentrations. 46% of food contact papers and 20% of paperboard samples had detectable fluorine.
Robel et al., 2017[9]	Fast food packaging, popcorn bags	N=6 food contact materials	Washington, US	4/6 food contact materials had total F above LOD
Ritter et al., 2017[10]	Wrappers for tacos, pizza, burgers, sandwiches. Paperboard from fast-food fries, chicken, drink cups, etc. Popcorn bags	N=50 food contact materials	Washington, US	8 of the food contact paper samples showed <LOQ, 10 of food contact papers ranged from 57-331 nmol F/cm^2. Paperboard food-contact items and popcorn bags also showed hits ranging from 42-285 nmol F/cm2 and 161-445 nmol F/cm2, respectively.

Table S2. Summary of targeted PFAS measured in fast food packaging in previous studies. Please see additional excel file.

Table S3. Sample type and descriptions. Specific sample numbers are provided for all samples which underwent targeted and non-targeted analyses. Sampling round one refers to samples collected between February to March 2020, and round two refers to samples collected in August 2020.

Sample Type	Description	Sampling Round
Burger	Paper wrapper	1
Sub sandwich	Paper wrapper	1
Burrito roll	Paper wrapper	1
Donut	Paper bag	1
Burger	Paper wrapper	1
Pastry	Paper bag	1
Pastry	Paper bag	1
Burger	Paper bag	1
Burger	Paper wrapper	1
Burrito bowl 1	Molded fiber bowl	1, 2
Popcorn bag 7	Paper bag	1, 2
Popcorn bag 8	Paper bag, plastic lining	1, 2
Pastry bag 5	Paper bag	1, 2
Pita wrap	Paper wrapper	1
Pita wrap	Paper wrapper	1
Sandwich	Paper wrapper	1
Burger	Paper wrapper	1
Burger	Paper wrapper	1
Burger	Paper wrapper	1
Sandwich	Paper wrapper	1
Bagel	Paper bag	1
Sub sandwich	Paper wrapper	1

Sub sandwich	Paper wrapper	1
Donut bag	Paper bag	1
Sandwich	Paper wrapper	1
Salad bowl 3	Molded fiber bowl	1, 2
Pita wrap	Paper wrapper	1
Pita wrap	Paper wrapper	1
Burger	Paper wrapper	1
Burrito roll	Paper wrapper	1
Sub sandwich	Paper wrapper	1
Donut bag 4	Paper bag	1, 2
Burger	Paper wrapper	1
Pastry	Paper bag	1
Pastry	Paper bag	1
Burrito bowl 2	Molded fiber bowl	1, 2
Sandwich	Paper wrapper	1
Sandwich	Paper bag	1
Salad	Molded fiber bowl	1
Burrito roll	Paper wrapper	1,2
Pastry 6	Paper bag	1
Pastry	Paper bag	1

Table S4. List of analytes included in targeted analysis for LC-MS/MS and GC-MS along with their Limits of Detection (LODs). A description of the calculation of LODs is given above. The LODs ranged from 0.001 ng for pefluoropentanesulfonic acid (PFPeS) to 0.62 ng for Perfluoro-2-propoxypropanoic acid (GenX). [3] PFAS indicated in shaded area were analyzed by targeted and non-targeted analysis.

LC-MS/MS								GC-MS	
Analyte	LOD (ng)	Analyte	LOD (ng)	Analyte	LOD (ng)	Analyte	LOD (ng)	Analyte	LOD (ng)
PFPrA	0.008	PFTeDA	0.007	PFECHS	0.003	FHxSA	0.003	4:2 FTOH	0.37
PFBA	0.01	PFHxDA	0.01	Cl-PFOS	0.005	FOSA	0.003	6:2 FTOH	0.22
PFPeA	0.009	GenX	0.62	6:2 FTCA	0.076	MeFOSA	0.004	8:2 FTOH	0.16
PFHxA	0.007	PFPrS	0.007	8:2 FTCA	0.08	EtFOSA	0.002	10:2 FTOH	0.22
PFHpA	0.005	PFBS	0.001	10:2 FTCA	0.074	6:2 PAP	0.03	MeFOSE	0.23
PFOA	0.004	PFPeS	0.001	4:2 FTSA	0.003	8:2 PAP	0.028	EtFOSE	0.22
PFNA	0.007	PFHxS	0.002	6:2 FTSA	0.005	6:2 diPAP	0.002	6:2 FTAc	0.032
PFDA	0.007	PFHpS	0.005	8:2 FTSA	0.003	6:2/8:2 diPAP	0.001	8:2 FTAc	0.039
PFUnDA	0.006	PFOS	0.005	6:2 Cl-PFESA	0.002	8:2 diPAP	0.001	10:2 FTAc	0.041
PFDoDA	0.009	PFNS	0.002	8:2 Cl-PFESA	0.004	6:2 FTUCA	0.001	6:2 FTMAc	0.031
PFTrDA	0.006	PFDS	0.008	FBSA	0.002	5:3 FTCA	0.001	8:2 FTMAc	0.034

Table S5. List of targeted PFAS measured by LC-MS/MS.

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
PFPrA	Perfluoropropanoic acid	422-64-0	$C_3HF_5O_2$	0.532	164.03	162.9	64	119.0	5	F_3CCF_2COOH
PFBA	Perfluorobutanoic acid	375-22-4	$C_4HF_7O_2$	2.243	214.04	213.0	64	169	5	$F_3C(CF_2)_2COOH$
PFPeA	Perfluoropentanoic acid	2706-90-3	$C_5HF_9O_2$	3.518	264.05	263.0	64	218.9 140.8	5 5	$F_3C(CF_2)_3COOH$
PFHxA	Perfluoro-n-hexanoic acid	307-24-4	$C_6HF_{11}O_2$	5.008	314.05	313.0	73	268.9 119	5 21	$F_3C(CF_2)_4COOH$
PFHpA	Perfluoro-n-heptanoic acid	375-85-9	$C_7HF_{13}O_2$	6.646	364.06	363.0	78	319 169	5 17	$F_3C(CF_2)_5COOH$
PFOA	Perfluoro-n-octanoic acid	335-67-1	$C_8HF_{15}O_2$	8.186	414.07	413.1	83	369 169	5 17	$F_3C(CF_2)_6COOH$
PFNA	Perfluoro-n-nonanoic acid	375-95-1	$C_9HF_{17}O_2$	9.542	464.08	463.1	83	419 218.9	5 17	$F_3C(CF_2)_7COOH$
PFDA	Perfluoro-n-decanoic acid	335-76-2	$C_{10}HF_{19}O_2$	10.712	514.08	513.0	93	468.9 269	5 17	$F_3C(CF_2)_8COOH$
PFUnDA	Perfluoro-n-undecanoic acid	2058-94-8	$C_{11}HF_{21}O_2$	11.725	564.09	563.0	102	518.9 268.9	5 17	$F_3C(CF_2)_9COOH$

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
PFDoDA	Perfluoro-n-dodecanoic acid	307-55-1	$C_{12}HF_{23}O_2$	12.601	614.10	613.0	102	569 269	9 21	$F_3C(CF_2)_{10}COOH$
PFTrDA	Perfluoro-n-tridecanoic acid	72629-94-8	$C_{13}HF_{25}O_2$	13.347	664.11	663.1	107	619 169	9 29	$F_3C(CF_2)_{11}COOH$
PFTeDA	Perfluoro-n-tetradecanoic acid	376-06-7	$C_{14}HF_{27}O_2$	13.998	714.11	713.1	112	668.9 169	13 29	$F_3C(CF_2)_{12}COOH$
PFHxDA	Perfluoro-n-hexadecanoic acid	67905-19-5	$C_{16}HF_{31}O_2$	15.041	814.13	813.1	121	768.9 168.9	13 37	$F_3C(CF_2)_{14}COOH$
Gen X	Perfluoro-2-propoxypropanoic acid	13252-13-6	$C_6HF_{11}O_3$	5.866	330.05	329.0	156	284.9 169.0	5 13	$F_3C(CF_2)_2OCF(COOH)CF_3$
PFPrS	Perfluoro-1-propanesulfonic acid	423-41-6	$C_3HF_7SO_3$	2.748	250.09	249.1	140	80 98.9	37 33	$F_3C(CF_2)_2SO_3H$
PFBS	Perfluoro-1-butanesulfonic acid	375-73-5	$C_4HF_9SO_3$	3.876	300.10	299.0	149	80 98.9	37 37	$F_3C(CF_2)_3SO_3H$
PFPeS	Perfluoro-1-pentanesulfonic acid	2706-91-4	$C_5HF_{11}SO_3$	5.336	350.11	349.0	175	80 98.9	45 37	$F_3C(CF_2)_4SO_3H$
PFHxS	Perfluoro-1-hexanesulfonic acid	355-46-4	$C_6HF_{13}SO_3$	6.885	400.11	399.0	179	80 98.9	45 41	$F_3C(CF_2)_5SO_3H$
PFHpS			$C_7HF_{15}SO_3$	8.357	450.12	449.0	183	80	49	$F_3C(CF_2)_6SO_3H$

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
	Perfluoro-1-heptanesulfonic acid	375-92-8						98.9	45	
PFOS	Perfluoro-1-octanesulfonic acid	1763-23-1	$C_8HF_{17}SO_3$	9.647	500.13	499.0	208	80 98.9	101 49	$F_3C(CF_2)_7SO_3H$
PFNS	Perfluoro-1-nonanesulfonic acid	68259-12-1	$C_9HF_{19}SO_3$	10.776	549.93	549.0	218	80 98.9	105 49	$F_3C(CF_2)_8SO_3H$
PFDS	Perfluoro-1-decanesulfonic acid	335-77-3	$C_{10}HF_{21}SO_3$	11.764	600.14	598.9	232	80 98.9	137 53	$F_3C(CF_2)_9SO_3H$
PFECHS	Perfluoro-4-ethylcyclohexanesulfonic acid	646-83-3	$C_8HF_{15}SO_3$	8.096	462.13	461.0	150	380.9 98.9	29 29	$F_5C_2(C_6F_{10})$ (*para*-) SO_3H
Cl-PFOS	8-Chloroperfluoro-1-octanesulfonic acid	777011-38-8	$C_8HF_{16}ClSO_3$	9.897	516.58	515.0	203	80 98.9	105 49	$ClF_2C(CF_2)_7SO_3H$
6:2 Cl-PFESA	9-Chlorohexadecafluoro-3-oxanonane-1-sulfonic acid	756426-58-1	$C_8HClF_{16}O_4S$	9.379	532.58	530.9	161	350.9 83.0	29 29	$ClF_2C(CF_2)_5O(CF_2)_2SO_3H$
8:2 Cl-PFESA	11-Chloroeicosafluoro-3-oxaundecane-1-sulfonic acid	763051-92-9	$C_{10}HClF_{20}O_4S$	10.812	632.60	630.9	171	450.9 83.0	33 33	$ClF_2C(CF_2)_7O(CF_2)_2SO_3H$
FBSA	Perfluoro-1-butanesulfonamide	30334-69-1	$C_4H_2F_9NO_2S$	5.002	299.12	298.0	98	78.0 48.1	25 93	$F_3C(CF_2)_3SO_2NH_2$
FHxSA	Perfluoro-1-hexanesulfonamide	41997-13-1	$C_6H_2F_{13}NO_2S$	8.226	399.13	397.9	117	78.0 48.1	29 100	$F_3C(CF_2)_5SO_2NH_2$

