Supporting Information for

A North American and Global Survey of Perfluoroalkyl Substances in Surface Soils: Distribution Patterns and Mode of Occurrence

Keegan Rankin[a1], Scott A. Mabury[a], Thomas M. Jenkins[b] and John W. Washington[c2]

[a]Department of Chemistry, University of Toronto,
80 St. George Street, Toronto, Ontario, Canada, M5S 3H6

[b]Senior Environmental Employment Program, United States Environmental Protection Agency,
960 College Station Road, Athens, Georgia, 30605

[c]Ecosystems Research Division, National Exposure Research Laboratory, Office of Research and Development, United States Environmental Protection Agency,
960 College Station Road, Athens, Georgia, 30605

[1]Present address: Xerox Research Centre of Canada, 2660 Speakman Drive, Mississauga, Ontario, L5K 2L1, Canada

[2]To whom correspondence should be addressed: email: washington.john@epa.gov;
phone: 1 (706) 355-8227

Title **Page**

Experimental

Chemicals. Unless noted, all chemicals used in this study were of the highest purity offered by the suppliers, uniformly ≥97% purity. Unlabeled and the twelve mass-labeled perfluorocarboxylic acids and telomer acids, for use as matrix and recovery standards, all were purchased as certified standards from Wellington Laboratories through TerraChem (Shawnee Mission, KS, USA). Tetrabutylammonium hydrogen sulfate (TBAHS) and sodium carbonate were purchased from Aldrich Chemical (Milwaukee, WI, USA). Acetonitrile (ACN), glacial acetic acid, methanol (MeOH) and methyl tert-butyl ether (MTBE) were purchased from Fisher Chemical (Fairlawn, NJ, USA). Oasis HLB solid-phase extraction (SPE) cartridges, 35-cm^3 capacity, were purchased from Waters (Milford, MA, USA). For the ion-pairing agent, a TBAHS mixture (TBA-mix) was prepared by slowly combining two parts 0.25 M Na_2CO_3 solution and one part 0.50 M TBAHS solution by volume to avoid spillage caused by CO_2 generation. The resulting mixture was polished by passage through HLB cartridge to remove PFOA (as detailed below for polished water), which we observed to be present in the TBAHS product as purchased.

Sample-Collection Details. Most sample collectors were known to the authors, but to achieve a better geographic distribution, some were solicited based on their status as authors of published papers and/or university faculty profiles available on the internet. Scientists in geoscience, soils, chemistry, and engineering departments were preferred due to expected training in collecting uncontaminated samples, but presented with special opportunities to choice regions, a medical doctor and a professor of mathematics also agreed to collect samples for us.

Identical sampling kits, which contained instructions and everything needed to sample, were prepared at the EPA/Athens laboratory and were shipped to each location. Sampling kits were prepared entirely with new, unused materials and included: 1) placing nitrile gloves in a zip-lock baggie; 2) placing a methanol-washed stainless-steel trowel in a zip-lock baggie; 3) placing Ottawa sand, purchased from a laboratory supplier, in a polypropylene co-polymer (PPCO) methanol-washed sample container as a field collection blank; 4) adding a second PPCO, methanol-washed sample container as the sample container; 5) return postage and labeling; and 6) sampling and return instructions.

In the instructions, collectors were asked to obtain a surface-soil sample from a nearby location they deemed to have limited recent human impact following the provided written instructions: 1) don nitrile gloves; 2) clear natural, unhumified litter (e.g., undegraded plant matter) from the sampling location; 3) open zip-lock baggie and remove the trowel without touching the blade; 4) open the sample bottle containing the Ottawa sand, pour all the sand onto the trowel blade, pour the sand back into the blank sample bottle and close the sample bottle (collection blank); 5) sample the soil from the surface to about 10-cm depth, placing the soil into the sample bottle, taking care to touch the soil only with the trowel blade and the sample-bottle lip and close the sample bottle; 5) return the sample bottles and trowel to the zip-lock baggie, seal the baggie, and return the baggie to the mailing box; and 6) seal the box and mail back to the Office of Research and Development (ORD) of the United States Environmental Protection Agency (USEPA) in Athens, GA by the return-delivery method organized by the EPA. In all cases, the EPA provided the most expeditious mode of delivery that could be established; this varied in some cases due to availability of courier services and/or export laws of the country of origin.

The sampling instructions did not request the sampler to record the exact sample location because Federal managers determined that such a request would constitute a breach of the

Federal Paperwork Reduction Act of 1980. Despite this, in many instances, the samplers volunteered GPS coordinates, a map or a description of the sampling location. In the remainder, the authors used the best available information to assign the sample location.

Once the sample was received at the laboratory, large particulates were removed from all soil samples using a methanol-washed 2-mm stainless steel sieve. Soil samples then were returned to their original containers and stored at 4 ^{o}C until extraction.

Polished Water. Polished water was achieved by passing 18 MΩ water through an Oasis 35 cc HLB cartridge into a two-liter Erlenmeyer flask that was purchased, methanol washed, then dedicated solely to this use. Once a total of 6 L of the 18 MΩ water was passed through the HLB cartridge it was replaced.

SPE Manifold. Soil extracts were blown to dryness using a solid phase extraction (SPE) manifold as depicted in Figure S7. The samples were placed in the manifold and placed under vacuum. Air was passed through nylon filters and directed into the sample vials with methanol rinsed stainless steel needles. In order to increase the rate of evaporation of the 90:10 acetonitrile(ACN):polished water(PW), a heating pad was fastened to the manifold.

EPA LC-MS/MS Parameters. The LC strong-needle, weak-needle and seal washes were as follows: 60:40 ACN:PW, PW and 10:90 ACN:PW, respectively. A 20 µL aliquot of the extract was injected onto the BEH C18 column and separated with using ACN and PW adjusted to pH 3 with acetic acid as the mobile phases.

University of Toronto LC-MS/MS Parameters. All parameters were the same as above except the injection volume was 5 µL.

Dry-Weight Determination. Extracted soils were left in the PPCO tubes and placed in a vacuum desiccator for several days. The tubes were re-weighed until a constant weight was obtained, which was then used to calculate the dry weight PFCA and PFSA concentrations.

116 **Additional Discussion**

117 **Quality Metrics:** The Ottawa sand field blanks were quantitated for the two dominant
118 analytes we detected in this study, PFOA and PFOS (Table S5-S7). Most sand blanks were
119 found to contain low, and relatively constant concentrations of both analytes, suggesting no
120 sample-collection or –transit contamination. The sand blanks for three samples of remote origin,
121 Buea, Cameroon (AF03), Mabira, Uganda (AF05) and El Yunque, Puerto Rico (NA23), returned
122 anomalous PFAS concentrations suggesting the potential for collection or transit contamination
123 of these samples (Table S8). Reviewing the data for these three samples, in all cases analyte
124 concentrations fell toward the low end of our database, but they generally did not fall at the
125 lowest limits. There were no obvious packaging anomalies upon delivery receipt of these
126 samples, nor did we discern unusual concentrations or homologue distributions for the samples
127 themselves. Based on these observations that, by all metrics other than the sand blanks, these
128 samples were unexceptional, we retained these samples in our study, but results for these
129 samples should be regarded with a degree of caution.

130 As expected given the sensitivity of our instruments, low levels of PFASs, <50 pg/g
131 (parts per trillion) solvent, were detected in the procedural blanks (Table S9-S12). Of the 11
132 procedural blanks, prepared and analyzed interspersed among all samples run over the course of
133 the investigation, one blank returned PFCA concentrations up to ~100x greater than all other
134 blanks (Table S8). This blank was the first item handled when extraction activities were
135 transferred from one team member to another. Because the second blank prepared by this team
136 member was similar to the other blanks (Table S8), and because the five samples prepared with
137 this blank (NA06, NA09, NA11, NA19, AS01) had lower concentrations of most analytes than
138 this blank, we omitted this blank from detection-limit definition and retained the five samples in
139 the study, so these five samples should be regarded with caution.