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
FOSA	Perfluoro-1-octanesulfonamide	754-91-6	$C_8H_2F_{17}NO_2S$	11.159	499.14	498.0	169	78 48.1	37 150	$F_3C(CF_2)_7SO_2NH_2$
MeFOSA	N-methylperfluoro-1-octanesulfonamide	31506-32-8	$C_9H_4F_{17}NO_2S$	12.808	513.17	512.0	160	169 218.9	29 25	$F_3C(CF_2)_7SO_2NHCH_3$
EtFOSA	N-ethylperfluoro-1-octanesulfonamide	4151-50-2	$C_{10}H_6F_{17}NO_2S$	13.375	527.20	526.0	165	169 219	29 29	$F_3C(CF_2)_7SO_2NHC_2H_5$
6:2 FTUCA	2H-Perfluoro-2-octenoic acid (6:2)	70887-88-6	$C_8H_2F_{12}O_2$	7.712	358.08	357	69	293 92.9	13 49	$F_3C(CF_2)_4FC=CHCOOH$
5:3 FTCA	5:3 Fluorotelomer carboxylic acid	914637-49-3	$C_8H_5F_{11}O_2$	7.597	342.10	341	108	237 216.9	13 25	$F_3C(CF_2)_4(CH_2)_2COOH$
6:2 FTCA	2-Perfluorohexyl ethanoic acid (6:2)	53826-12-3	$C_8H_3F_{13}O_2$	7.037	378.09	377.0	185	292.9 63.1	15 3	$F_3C(CF_2)_5CH_2COOH$
8:2 FTCA	2-Perfluorooctyl ethanoic acid (8:2)	27854-31-5	$C_{10}H_3F_{17}O_2$	9.927	478.10	477.0	215	392.9 63	15 3	$F_3C(CF_2)_7CH_2COOH$
10:2 FTCA	2-Perfluorodecyl ethanoic acid (10:2)	53826-13-4	$C_{12}H_3F_{21}O_2$	12.075	578.12	577.0	245	492.9 63	15 3	$F_3C(CF_2)_9CH_2COOH$
4:2 FTSA	1H,1H,2H,2H-perfluorohexane sulfonic acid (4:2)	757124-72-4	$C_6H_5F_9O_3S$	4.870	328.15	327.1	136	306.9 81	21 33	$F_3C(CF_2)_3(CH_2)_2SO_3H$
6:2 FTSA	1H,1H,2H,2H-perfluorooctane sulfonic acid (6:2)	27619-97-2	$C_8H_5F_{13}O_3S$	8.091	428.17	427.0	164	406.9 81	25 41	$F_3C(CF_2)_5(CH_2)_2SO_3H$
8:2 FTSA			$C_{10}H_5F_{17}O_3S$	10.676	528.18	527.0	179	506.9	29	$F_3C(CF_2)_7(CH_2)_2SO_3H$

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
	1H,1H,2H,2H-perfluorodecane sulfonic acid (8:2)	39108-34-4						81	41	
6:2 PAP	1H,1H,2H,2H-perfluorooctylphosphate	57678-01-0	$C_8H_6F_{13}O_4P$	3.513	444.08	443.0	108	97.0 79.0	17 100	$(O)P(OH)_2[O(CH_2)_2(CF_2)_5CF_3]$
8:2 PAP	1H,1H,2H,2H-perfluorodecylphosphate	57678-03-2	$C_{10}H_6F_{17}O_4P$	4.306	544.08	543.0	108	97.0 79.0	21 93	$(O)P(OH)_2[O(CH_2)_2(CF_2)_7CF_3]$
6:2 diPAP	Bis(1H,1H,2H,2H-perfluorooctyl)phosphate	57677-95-9	$C_{16}H_9F_{26}O_4P$	5.289	790.17	789.0	132	442.9 97.0	17 37	$(O)P(OH)[O(CH_2)_2(CF_2)_5CF_3]_2$
6:2/8:2 diPAP	(1H,1H,2H,2H-perfluorooctyl-1H,1H,2H,2H-perfluorodecyl)phosphate	943913-15-3	$C_{18}H_9F_{30}O_4P$	5.478	890.20	889.0	156	443.0 96.9	21 33	$(O)P(OH)[O(CH_2)_2(CF_2)_5CF_3][O(CH_2)_2(CF_2)_7CF_3]$
8:2 diPAP	Bis(1H,1H,2H,2H-perfluorodecyl)phosphate	678-41-1	$C_{20}H_9F_{34}O_4P$	5.622	990.20	989.0	151	542.9 97.0	25 37	$(O)P(OH)[O(CH_2)_2(CF_2)_7CF_3]_2$
M3PFBA (surrogate standard, SS)	Perfluoro-n-[2,3,4-$^{13}C_3$]butanoic acid		CHF_7O_2 + $^{13}C_3$	2.242	217.04	216.0	64	172	5	
MPFHxA (SS)	Perfluoro-n-[1,2-$^{13}C_2$]hexanoic acid		$C_4HF_{11}O_2$ + $^{13}C_2$	4.999	316.05	315.1	78	270	5	
MPFOA (SS)	Perfluoro-n-[1,2,3,4-$^{13}C_4$]octanoic acid		$C_4HF_{15}O_2$ + $^{13}C_4$	8.185	418.07	417.1	83	372	5	
MPFUnDA (SS)	Perfluoro-n-[1,2-$^{13}C_2$]undecanoic acid		$C_9HF_{21}O_2$ + $^{13}C_2$	11.725	566.09	565.1	97	520	9	
M2PFTeDA (SS)	Perfluoro-n-[1,2-$^{13}C_2$]tetradecanoic acid		$C_{12}HF_{27}O_2$ + $^{13}C_2$	13.997	716.11	715.1	116	669.9	13	
M3PFBS (SS)	Perfluoro-1-[2,3,4-$^{13}C_3$]butanesulfonic acid		CHF_9SO_3 + $^{13}C_3$	3.874	303.10	302.0	149	80	45	
MPFHxS (SS)	Perfluoro-1-hexane[$^{18}O_2$]sulfonic acid		$C_6HF_{13}SO$ + $^{18}O_2$	6.882	404.11	403.0	169	84	49	

Abbr.	Compound Name	CAS #	Formula	Retention time (min)	Mol. Wt.	Precursor ion [M-H/D]-	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)	Structure
MPFOS (SS)	Perfluoro-1-[1,2,3,4-$^{13}C_4$]octanesulfonic acid		$C_4HF_{17}SO_3$ + $^{13}C_4$	9.646	504.13	503.0	198	80	93	
dMeFOSA (SS)	N-methyl-d_3-perfluoro-1-octanesulfonamide		$C_9HF_{17}NO_2S$ + D_3	12.799	516.17	515.0	160	169	29	
M2-8:2 FTCA (SS)	2-Perfluorooctyl-[1,2-$^{13}C_2$]-ethanoic acid(8:2)		$C_8H_3F_{17}O_2$ + $^{13}C_2$	9.926	480.10	479.0	215	394	11	
M2-8:2 FTSA (SS)	1H,1H,2H,2H-perfluoro-1-[1,2-$^{13}C_2$]-decane sulfonic acid (8:2)		$C_8H_5F_{17}O_3S$ + $^{13}C_2$	10.675	530.18	529.0	195	509	33	
M2-8:2 PAP (SS)	1H,1H,2H,2H-[1,2-$^{13}C_2$]perfluorodecylphosphate		$C_8H_6F_{17}O_4P$ + $^{13}C_2$	4.305	546.08	545.0	113	97.0	17	
MPFBA (internal standard, IS)	Perfluoro-n-[1,2,3,4-$^{13}C_4$]butanoic acid		HF_7O_2 + $^{13}C_4$	2.240	218.04	217.0	64	172	5	
M8PFOA (IS)	Perfluoro-n-[$^{13}C_8$]octanoic acid		$HF_{15}O_2$ + $^{13}C_8$	8.184	422.07	421.1	83	376	5	
M7PFUnDA (IS)	Perfluoro-n-[1,2,3,4,5,6,7-$^{13}C_7$]undecanoic acid		$C_4HF_{21}O_2$ + $^{13}C_7$	11.724	571.09	570.0	97	525	9	
M3PFHxS (IS)	Perfluoro-1-[1,2,3-$^{13}C_3$]hexanesulfonic acid		$C_3HF_{13}SO_3$ + $^{13}C_3$	6.883	403.11	402.0	184	80	45	
M8PFOS (IS)	Perfluoro-[$^{13}C_8$]octanesulfonic acid		$HF_{17}SO_3$ + $^{13}C_8$	9.637	508.13	507.0	203	79.9	97	
M4-6:2 diPAP (IS)	Bis(1H,1H,2H,2H-[1,2-$^{13}C_2$]perfluorooctyl)phosphate		$C_{12}H_9F_{26}O_4P$ + $^{13}C_4$	5.288	794.17	793.0	137	445.0	21	

Table S6. List of targeted PFAS measured by GC-MS.

Abbr.	Compound Name	CAS #	Formula	Mol. Wt.	Retention time (min)	Quantifier	Qualifier	Structure
4:2 FTOH	2-Perfluorobutyl ethanol (4:2)	2043-47-2	$C_6H_5F_9O$	264.09	5.840	265	227	$F_3C(CF_2)_3(CH_2)_2OH$
6:2 FTOH	2-Perfluorohexyl ethanol (6:2)	647-42-7	$C_8H_5F_{13}O$	364.10	7.569	365	327	$F_3C(CF_2)_5(CH_2)_2OH$
8:2 FTOH	2-Perfluorooctyl ethanol (8:2)	678-39-7	$C_{10}H_5F_{17}O$	464.12	9.993	465	427	$F_3C(CF_2)_7(CH_2)_2OH$
10:2 FTOH	2-Perfluorodecyl ethanol (10:2)	865-86-1	$C_{12}H_5F_{21}O$	564.13	12.460	565	527	$F_3C(CF_2)_9(CH_2)_2OH$
6:2 FTAc	1H,1H,2H,2H-perfluorooctyl acrylate	17527-29-6	$C_{11}H_7F_{13}O_2$	418.15	6.450	419	399	$F_3C(CF_2)_5(CH_2)_2COOCH=CH_2$
8:2 FTAc	1H,1H,2H,2H-Perfluorodecyl acrylate	27905-45-9	$C_{13}H_7F_{17}O_2$	518.17	9.100	519	499	$F_3C(CF_2)_7(CH_2)_2COOCH=CH_2$
10:2FTAc	1H,1H,2H,2H-Perfluorododecyl acrylate	17741-60-5	$C_{15}H_7F_{21}O_2$	618.18	11.916	619	599	$F_3C(CF_2)_9(CH_2)_2COOCH=CH_2$
6:2 FTMAc	1H,1H,2H,2H-perfluorooctyl methacrylate	2144-53-8	$C_{12}H_9F_{13}O_2$	432.18	7.672	433	413	$F_3C(CF_2)_5(CH_2)_2COOC(=CH_2)CH_3$
8:2 FTMAc	1H,1H,2H,2H-heptadecafluorodecyl methacrylate	1996-88-9	$C_{14}H_9F_{17}O_2$	532.19	10.413	533	513	$F_3C(CF_2)_7(CH_2)_2COOC(=CH_2)CH_3$
MeFOSE	2-(N-methylperfluoro-1-octanesulfonamido)-ethanol	24448-09-7	$C_{11}H_8F_{17}NO_3S$	557.22	19.068	558	540	$F_3C(CF_2)_7SO_2N(-CH_3)(CH_2)_2OH$
EtFOSE	2-(N-ethylperfluoro-1-octanesulfonamido)-ethanol	1691-99-2	$C_{12}H_{10}F_{17}NO_3$	571.25	19.194	572	554	$F_3C(CF_2)_7SO_2N(-C_2H_5)(CH_2)_2OH$
M4-4:2 FTOH (SS)	2-Perfluorobutyl-[1,1,2,2-$^2H_4$]-ethanol(4:2)		$C_6HF_9O + D_4$	268.09	5.776	269	230	
M2-8:2 FTOH (SS)	2-Perfluorooctyl-[1,2-$^{13}C_2$]-ethanol(8:2)		$C_8H_5F_{17}O + {}^{13}C_2$	466.12	9.985	467	429	
dMeFOSE (SS)	2-(N-methyl-d_3-perfluoro-1-octanesulfonamido)ethan-d_4-ol		$C_{11}H_1F_{17}NO_3S + D_7$	564.22	19.028	565	547	
M4-8:2 FTOH (IS)	2-Perfluorooctyl-[1,1-$^2H_2$]-[1,2-$^{13}C_2$]-ethanol(8:2)		$C_8H_3F_{17}O + {}^{13}C_2D_2$	468.12	9.946	469	431	

Table S7: Surrogate (SS) and internal (IS) standards used to calculate PFAS concentrations.

Native PFAS	SS for correction	IS for quantitation	Surrogate standards	IS for quantitation
PFPrA	M3PFBA	MPFBA	M3PFBA	MPFBA
PFBA	M3PFBA	MPFBA	MPFHxA	M8PFOA
PFPeA	M3PFBA	MPFBA	MPFOA	M8PFOA
PFHxA	MPFHxA	M8PFOA	MPFUnDA	M7PFUnDA
PFHpA	MPFHxA	M8PFOA	M2PFTeDA	M7PFUnDA
PFOA	MPFOA	M8PFOA	M3PFBS	M3PFHxS
PFNA	MPFOA	M8PFOA	MPFHxS	M3PFHxS
PFDA	MPFUnDA	M7PFUnDA	MPFOS	M8PFOS
PFUnDA	MPFUnDA	M7PFUnDA	M2-8:2 FTCA	M8PFOA
PFDoDA	M2PFTeDA	M7PFUnDA	M2-8:2 FTSA	M8PFOS
PFTrDA	M2PFTeDA	M7PFUnDA	dMeFOSA	M8PFOS
PFTeDA	M2PFTeDA	M7PFUnDA	M4-4:2 FTOH	M4-8:2 FTOH
PFHxDA	M2PFTeDA	M7PFUnDA	M2-8:2 FTOH	M4-8:2 FTOH
PFPrS	M3PFBS	M3PFHxS	dMeFOSE	M4-8:2 FTOH
PFBS	M3PFBS	M3PFHxS	M2-8:2 PAP	M4-6:2 diPAP
PFPeS	MPFHxS	M3PFHxS		
PFHxS	MPFHxS	M3PFHxS		
PFHpS	MPFOS	M3PFHxS		
PFOS	MPFOS	M8PFOS		
PFNS	MPFOS	M8PFOS		
PFDS	MPFOS	M8PFOS		
PFECHS	MPFOS	M8PFOS		
Cl-PFOS	MPFOS	M8PFOS		
6:2 Cl-PFESA	MPFHxS	M8PFOS		
8:2 Cl-PFESA	MPFOS	M8PFOS		
4:2 FTSA	M2-8:2 FTSA	M3PFHxS		
6:2 FTSA	M2-8:2 FTSA	M3PFHxS		
8:2 FTSA	M2-8:2 FTSA	M8PFOS		
6:2 FTUCA	M2-8:2 FTCA	M8PFOA		
5:3 FTCA	M2-8:2 FTCA	M8PFOA		
6:2 FTCA	M2-8:2 FTCA	M8PFOA		
8:2 FTCA	M2-8:2 FTCA	M8PFOA		
10:2 FTCA	M2-8:2 FTCA	M8PFOA		
FBSA	M3PFBS	M3PFHxS		
FHxSA	MPFHxS	M3PFHxS		

FOSA	MPFOS	M8PFOS		
MeFOSA	dMeFOSA	M8PFOS		
EtFOSA	dMeFOSA	M8PFOS		
4:2 FTOH	M4-4:2 FTOH	M4-8:2 FTOH		
6:2 FTOH	M2-8:2 FTOH	M4-8:2 FTOH		
8:2 FTOH	M2-8:2 FTOH	M4-8:2 FTOH		
10:2 FTOH	M2-8:2 FTOH	M4-8:2 FTOH		
MeFOSE	dMeFOSE	M4-8:2 FTOH		
EtFOSE	dMeFOSE	M4-8:2 FTOH		
6:2 FTAc	M4-4:2 FTOH	M4-8:2 FTOH		
8:2 FTAc	M2-8:2 FTOH	M4-8:2 FTOH		
10:2 FTAc	M2-8:2 FTOH	M4-8:2 FTOH		
6:2 FTMAc	M2-8:2 FTOH	M4-8:2 FTOH		
8:2 FTMAc	M2-8:2 FTOH	M4-8:2 FTOH		
6:2 PAP	M2-8:2 PAP	M4-6:2 diPAP		
8:2 PAP	M2-8:2 PAP	M4-6:2 diPAP		
6:2 diPAP	M2-8:2 PAP	M4-6:2 diPAP		
6:2/8:2 diPAP	M2-8:2 PAP	M4-6:2 diPAP		
8:2 diPAP	M2-8:2 PAP	M4-6:2 diPAP		

Table S8: Replicate values for the targeted analysis of a field duplicate sample. Only compounds measured above the LOD are reported.

Sample Type	**Description**	**PFPrA**	**PFBA**	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**	**PFTeDA**	**PFHxDA**	**6:2 diPAP**	**∑ PFAS**
Popcorn	Paper bag, plastic lining	3.93	3.29	3.92	4.35	4.01	4.91	7.46	6.73	3.90	4.72	5.07	2.01	1.05	55.33
		3.98	4.65	4.14	4.09	4.79	4.56	6.77	7.38	4.33	4.28	4.66	2.12	1.23	56.97
% Difference		1	29	5	6	16	8	10	9	10	10	9	5	15	3

Table S9: Summary of duplicate samples measured by PIGE spectroscopy.

Sample Type	Description	Duplicate Type	Total F (µg F/m^2)	% Difference
Burrito roll	Paper wrapper	Field	< LOD	N/A
			< LOD	
Popcorn bag 8	Paper bag, plastic lining	Field	< LOD	N/A
			< LOD	
Pita wrap	Paper wrapper	Field	< LOD	N/A
			< LOD	
Burrito roll	Foil wrapper	Field	< LOD	N/A
			< LOD	
Pastry	Paper bag	Lab	< LOD	N/A
			< LOD	
Pita wrap	Paper wrapper	Lab	< LOD	N/A
			< LOD	
Donut bag	Paper bag	Lab	11600	1
			11700	
Donut bag 4	Paper bag	Lab	36000	3
			37200	
Burrito roll	Paper wrapper	Lab	< LOD	N/A
			< LOD	
Popcorn bag 7	Paper bag	Sample Period	<LOQ	N/A
			11400	
Donut bag	Paper bag	Sample Period	11700	33
			17600	
Pastry bag 5	Paper bag	Sample Period	21400	10
			23800	
Pastry bag 6	Paper bag	Sample Period	26700	5
			28000	
Donut bag 4	Paper bag	Sample Period	37200	19
			30100	
Burrito bowl 2	Molded fiber bowl	Sample Period	931000	8
			1010000	
Burrito bowl 1	Molded fiber bowl	Sample Period	1300000	4
			1240000	
Salad bowl 3			931000	21

	Molded fiber bowl	Sample Period	1180000	

Table S10. Summary of total F in all food packaging samples, determined by PIGE. The number of samples does not include duplicates.

Category	**Level of total F (μg/m^2)**	**Number of Food Packaging Samples**	**Percentage of Food Packaging Samples**
None	<3580 (165-3540)	23	55%
Low	>3580 – <10800 (4600-9600)	8	19%
High	>10800 (11400-1300000)	11	26%
	TOTAL	42	100%

For bowls (thickness > 620 μm): LOD = 20600 μg F/m^2, LOQ=62500 μg F/m^2; for paper bags and paper wrapper (thickness ⩽180): LOD = 3580 μg F/m^2, LOQ=10800 μg F/m^2. For all four bowls, the total F ranged from 1010000 to 1300000 μg F/m^2, which was higher than 62500 μg F/m^2 (LOQ) and much higher than 10800 μg F/m^2 (LOQ for paper bag and paper wrapper). So we use 3580 μg F/m^2 (LOD) and 10800 μg F/m^2 (LOQ) for paper bag and paper wrapper to divide them into three categories, None, Low, and High (see table S10).