140 For all data, we subtracted the mean concentrations of the ten process blanks from the
141 soil values we report herein. Regarding recovery, spiked M8PFOA ranged from 77-132% with
142 an average of 108% for all samples (Table S2-S4). We report numerical concentrations only if
143 they exceeded LOQ; values falling below LOQ, but above LOD, are reported as <LOQ, and
144 values below LOD are reported as <LOD. Finally, all eight samples associated with anomalous
145 sand (AF03, AF05, NA23) or process blanks (NA06, NA09, NA11, NA19, AS01), which should
146 be regarded with caution, are annotated as such in the SI tables.
147 **PFOA and PFOS Isomers.** Although qualitative, the distribution of PFOA and PFOS
148 isomers presented in Tables S25-S27 were investigated as a function of ΣPFCA and ΣPFSA
149 concentrations. When the percentage of linear PFOA was plotted against the log of ΣPFCA
150 concentration, a positive correlation ($r \cong 0.691$) was observed (Figure S8). A positive
151 correlation between the percentage of linear PFOS and the log of ΣPFSA concentration was also
152 observed (Figure S9), but was less pronounced ($r \cong 0.299$). The percent linear of historical ECF-
153 produced PFOA and PFOS was roughly 78% (De Silva et al. 2009) and 70% (Benskin et al.
154 2010), respectively, while the telomerization process produces 100% linear. The observation
155 that our results range from about 20% to 100% linear for ~60 data points in a statistically
156 significant pattern for both PFOA and PFOS (Figures S8 and S9) suggests that environmental
157 processes, e.g., variable sorption rates among isomers, are affecting isomer distribution in soils;
158 however, further research is necessary to ascertain this statistical relationship reflects any real
159 process.
160 **Relationship with total organic carbon (TOC).** As sorption of PFCAs and PFSAs in
161 solid matrices is believed to be driven by partitioning to OC (Higgins and Luthy 2006, You et al.
162 2010, Ahrens et al. 2011), TOC was measured for each soil sample and ranged from 0.1 for the
163 Antarctic sample (AN01) to 38.9% for Akumal, Mexico (NA06) with data presented in SI Tables

S2-S4. Unlike previous studies that reported an increase in sorption with the number of perfluorinated carbons (≥7 C_F's) (Higgins and Luthy 2006, You et al. 2010, Ahrens et al. 2011), no chain length dependency was observed in this study. Since volatility decreases with the number of perfluorinated carbons, the flux of longer chain congeners into the soil will depend on distance from local or regional sources. Gellrich et al. (2012) recently demonstrated that shorter chain congeners are only eliminated from a soil column in the presence of long chain congeners. These two factors may preclude any relationship between TOC and the number of perfluorinated carbons.

In addition, unlike POPs such as PBDEs and PCBs that have been demonstrated to have a correlation with TOC in background soils (Nam et al. 2008, Liu et al. 2011), no relationship was observed between TOC and total concentrations of PFCA or PFSA. Several factors such as the proximity to emission sources, precipitate and other soil properties differ significantly amongst sampling locations, which may preclude any direct comparison with TOC. Locations nearer emission sources have higher total concentrations of PFCA and PFSA, but may have a low fraction of TOC such as NA28-31 with TOC values ranging from 0.44 to 2.54%. Whereas several rural or remote locations could have higher TOC, but significantly lower PFCA and PFSA concentrations such as NA15 having a TOC value of 3.26%. The volume of precipitate could also impact the retention of PFCAs and PFSAs in the surface layer due to their water solubility. Davis et al. (2007) demonstrated that PFOA emitted from a nearby fluorochemical manufacturing facility was deposited in the surface soil, but was transported into the groundwater with increasing precipitation. However, further studies are needed to fully understand the impact precipitate has on the retention of PFCAs and PFSAs in the surface layer. Lastly, there are inherent differences in soil properties such as pH and cation exchange capacity (CEC), because of the geographical differences between sampling locations, which could suppress the sorption of PFCAs and PFSAs to TOC. For example, the bridging of anionic

189 PFCAs and PFSAs with cations such as Fe^{3+} and Ca^{2+} to the soils having net negative surfaces
190 has been proposed as an important sorption mechanism for PFOS to sediment (You et al. 2010).
191 Unfortunately, due to limited amounts of sample, further soil analyses were not performed in this
192 study, but this is an important area of research to completely understand the fate of PFCAs and
193 PFSAs in soils.
194 **Acknowledgements and sincere thanks to the samplers.** We express our deep gratitude to the
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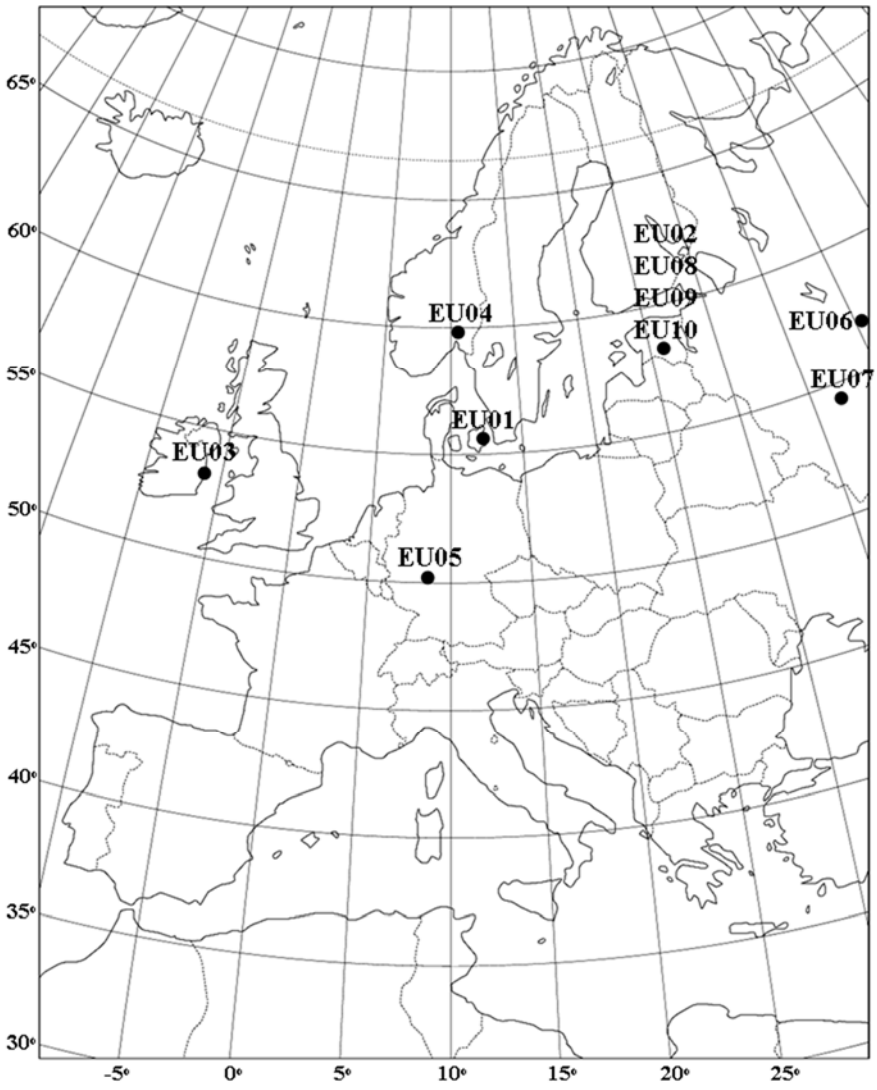


249

250 Figure S1: Approximate sampling locations in Europe (EU).

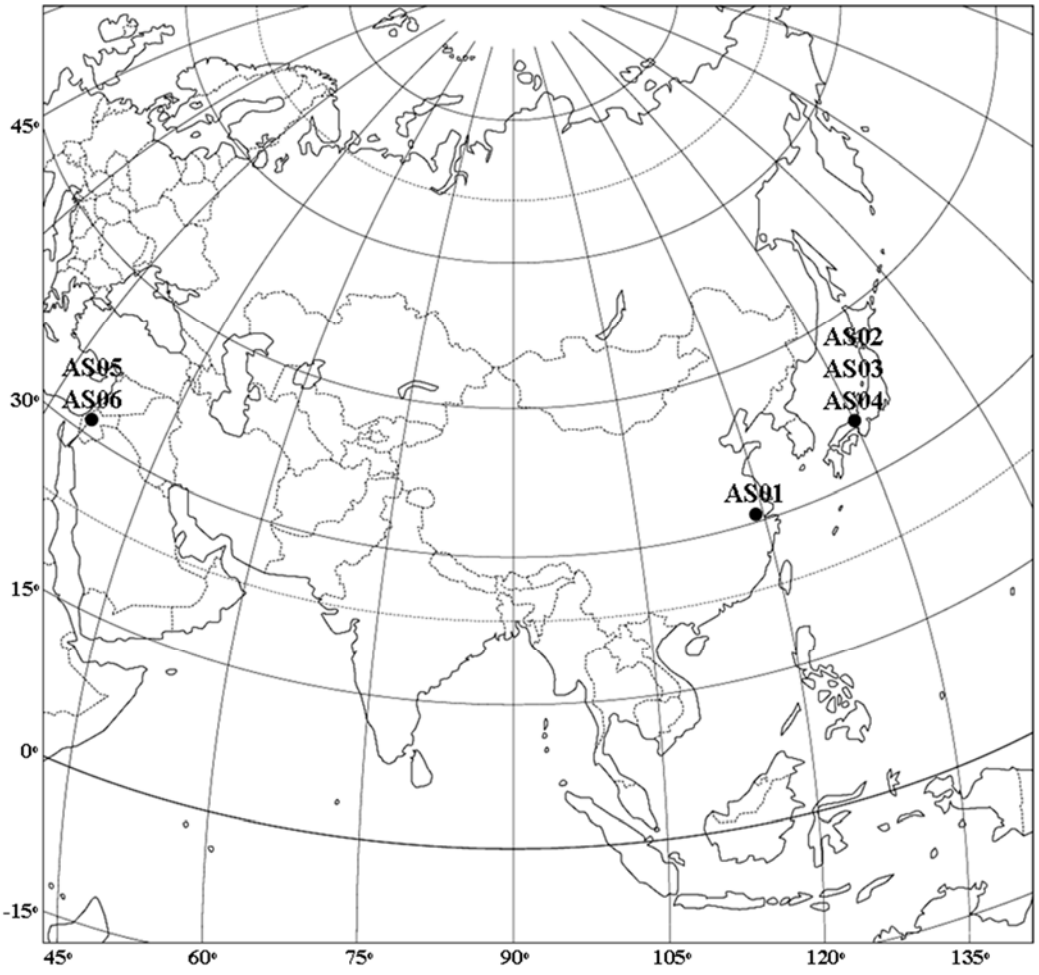
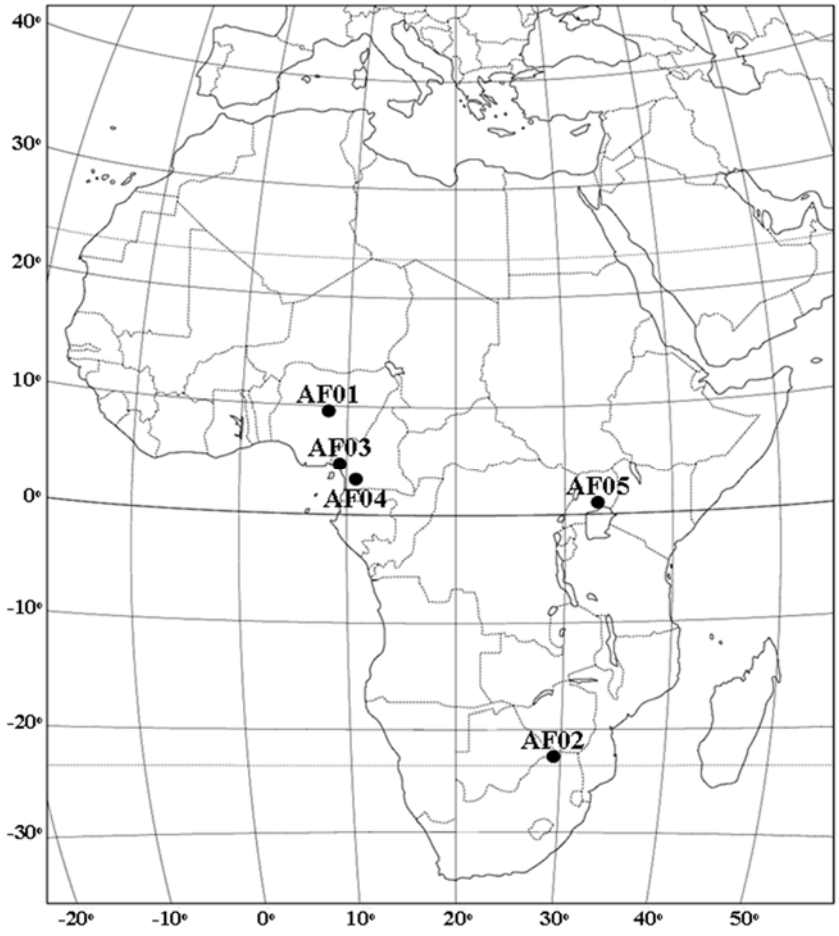


Figure S2: Approximate sampling locations in Asia (AS).



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256 Figure S3: Approximate sampling locations in Africa (AF).

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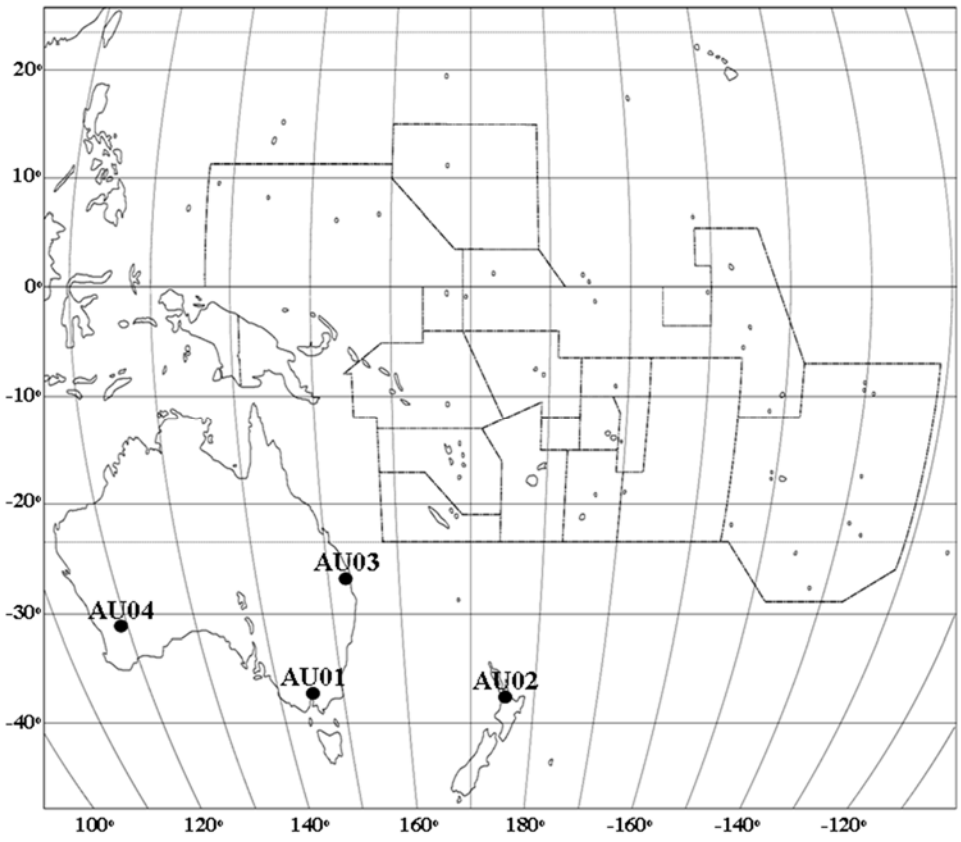

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259 Figure S4: Approximate sampling locations in continental Australia (AU).