Table S11. List of fast food packaging with total fluorine > LOQ determined by PIGE (see Table S10 for list of LOQ values). Only the maximum value from each sample is listed.

Item Name (Food)	**Item Size**	**Category (material)**	**Description (colour, material, location)**	**Total F (µg F/m^2)**
Burrito bowl 1	Regular	Molded fiber bowl	Brown molded fiber, exterior	1300000
Burrito bowl 2	Small	Molded fiber bowl	Brown molded fiber, interior	1010000
Salad bowl 3	Regular	Molded fiber bowl	Brown molded fiber, interior	1180000
Salad bowl	Regular	Molded fiber bowl	Brown molded fiber, exterior	1070000
Donut bag 4	Regular	Paper bag	White paper, interior	30100
Pastry bag 5	Regular	Paper bag	Brown paper, interior	23800
Pastry bag 6	Regular	Paper bag	Brown paper, interior	28000
Popcorn bag 7	Very small (grab)	Paper bag	White paper, interior	11400
Donut	Regular	Paper bag	Brown paper, interior	17600
Donut bag	Regular	Paper bag	White paper, interior	17600
Burger	Small (buddy)	Paper bag	White paper, interior	12100

Table S12a. Full results for all samples selected for targeted analysis using LC-MS/MS and GC-MS. Concentrations of each analyte are given in ng/g (ppb). Cells containing values <LOD are left empty. Only compounds detected in any sample above the LOD are listed. Since popcorn bag 8 had a field duplicate, the numbers showed in both tables (S12a &b) are average value.

	Compound Concentration (ng/g)																							
Category	**PFPrA**	**PFBA**	**PFPeA**	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**	**PFTeDA**	**PFHxDA**	**PFBS**	**6:2 FTSA**	**6:2 PAP**	**8:2 PAP**	**6:2 diPAP**	**6:2 FTOH**	**8:2 FTOH**	**6:2 FTAc**	**6:2 FTMAc**	**6:2 FTUCA**	**5:3 FTCA**	**∑PFAS**
Burrito bowl 1	1.28	0.75	0.84	1.45	0.15											58.0	27.2		294			598	4.94		987
Burrito bowl 2	1.54	1.23		8.50	1.59	0.06		0.19		0.06					0.20	11.9	10.3		885	8.00	28.0	681	1.70	0.01	1639
Salad bowl 3	0.66			4.12	0.24									0.04					486	9.82		430	1.41	0.03	932
Donut paper bag 4		11.3		0.65															1483		17.5	5668			7181
Pastry bag 5		10.5		4.00	0.65														1126	79.9		3407			4628
Pastry bag 6		16.5																	1160		30.2	4672			5879
Popcorn bag 7				2.39	1.43				1.57	1.28	2.74	2.53							1735		232	4575			6554
Popcorn bag 8 (paper with plastic lining)	3.95	3.97		4.03	4.22	4.40	4.73	7.11	7.06	4.12	4.50	4.86	2.06					1.14							56.2

b. Re-analyzed results for all samples selected for targeted analysis using LC-MS/MS and GC-MS after being in storage for ~2 years. Concentrations of each analyte are given in ng/g (ppb). Cells containing values <LOD are left empty. Only compounds detected in any sample above the LOD are listed.

	Compound Concentration (ng/g)																							
Category	**PFPrA**	**PFBA**	**PFPeA**	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUnDA**	**PFDoDA**	**PFTrDA**	**PFTeDA**	**PFHxDA**	**PFBS**	**6:2 FTSA**	**6:2 PAP**	**8:2 PAP**	**6:2 diPAP**	**6:2 FTOH**	**8:2 FTOH**	**6:2 FTAc**	**6:2 FTMAc**	**6:2 FTUCA**	**5:3 FTCA**	**∑PFAS**
Burrito bowl 1	9.11	0.68	0.64	4.14	0.40				0.06										995			206	30.7		1247
Burrito bowl 2	5.59	3.19	1.46	15.4	2.60	0.20				0.19		0.11							1252		49.6	173	8.70	0.34	1512
Salad bowl 3	5.18	1.39	0.54	6.61	0.44				0.08										852		2.01	22.9	8.21	0.26	900
Donut paper bag 4				1.12															1022		15.8	49.8			1089
Pastry bag 5	3.01			6.15	1.17														1190		10.2				1211
Pastry bag 6																			957		34.4				991
Popcorn bag 7	7.49		1.28	0.71					1.88	0.77			2.49						2256		164				2434
Popcorn bag 8 (paper with plastic lining)	12.3	6.68	5.11	4.89	4.59	4.97	5.61	7.13	7.20	6.75	6.42	5.08	1.84						51.9						130

Table S13. Summary of 22 PFAS tentatively identified using non-targeted analysis.

Confidence levels were assigned according to the information provided in the 'Fragments' column. The mass spectra of representative classes of PFAS are shown in Figures S6 to S11.

Compound Group	Compound	$[M-H]^-$	m/z	RT (min)	Confidence Level	Fragments	Present in Sample(s)
A, Fluorotelomer Unsaturated Carboxylic Acid	6:2 FTUCA	$C_8HF_{12}O_2$	356.9789	6.15	1	$[M-HF-CO_2-H]^-$, $[M-CF_2-HF-CO_2-H]^-$	1, 2, 3
B, n:3 Fluorotelomer Carboxylic Acids	5:3 FTCA	$C_8H_4F_{11}O_2$	341.0035	6.31	2	$[M-C_2H_4-CO_2-H]^-$, $[M-2HF-H_2-CO_2-H]^-$, $[M-4HF-CO_2-H]^-$	2, 3
	6:3 FTCA	$C_9H_4F_{13}O_2$	391.0007	6.66	2	$[M-C_2H_4-CO_2-H]^-$, $[M-2HF-H_2-CO_2-H]^-$, $[M-3HF-CO_2-H]^-$, $[M-4HF-CO_2-H]^-$, $[M-CF_2-4HF-CO_2-H]^-$	2
C, Multiple H-Substituted-Ether-Substituted-Polyfluoroalkyl (linear) Carboxylic Acid	n=6	$C_9H_4F_{13}O_3$	406.9956	6.55	3	$[M-3HF-CO_2-H]^-$, $[M-2HF-OCH_2-CO_2-H]^-$, $[M-4HF-CO_2-H]^-$	2
D, Double Bond Perfluoroalkyl (linear) Carboxylic Acids	n=3	$C_{11}F_{19}O_2$	524.9600	5.52	3	$[M-CO_2-H]^-$	8
	n=4	$C_{12}F_{21}O_2$	574.9567	5.82	3	$[M-CO_2-H]^-$, $[M-CF_2-2CF-5CF_2-CO_2-H]^-$	8
	n=5	$C_7HF_{12}O_2$	344.9785	5.43	4	No fragments found	8

E, H-substituted Perfluoroalkyl (linear) Carboxylic Acids	n=6	$C_8HF_{14}O_2$	394.9753	5.86	3	$[M-6CF_2-CHF-CO_2-H]^-$	8
	n=7	$C_9HF_{16}O_2$	444.9721	6.21	4	No fragments found	8
	n=8	$C_{10}HF_{18}O_2$	494.9689	6.48	3	$[M-6CF_2-CHF-CO_2-H]^-$	8
	n=9	$C_{11}HF_{20}O_2$	544.9669	6.74	3	$[M-HF-CO_2-H]^-$, $[M-6CF_2-CHF-CO_2-H]^-$	5, 8
	n=10	$C_{12}HF_{22}O_2$	594.9625	6.96	3	$[M-6CF_2-CHF-CO_2-H]^-$	5, 8
	n=11	$C_{13}HF_{24}O_2$	644.9593	7.13	3	$[M-HF-CO_2-H]^-$, $[M-6CF_2-CHF-CO_2-H]^-$	8
	n=12	$C_{14}HF_{26}O_2$	694.9561	7.28	3	$[M-HF-CO_2-H]^-$, $[M-6CF_2-CHF-CO_2-H]^-$	8
	n=13	$C_{15}HF_{28}O_2$	744.9529	7.41	3	$[M-HF-CO_2-H]^-$, $[M-6CF_2-CHF-CO_2-H]^-$	8
F, Perfluoroalkyl (linear) Dicarboxylic Acids	n=7	$C_9HF_{14}O_4$	438.9651	3.91	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=8	$C_{10}HF_{16}O_4$	488.9619	4.55	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=9	$C_{11}HF_{18}O_4$	538.9587	5.07	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=10	$C_{12}HF_{20}O_4$	588.9562	5.52	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=11	$C_{13}HF_{22}O_4$	638.9536	5.82	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=12	$C_{14}HF_{24}O_4$	688.9491	6.09	2	$[M-HF-CO_2-H]^-$, $[M-HF-2CO_2-H]^-$	8
	n=13	$C_{15}HF_{26}O_4$	738.9459	6.30	2	$[M-HF-2CO_2-H]^-$	8