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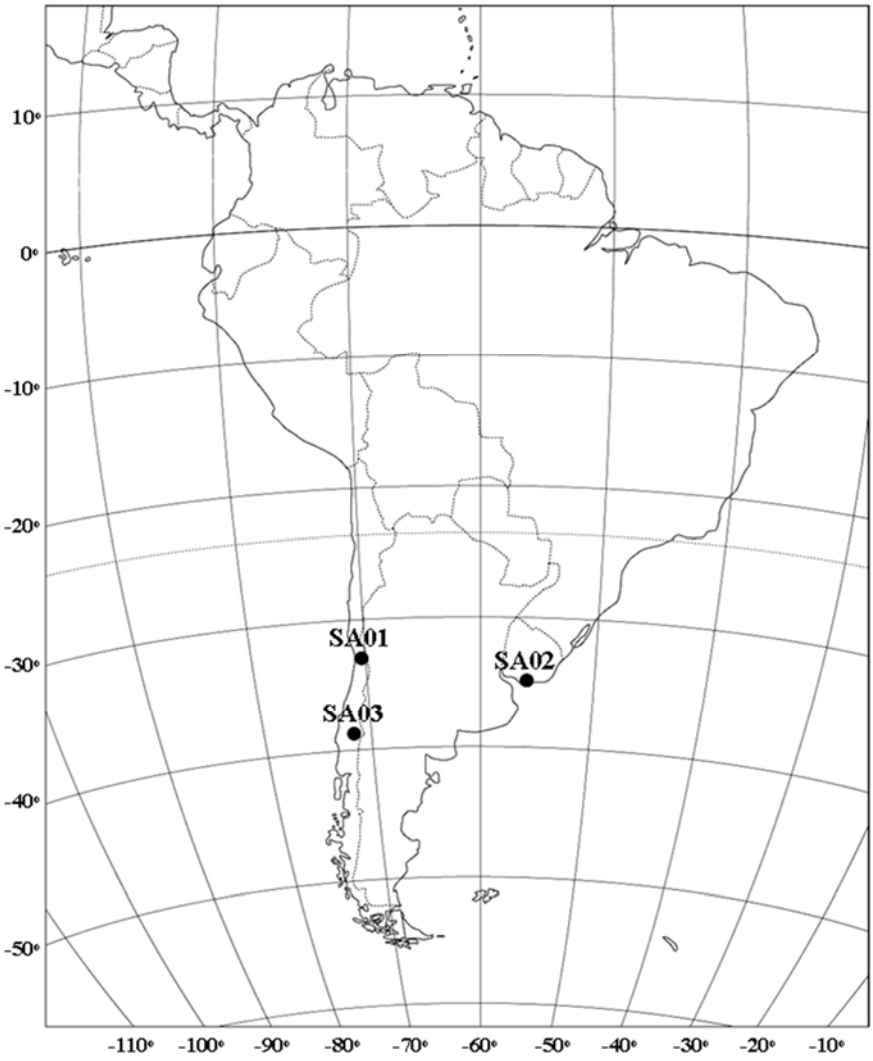

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262 Figure S5: Approximate sampling locations in South America (SA).

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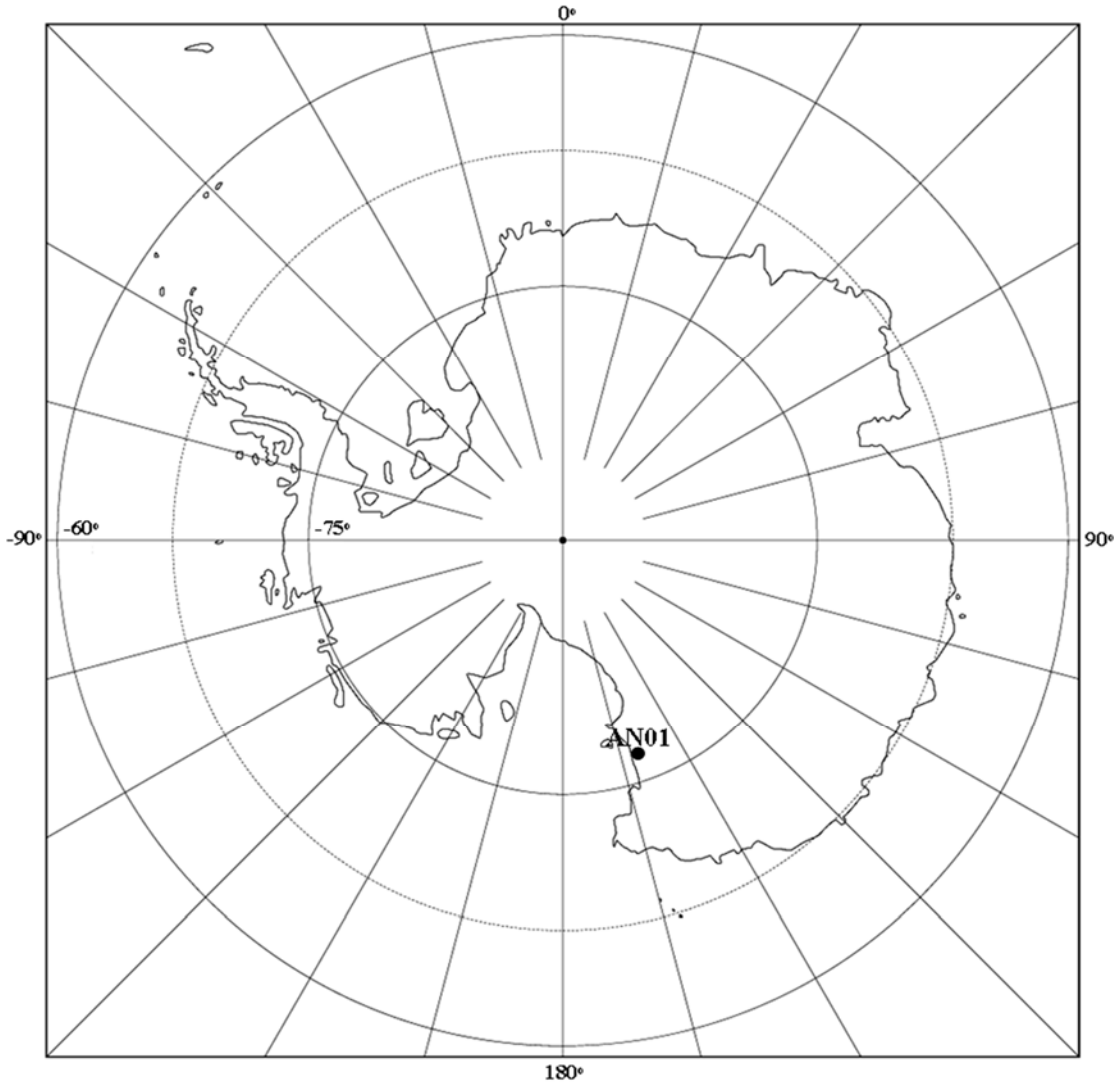


Figure S6: Approximate sampling locations in Antarctica (AN).

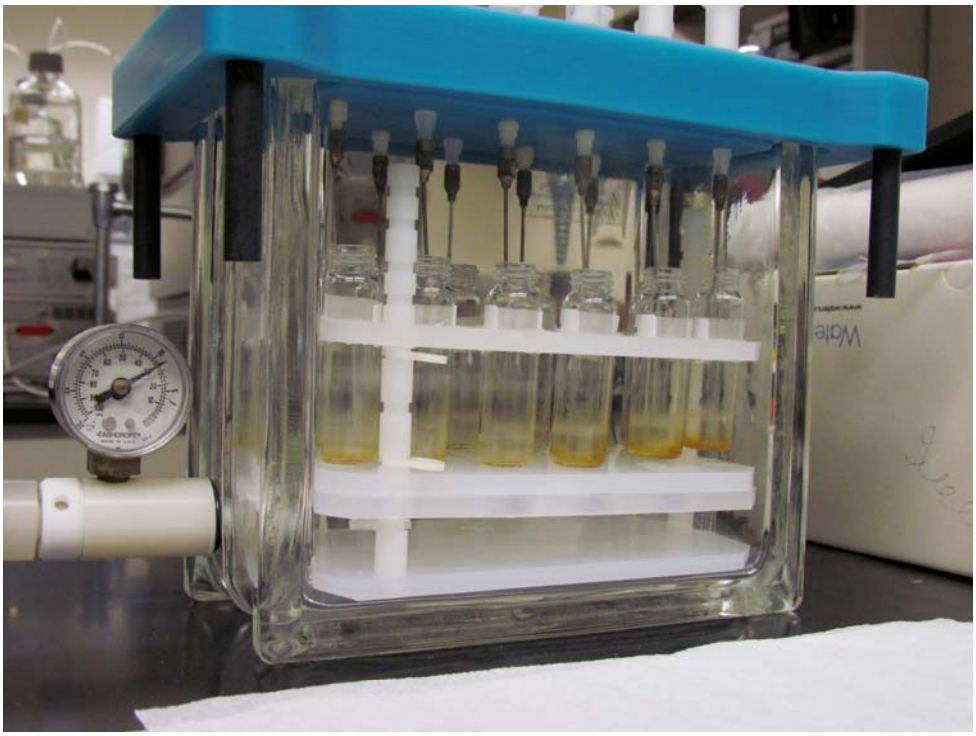
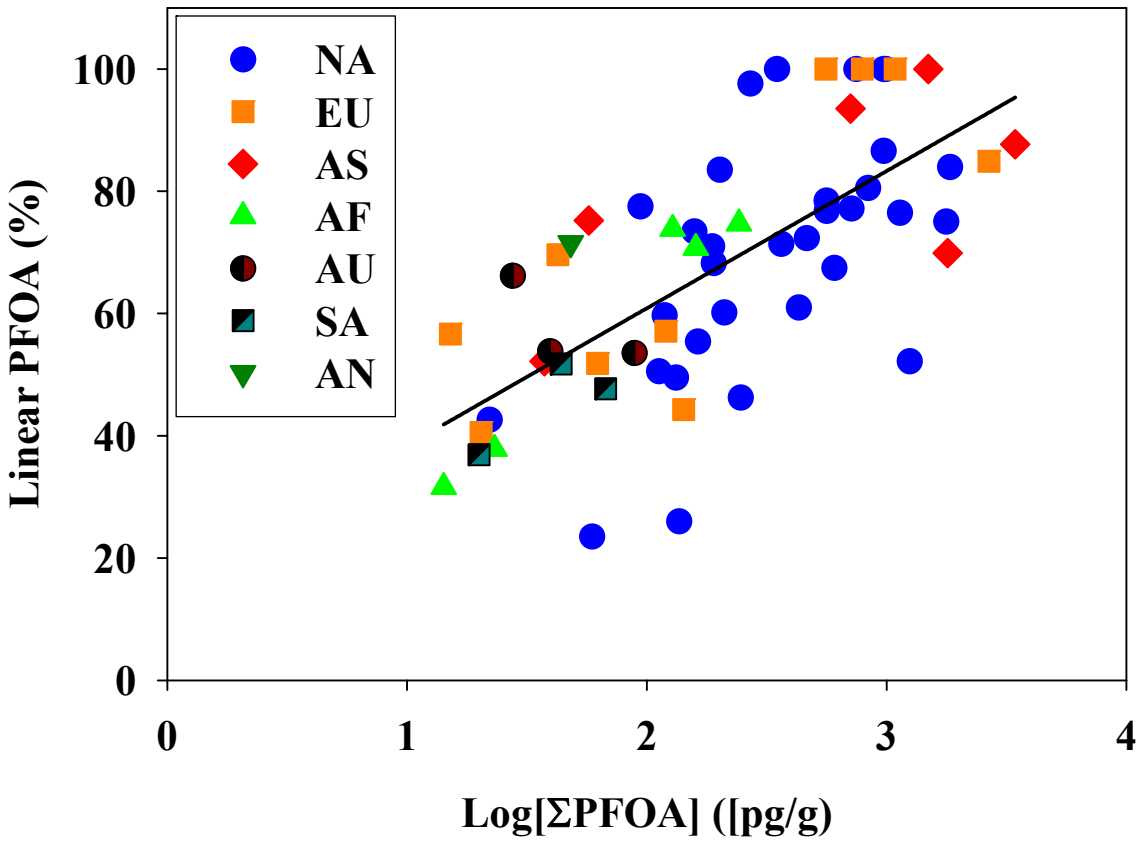