Figures

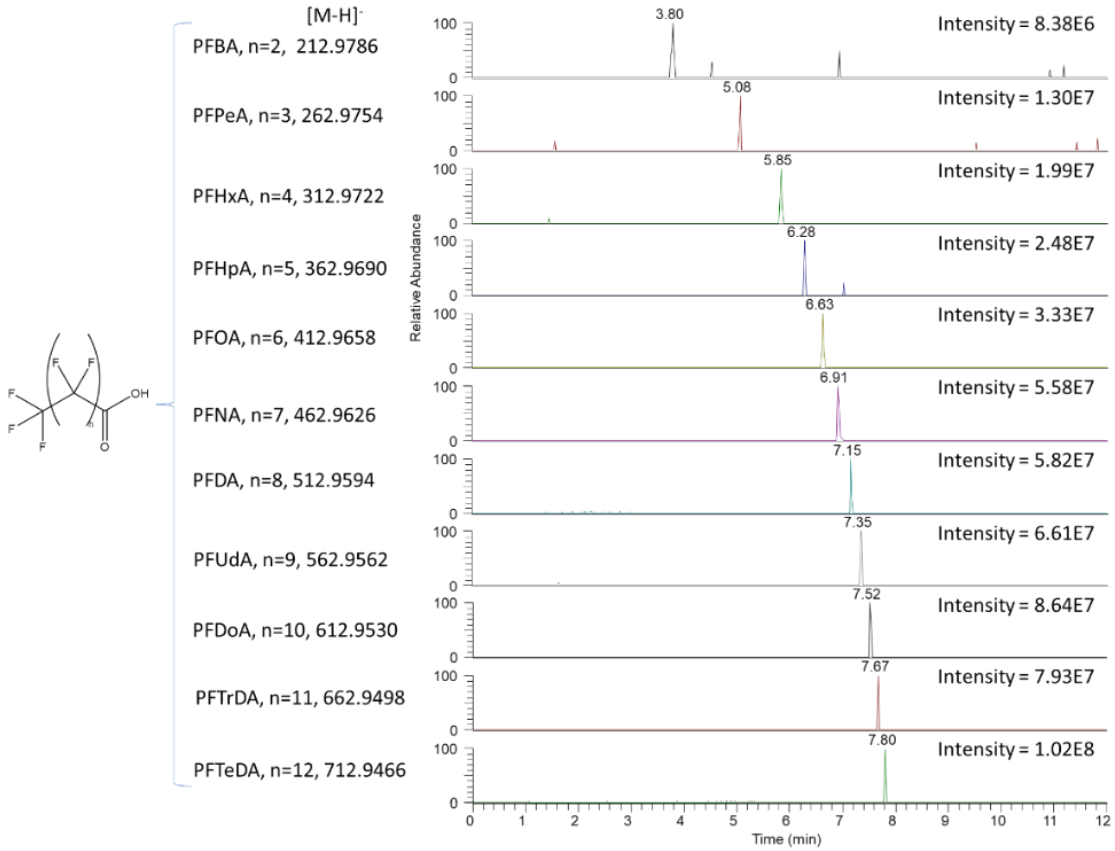


Figure S1. Chromatograms for the $[M-H]^-$ ion of each PFCA standard used for non-targeted analysis QA/QC testing. The retention time is listed above each peak, and the intensity of each compound at a concentration of 100 ppb is listed on the right.

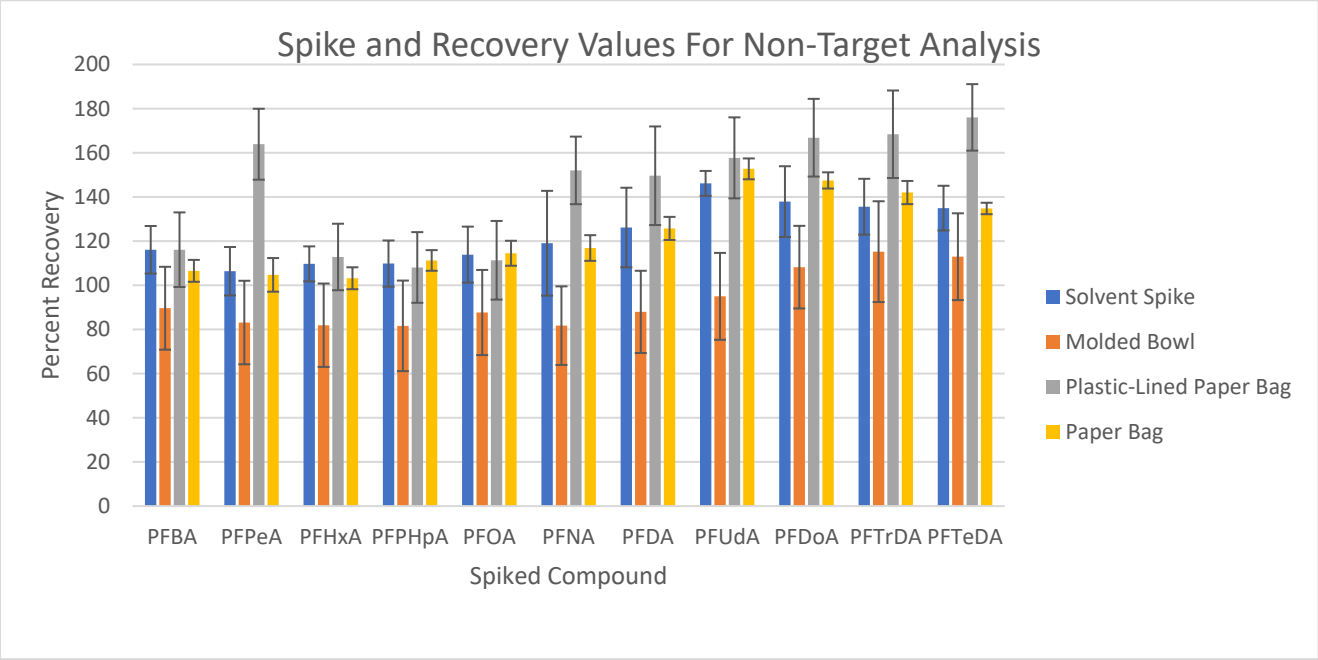


Figure S2. Spike and recovery values for PFCA compounds from PFBA – PFTeDA in three representative sample types. Spiked solvent was used to determine compound loss due to the extraction method. The bars represent averages of three replicates for each compound in each material, and the error bars represent the error-propagated standard deviation for each of the averaged values.

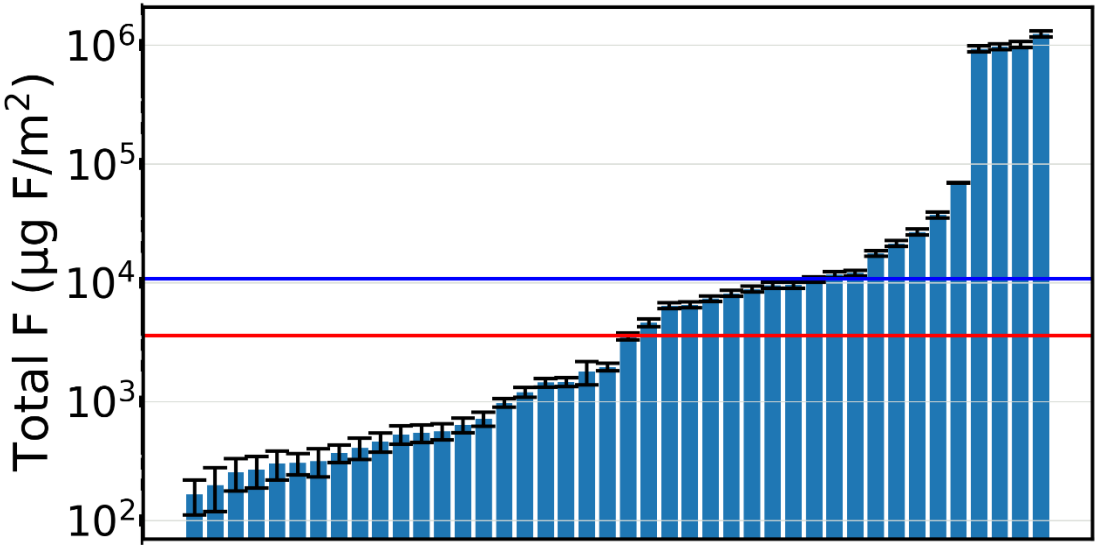


Figure S3. Histogram of screening data showing total F in fast food packaging samples analyzed using PIGE. The highest measured value for each individual sample was used in this analysis. The error bars represent the error of the PIGE measurement, calculated using the calibration. The lower (red) and upper (blue) horizontal lines represent the LOD (3580 µg F/m^2) and LOQ (10800 µg F/m^2) for paper wrapper and paper bag thickness less than 370 µm, respectivly. Note that all samples which are reported below the lower horizontal line (3580 ppm F) were measured below the limit of detection and are included for frequency visualisation only.

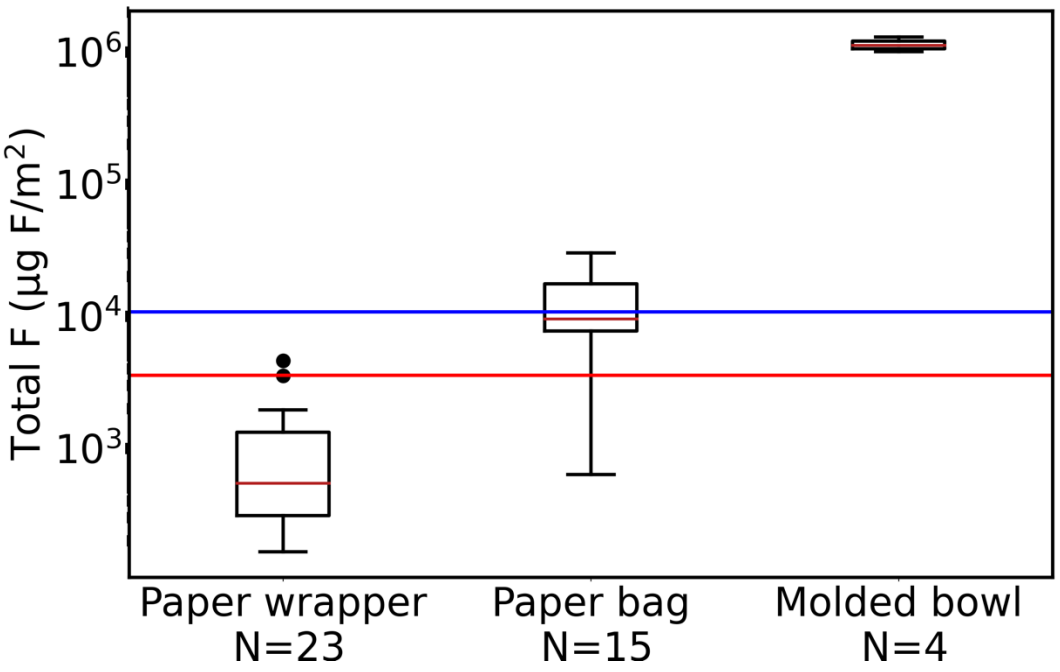
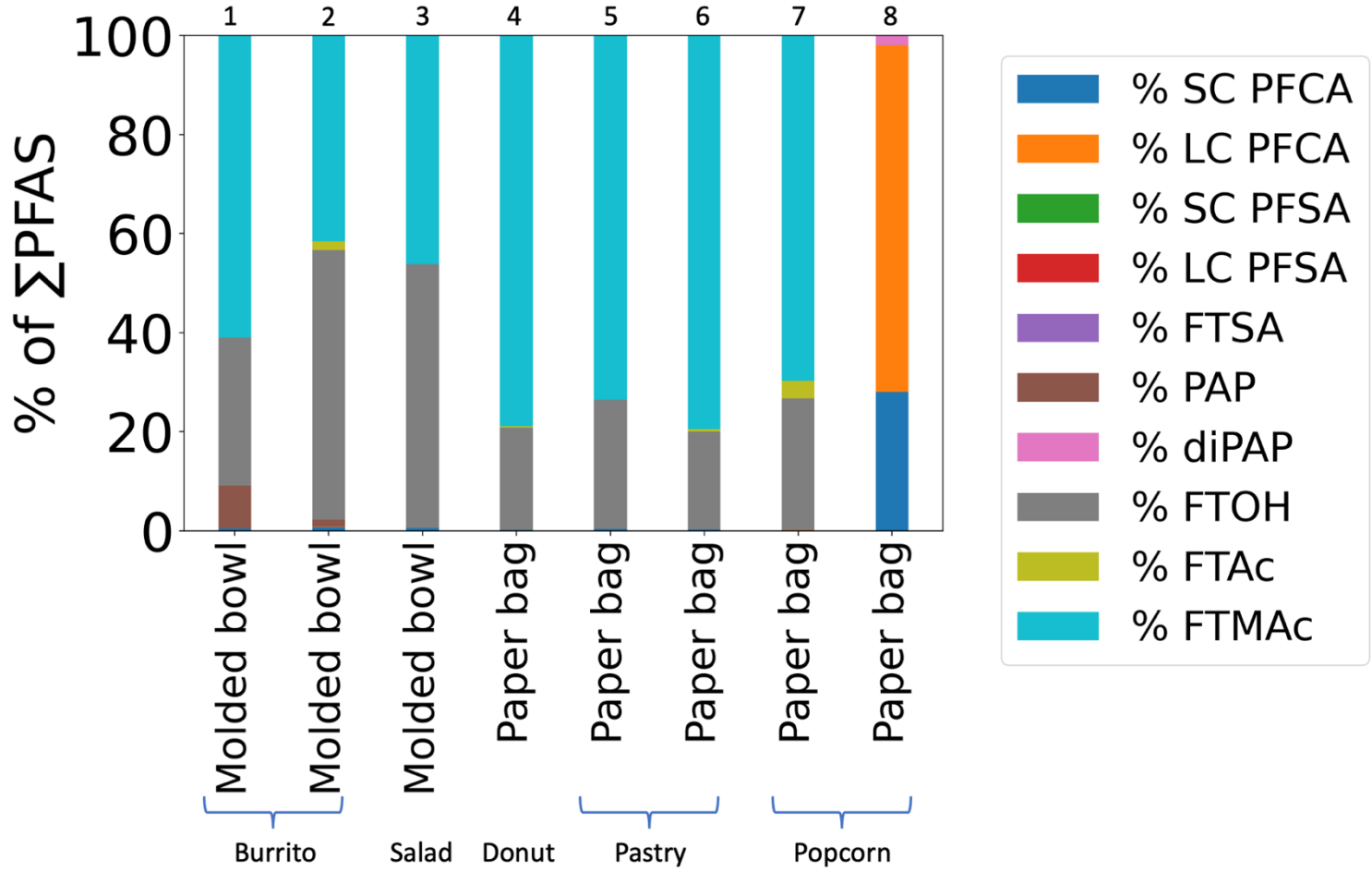


Figure S4. Boxplots showing total F concentration in food packaging. For each sample, the highest measurement was reported. The lower (red) and upper (blue) horizontal lines represent the LOD (3580 µg F/m^2) and LOQ (10800 µg F/m^2) for paper wrapper and paper bag thickness less than 370 µm, respectivly.

Figure S5. Stacked bar chart illustrating the PFAS composition of nine samples selected for targeted analysis. SC and LC refer to short chain and long chain compounds.



Compound	# of Compounds /Class	Compound Class	Sample Type (Intensity)							
			Burrito Bowl 1	Burrito Bowl 2	Salad Bowl 3	Donut Bag 4	Pastry Bag 5	Pastry Bag 6	Popcorn Bag 7	Popcorn Bag 8
6:2 Fluorotelomer Unsaturated Carboxylic Acid	1	a	9.64×10^6	1.66×10^6	1.88×10^6					
n:3 Fluorotelomer Carboxylic Acid, n=6	2	b		6.56×10^4	6.95×10^4					
Multiple H-Substituted-Ether-Substituted-Perfluoroalkyl (linear) Carboxylic Acids, n=6	1	c		1.58×10^4						
Double Bond Perfluoroalkyl (linear) Carboxylic Acids, n=4	2	d								1.43×10^5
H-substituted Perfluoroalkyl (linear) Carboxylic Acids, n=9	9	e					2.45×10^4			1.60×10^5
Perfluoroalkyl (linear) Dicarboxylic Acids, n=11	7	f								3.61×10^5

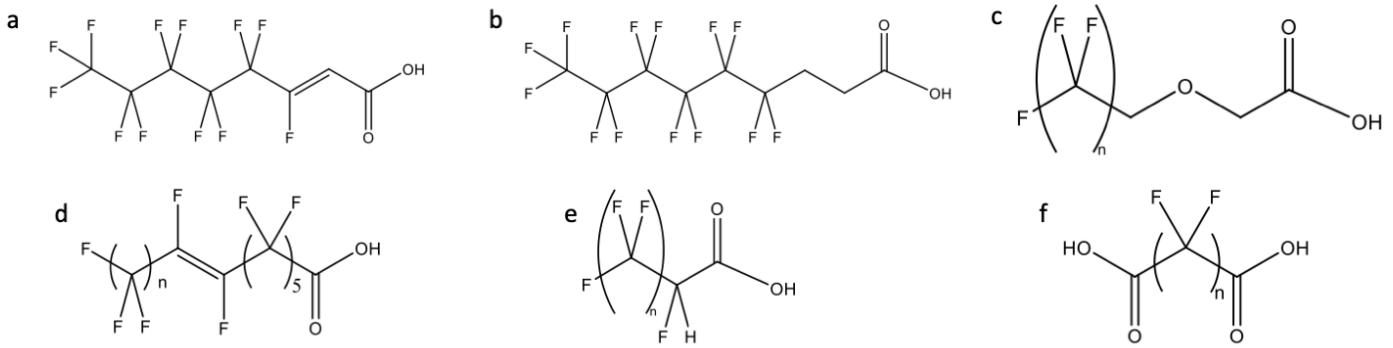


Figure S6: Summary of all compound groups identified using non-targeted analysis. The blank-subtracted intensity of a representative compound from each class is listed in the table and shaded such that a darker color indicates a higher intensity measurement. Structural diagrams for each class are illustrated below the table. For compound groups c and e, it was not possible to locate the exact positions of the H-substitutions, and for group d, it was not possible to locate the exact position of the double bond.

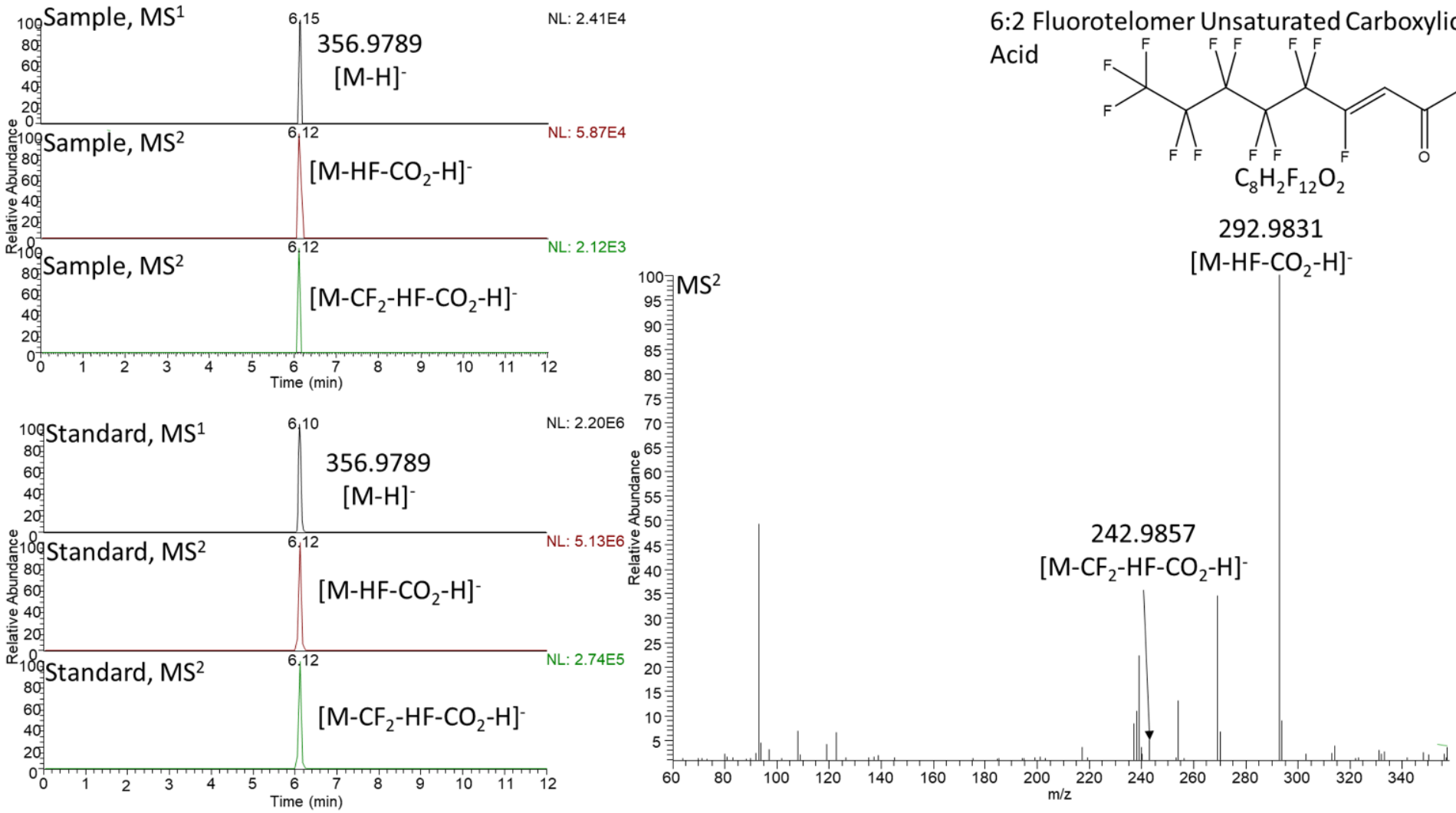


Figure S7. Extracted ion chromatograms and a representative mass spectrum for compound group A, 6:2 FTUCA. The MS^2 fragments of this compound were confirmed by comparing their chromatographic profiles with corresponding precursor ions. The fragments from this compound were compared to those from an authentic standard.