Figure S7: SPE manifold adapted for solvent evaporation.



273

274 Figure S8: Percentage of linear PFOA as a function of the log scale of ΣPFCAs. Note that the

275 line of least squared error in the plot yields a correlation coefficient of $r = 0.691$, which is

276 significant at $P < 0.0001$ for 59 degrees of freedom.

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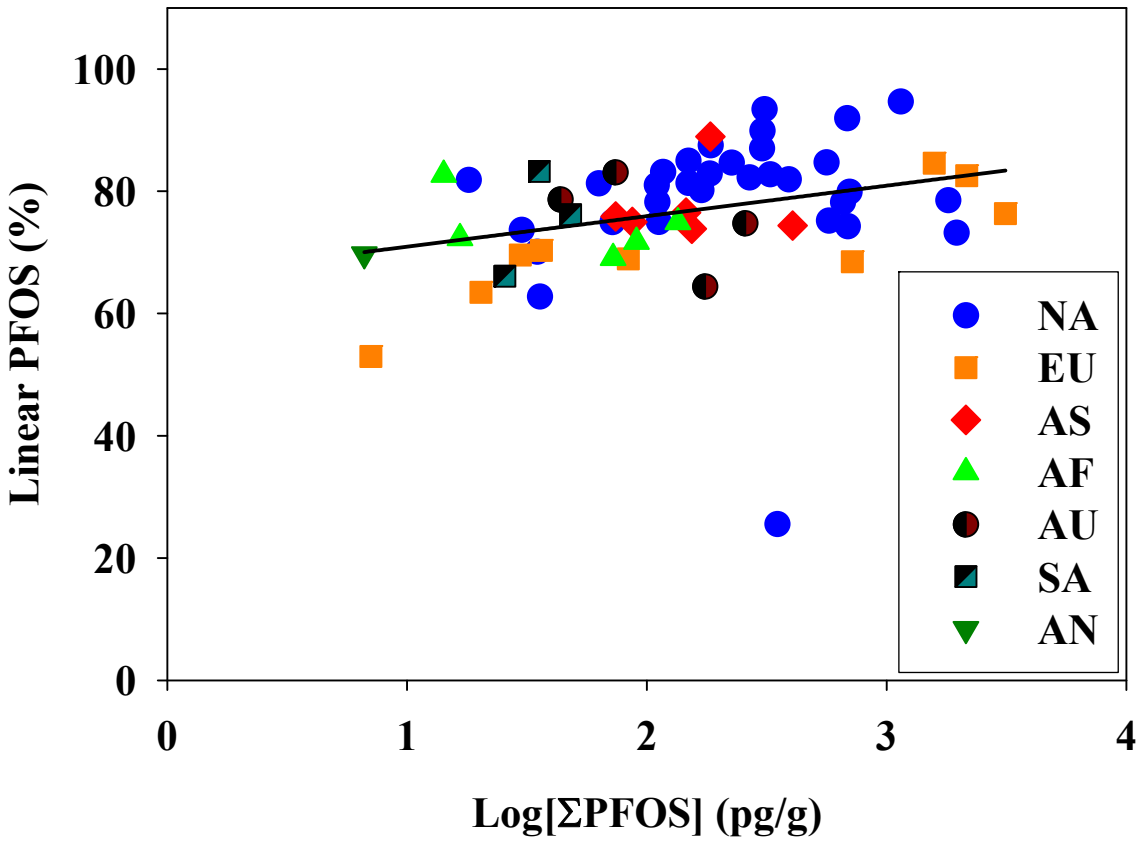

279

280 Figure S9: Percentage of linear PFOS as a function of the log scale of ΣPFSAs. Note that the

281 line of least squared error in the plot yields a correlation coefficient of $r = 0.299$, which is

282 significant at $P < 0.0205$ for 59 degrees of freedom.