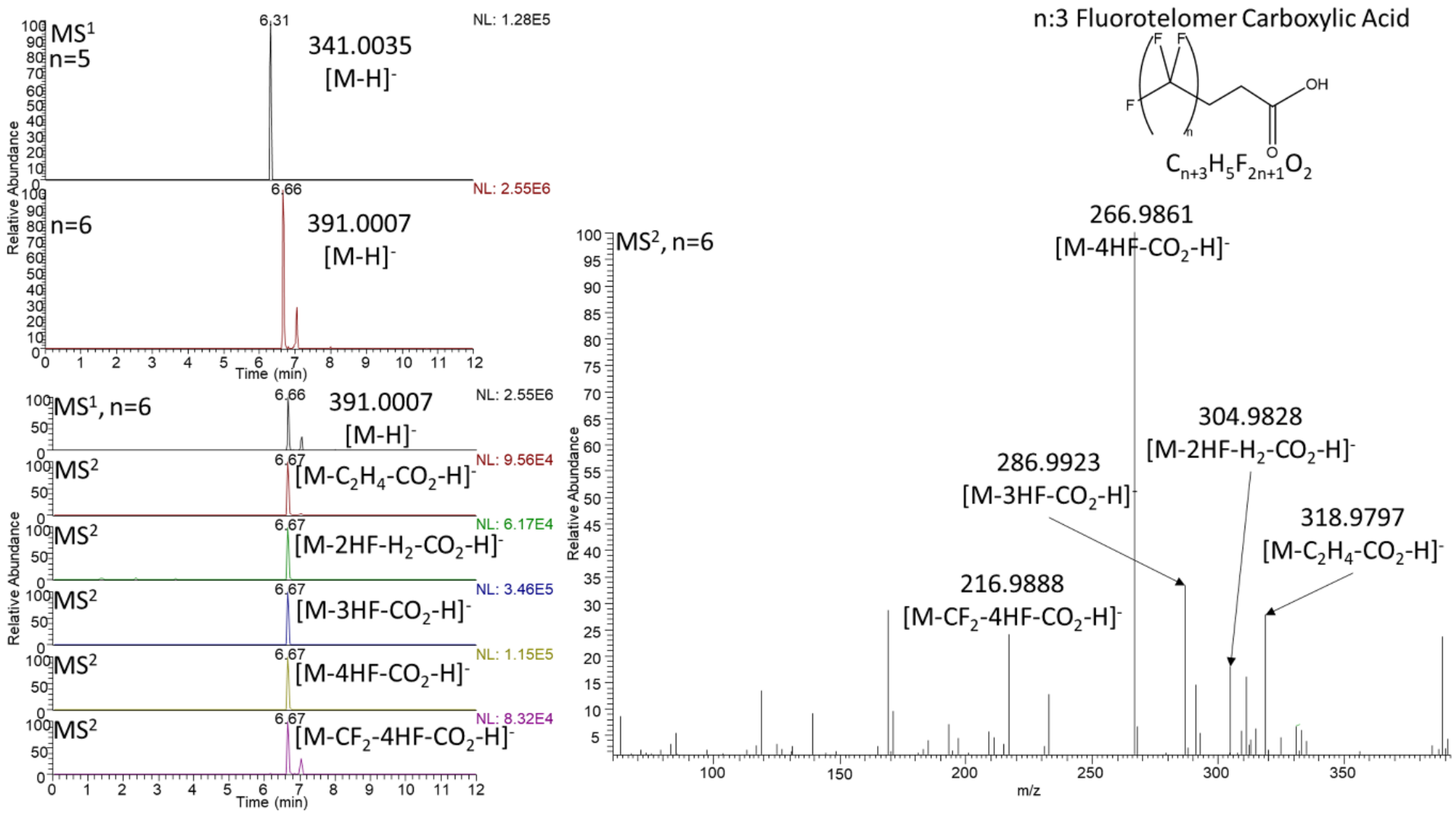


Figure S8. Extracted ion chromatograms and a representative mass spectrum for compound group B, n:3 fluorotelomer carboxylic acids. The fragments from this compound were compared to those reported in previous studies. [9,11,13,26,27]

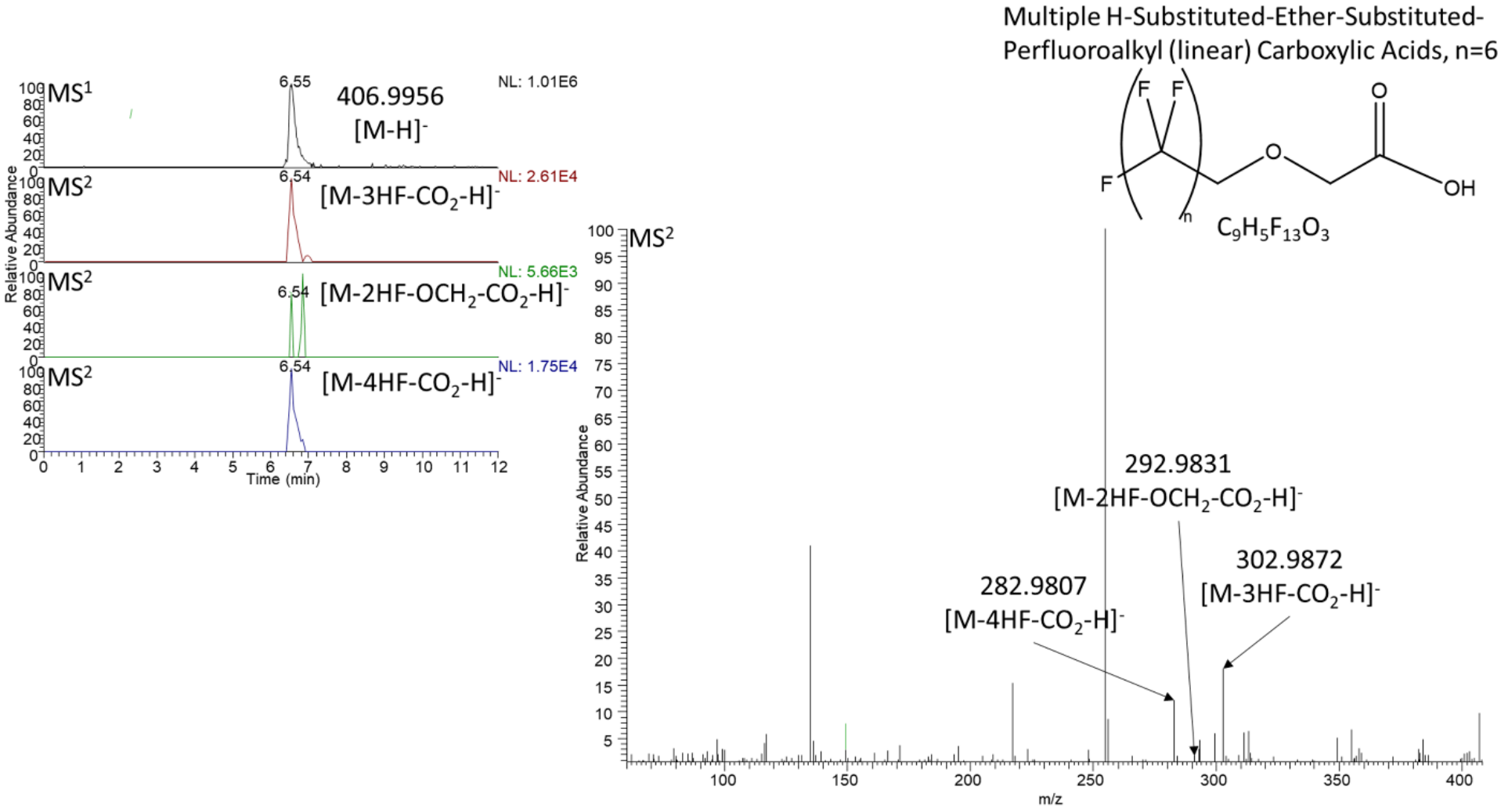


Figure S9. Extracted ion chromatograms and a mass spectrum for compound group C, multiple H-substituted-ether-substituted-polyfluoroalkyl (linear) carboxylic acids. The fragments from this compound were compared to those reported in previous studies.[14]