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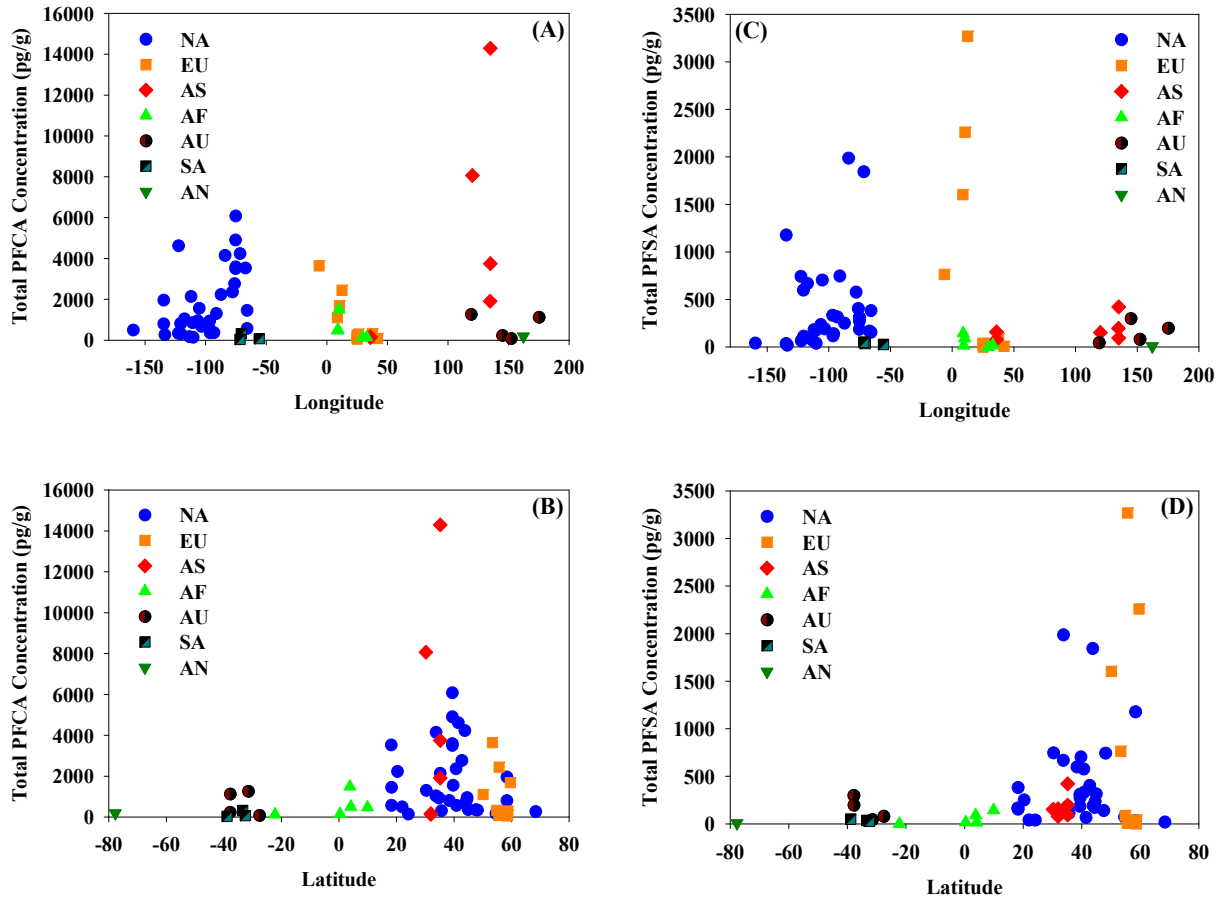
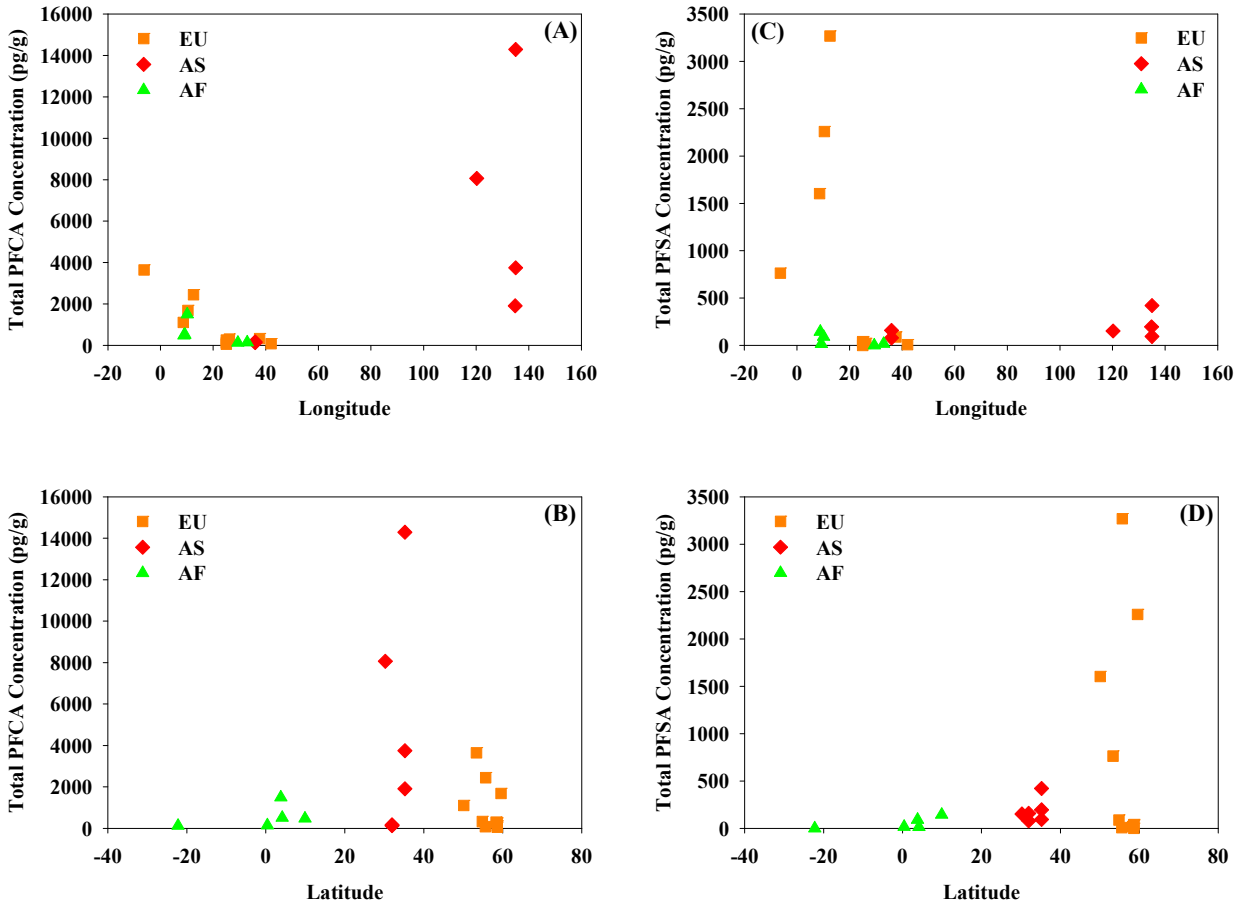


284

285 Figure S10: Longitudinal and latitudinal distribution of total PFCAs (**A** and **B**) and PFSAs (**C**

286 and **D**) in North America (NA), Europe (EU), Asia (AS), Africa (AF), Australia (AU), South

287 America (SA) and Antarctica (AN).



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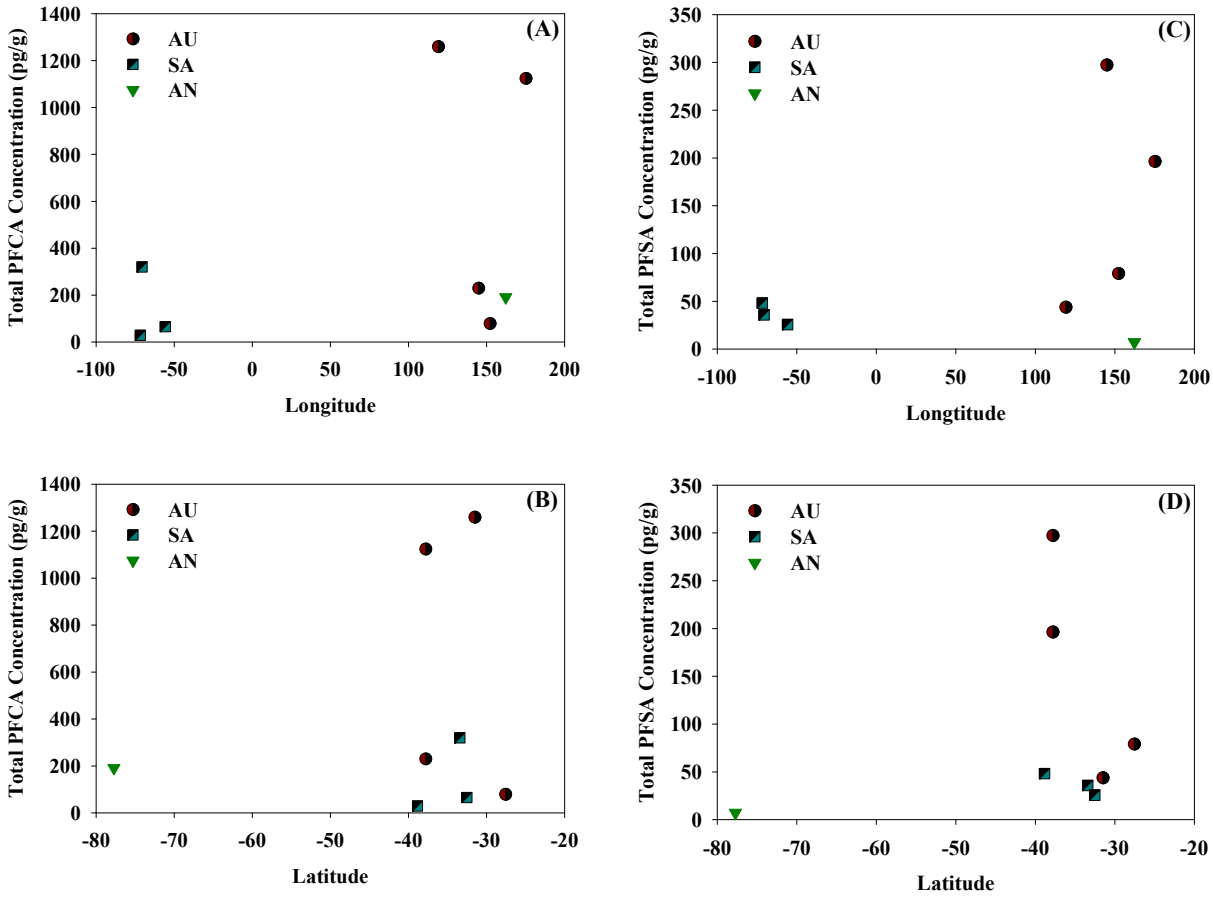
289 Figure S11: Longitudinal and latitudinal distribution of total PFCAs (**A** and **B**) and PFSAs (**C**

290 and **D**) in Europe (EU), Asia (AS) and Africa (AF).



291

292 Figure S12: Longitudinal and latitudinal distribution of total PFCAs (**A** and **B**) and PFSAs (**C**

293 and **D**) in Australia (AU), South America (SA) and Antarctica (AN).

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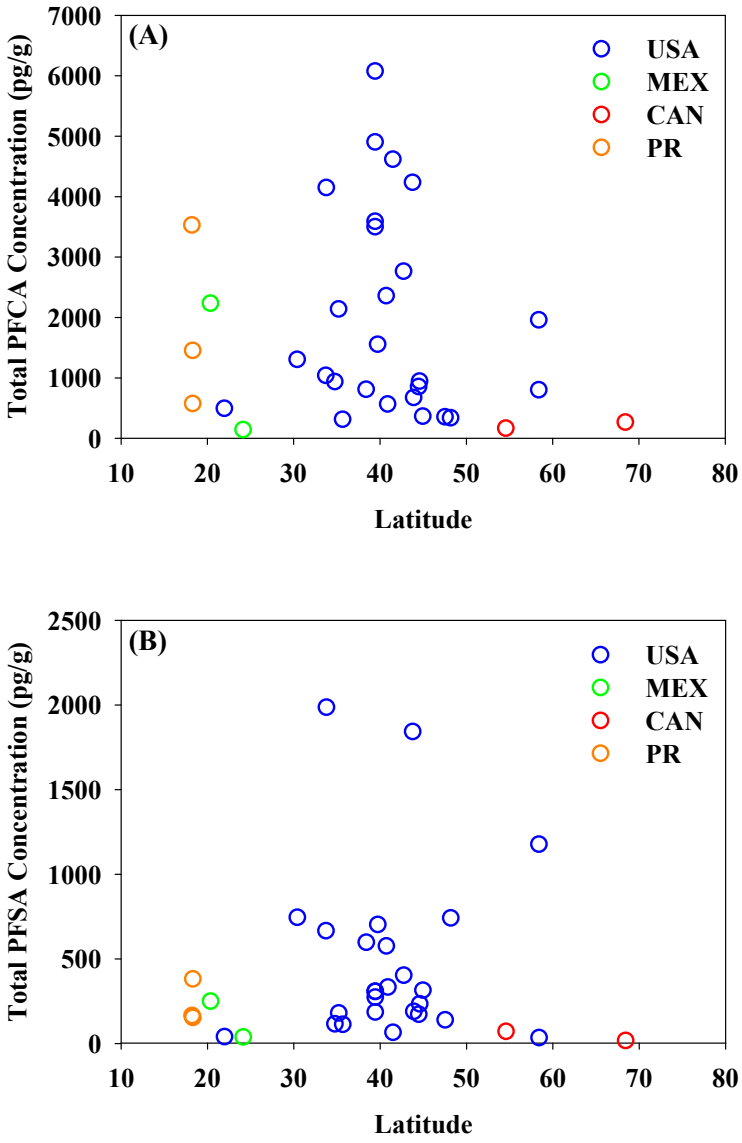

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298 Figure S13: Latitudinal distribution of total PFCAs (**A**) and PFSAs (**B**) in North American (NA)

299 surface soils.

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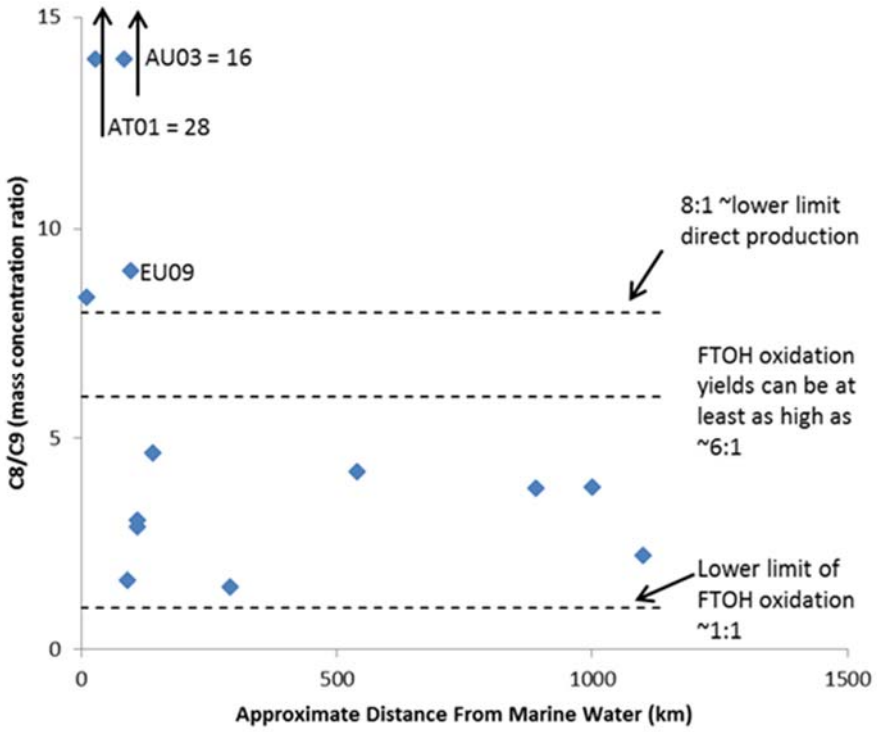

Figure S14: C8/C9 vs distance from marine water for the Antarctic sample (AN01) and 12 samples having lower ΣPFCAs than AN01 (selected to represent samples not showing evidence of local PFCA sources). Three samples that were non-detect for C9 were calculated at C9 = ½ lowest C9 detection. Only samples located within 100 km of coastal waters have higher C8/C9 than that expected for PFCAs being generated from oxidation of FTOHs in the atmosphere.

Table S1: Study analytes and mass spectral detection parameters

Compound	Parent m/z	Primary			Secondary			Standards		
		Fragment m/z	Cone Potential	Collision Energy	Fragment m/z	Cone Potential	Collision Energy	Calibration	Matrix Internal	Fragment m/z
	(D/esu)	(D/esu)	(V)	(eV)	(D/esu)	(V)	(eV)			(D/esu)
Perfluorocarboxylic Acids (PFCAs; C6-C14)										
PFHxA	312.80	268.85	13	10	118.80	13	20	C6	M2 C6	270.00
PFHpA	362.70	318.80	13	10	168.85	13	18	C7	M2 C6	270.00
PFOA	412.70	368.75	14	10	168.85	14	18	C8	M4 C8	372.00
PFNA	462.70	418.70	15	11	218.85	15	18	C9	M5 C9	423.00
PFDA	512.90	468.70	15	11	218.85	15	20	C10	M2 C10	470.00
PFUA	562.70	518.70	15	12	218.85	15	20	C11	M2 C11	520.00
PFDoA	612.70	568.70	16	13	318.70	16	20	C12	M2 C12	570.00
PFTrA	662.75	618.70	16	13	318.70	16	22	C13	M2 C12	570.00
PFTeA	712.75	668.70	18	14	318.70	18	24	C14	M2 C12	570.00
Unsaturated Telomer Acids (FTUCAs; 5-3 through 14-2)										
5-3FTUCA	339.00	269.00	16	18				6-2FTUCA	M2 6-2FTUCA	294.00
7-3FTUCA	439.00	369.00	16	17				8-2FTUCA	M2 8-2FTUCA	394.00
9-3FTUCA	539.00	469.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
11-3FTUCA	639.00	569.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
13-3FTUCA	739.00	669.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
6-2FTUCA	357.00	293.00	16	17	242.70	16	40	6-2FTUCA	M2 6-2FTUCA	294.00
8-2FTUCA	457.00	393.00	16	18	342.70	16	40	8-2FTUCA	M2 8-2FTUCA	394.00
10-2FTUCA	557.00	493.00	16	17	443.00	16	38	10-2FTUCA	M2 10-2FTUCA	494.00
12-2FTUCA	657.00	593.00	16	17	543.00	16	38	10-2FTUCA	M2 10-2FTUCA	494.00
14-2FTUCA	757.00	693.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
Saturated Telomer Acids (FTCAs; 5-3 through 14-2)										
5-3FTCA	341.00	237.00	17	12				6-2FTUCA	M2 6-2FTUCA	294.00
7-3FTCA	441.10	337.10	17	12				8-2FTUCA	M2 8-2FTUCA	394.00
9-3FTCA	541.00	437.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
11-3FTCA	641.00	537.00	17	12				10-2FTUCA	M2 10-2FTUCA	494.00
13-3FTCA	741.00	637.00	17	12				10-2FTUCA	M2 10-2FTUCA	494.00
6-2FTCA	377.00	293.00	16	17				6-2FTUCA	M2 6-2FTUCA	294.00
8-2FTCA	477.00	393.00	16	17				8-2FTUCA	M2 8-2FTUCA	394.00
10-2FTCA	577.00	493.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
12-2FTCA	677.00	593.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
14-2FTCA	777.00	693.00	16	17				10-2FTUCA	M2 10-2FTUCA	494.00
Sulfonates (PFSAs; S6, S8, S10)										
S6		79.85	50	40	98.85	50	40	S6	M4 S6	83.90
S8		79.85	60	50	98.85	60	40	S8	M4 S8	83.90
S10		79.90	70	50				S10	M4 S8	83.90