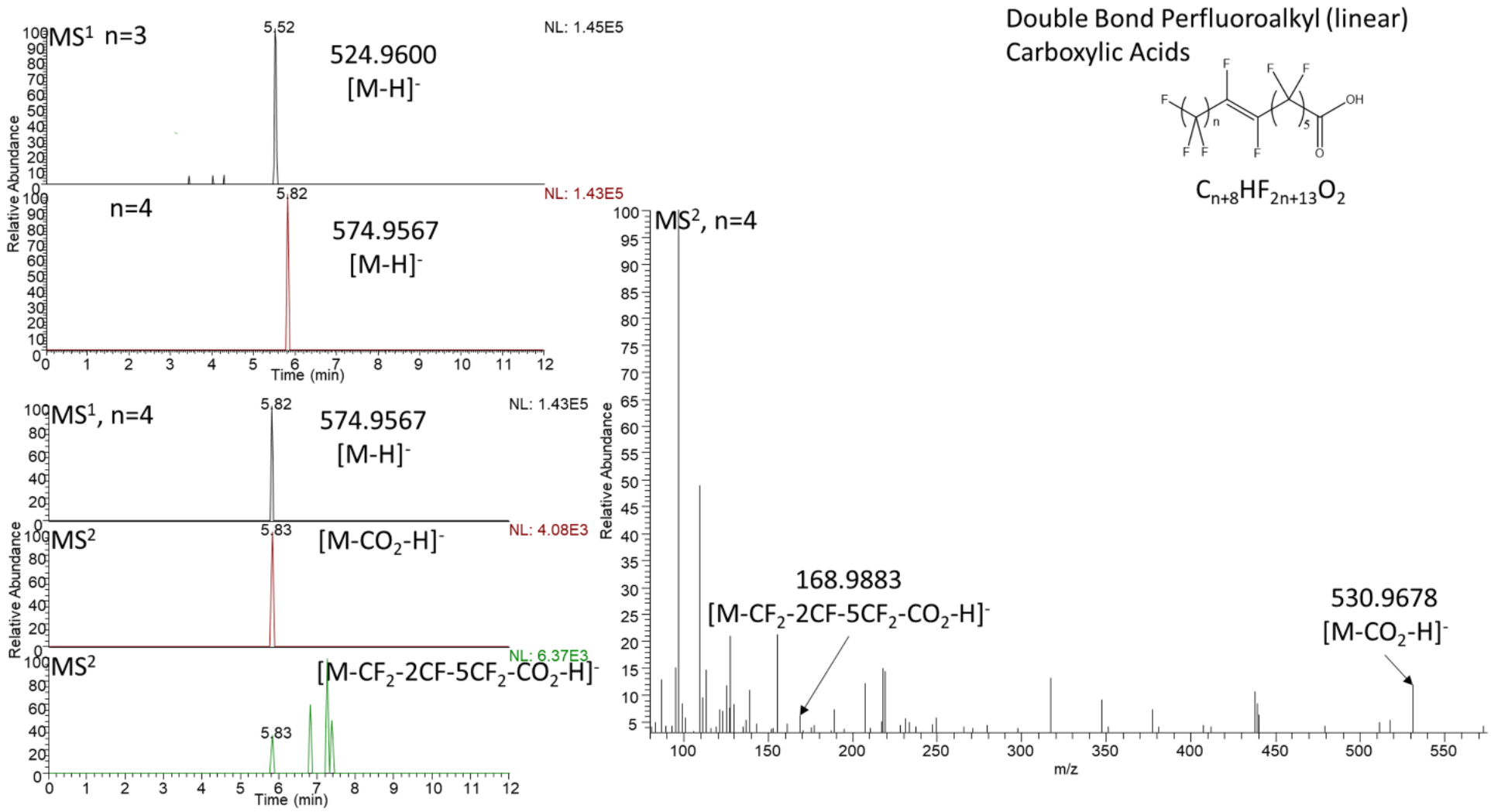


Figure S10. Extracted ion chromatograms and a representative mass spectrum for compound group D, double bond perfluoroalkyl (linear) carboxylic acids. The fragments from this compound were compared to those reported in previous studies.[14,15]

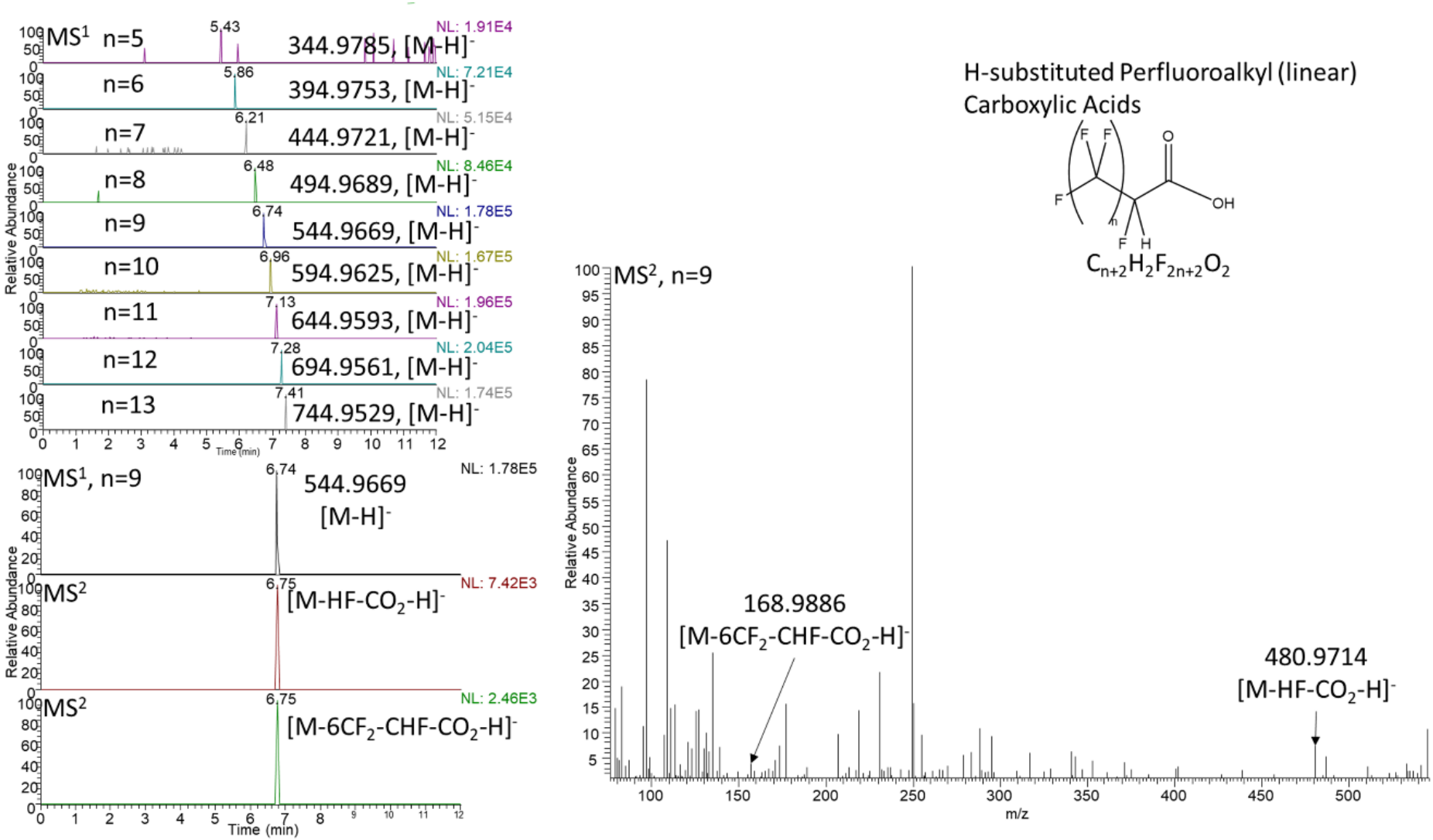


Figure S11. Extracted ion chromatograms and a mass spectrum for compound group E, H-substituted perfluoroalkyl (linear) carboxylic acids. The fragments from this compound were compared to those reported in previous studies.[12,14,16,17]

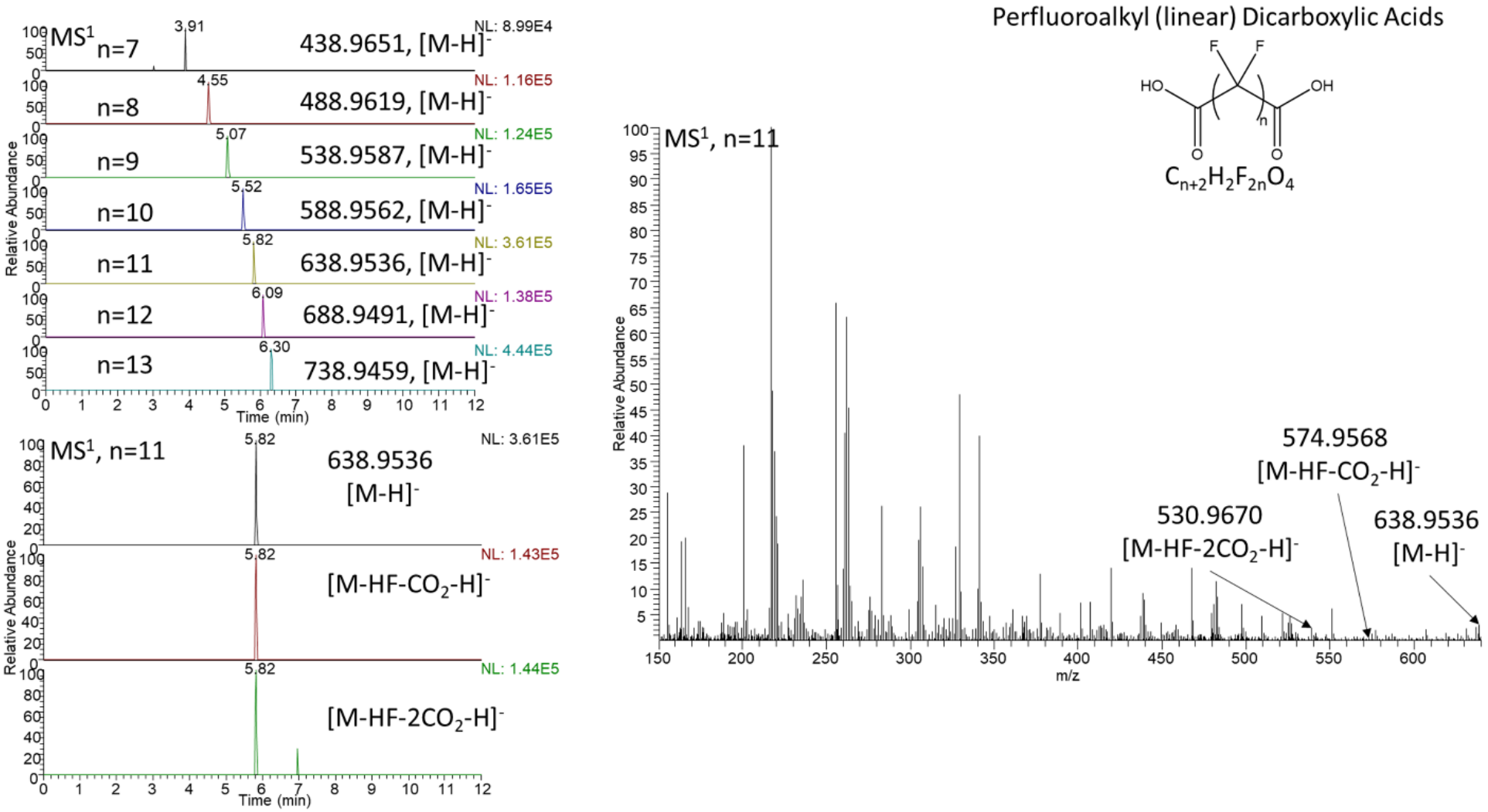


Figure S12. Extracted ion chromatograms and a representative mass spectrum for compound group F, perfluoroalkyl (linear) dicarboxylic acids. The fragments from this compound were compared to those reported in previous studies.[14]

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