316 Table S2: Sample locations, total organic carbon, PFOA percent recovery and summed analyte
317 concentration for soils collected from North America (NA).

Sample ID	Location	TOC (%)	Recovery (%)	ΣPFCA (pg/g)	ΣPFSA (pg/g)	ΣFTCA (pg/g)	ΣFTUCA (pg/g)
NA01	St. Paul (MN), USA (44.88312, -93.55454)	0.81	95.68 ± 6.06	367.33	315.13	0.55	0.00
NA02	Conyers (GA), USA (33.763513, -83.9929)	3.63	118.43 ± 8.56	4150.63	1986.68	44.00	105.55
NA03	Fertile (MN), USA (47.5306, -96.3032)	2.84	132.12 ± 33.68	357.25	139.31	0.00	0.00
NA04	Cortland (NY), USA (42.660222, -76.29124)	4.48	98.89 ± 17.87	2766.87	403.76	6.01	2.42
NA05	Seward (NE), USA (40.874639, -96.98603)	4.59	131.22 ± 10.10	567.17	333.85	0.00	0.27
NA06	Akumal, Mexico (20.35187, -87.36972)	38.87	77.64 ± 12.79	2234.04	250.17	1.75	0.00
NA07	Auke Bay (AK), USA (58.380861, -134.7324)	2.25	108.03 ± 12.11	802.44	34.98	0.00	0.00
NA08	Ada (OK), USA (34.581214, -96.85593)	0.74	120.92 ± 7.49	939.01	116.92	0.00	0.00
NA09	Mt. Zion (CO), USA (39.743956, -105.2431)	3.21	97.46 ± 11.57	810.86	598.06	1.72	0.00
NA10	Kaibab National Forest (AZ), USA (35.191833, -112.0088)	2.10	110.52 ± 1.96	2140.68	181.32	0.71	0.00
NA11	Whipple Dam State Park (PA), USA (40.709889, -77.881)	9.91	100.17 ± 10.77	2361.24	577.00	0.00	0.00
NA12	Cleveland National Forest (CA), USA (33.654709, -117.4055)	2.73	86.21 ± 17.53	1041.31	667.02	1.89	1.44
NA13	Shasta-Trinity National Forest (CA), USA (41.478892, -122.3592)	1.53	111.93 ± 11.63	4618.12	66.39	40.93	53.22
NA14	Baton Rouge (LA), USA (30.385292, -91.13874)	2.10	123.25 ± 12.43	1306.07	746.31	1.17	0.00
NA15	Inuvik (NWT), Canada (68.415056, -133.766)	3.26	121.08 ± 5.45	269.30	18.09	0.00	0.00
NA16	La Paz, Mexico (24.176825, -110.2942)	0.49	102.26 ± 11.62	144.82	39.08	0.00	0.00
NA17	Holderness (NH), USA (43.7662, -71.6709)	10.85	104.52 ± 12.46	4236.78	1843.32	0.00	0.00
NA18	Meanook (AB), Canada (54.611846, -113.3453)	3.07	119.74 ± 6.25	169.86	71.80	0.00	0.00
NA19	Waimea (HI), USA (21.96836, -159.6848)	14.97	129.54 ± 26.84	496.13	40.21	0.00	0.00
NA20	Maraguez, Puerto Rico (18.099437, -66.59098)	2.18	110.11 ± 9.81	3531.38	165.21	13.56	8.68
NA21	Limon (CO), USA (39.265221, -103.6454)	10.70	115.61 ± 16.00	1557.62	703.53	0.00	2.74
NA22	Shandon (CA), USA (35.6521, -120.4118)	2.05	103.54 ± 1.26	314.20	112.92	0.00	0.61
NA23	El Yunque National Forest, Puerto Rico (18.294395, -65.78433)	6.30	96.38 ± 4.50	576.00	154.09	0.00	0.00
NA24	El Yunque National Forest, Puerto Rico (18.294395, -65.78433)	6.67	120.58 ± 23.15	1454.52	381.59	0.00	0.00
NA25	Yellowstone National Park (WY), USA (44.4628, -110.8264)	13.23	104.49 ± 17.03	857.07	173.30	1.02	20.28
NA26	Clearmont (WY), USA (44.5683, -106.5603)	6.14	110.27 ± 9.99	946.99	234.73	0.00	0.00
NA27	Keystone (SD), USA (43.8922, -103.4212)	3.82	116.29 ± 3.42	675.12	190.20	0.00	0.00
NA28	Penns Grove (NJ), USA (39.7158, -75.4555)	0.77	117.43 ± 23.81	3592.74	274.94	18.84	0.00
NA29	Penns Grove (NJ), USA (39.7123, -75.4529)	2.54	99.66 ± 7.09	6078.57	309.70	48.51	0.40
NA30	Penns Grove (NJ), USA (39.7015, -75.4493)	1.24	105.12 ± 19.65	4904.67	307.81	33.49	1.40
NA31	Penns Grove (NJ), USA (39.7014, -75.4481)	0.44	100.54 ± 17.88	3503.48	185.82	25.97	0.00
NA32	Fort Casey (WA), USA (48.164912, -122.6776)	6.08	122.48 ± 18.72	341.19	742.36	0.00	0.00
NA33	Juneau (AK), USA (58.371208, -134.594)	0.54	98.77 ± 12.71	1960.94	1177.24	37.25	0.00

318

Table S3: Sample locations, total organic carbon, PFOA percent recovery and summed analyte concentration for soils collected from Europe (EU), Asia (AS) and Africa (AF).

Sample ID	Location	TOC (%)	Recovery (%)	ΣPFCA (pg/g)	ΣPFSA (pg/g)	ΣFTCA (pg/g)	ΣFTUCA (pg/g)
EU01	Copenhagen, Denmark (55.676825, 12.556203)	3.98	115.32 ± 10.44	2444.09	3268.52	3.41	0.00
EU02	Vehendi, Estonia (58.20803, 26.11963)	0.66	111.14 ± 17.35	300.17	20.34	0.92	0.00
EU03	Dublin, Ireland (53.256998, -6.336594)	14.89	110.61 ± 8.06	3642.98	762.37	0.00	0.00
EU04	Olso, Norway (59.939333, 10.71825)	8.52	102.67 ± 10.94	1683.06	2258.28	1.22	0.00
EU05	Frankfurt, Germany (50.113478, 8.644352)	1.54	115.22 ± 2.56	1105.84	1602.62	3.83	0.00
EU06	Murom, Russia (55.582948, 42.046496)	0.37	94.26 ± 10.96	77.81	7.06	0.00	0.00
EU07	Puschino, Russia (54.842077, 37.64177)	3.18	106.97 ± 6.19	327.54	88.00	0.00	0.00
EU08	Vehendi, Estonia (58.219192, 26.132644)	1.09	107.29 ± 12.75	125.50	39.61	0.00	0.00
EU09	Vehendi, Estonia (58.20803, 26.11963)	0.54	101.00 ± 18.23	54.55	0.00	0.00	0.00
EU10	Vehendi, Estonia (58.210083, 26.107722)	1.05	108.49 ± 9.05	252.09	33.28	0.00	0.00
AS01	Hangzhou, China (30.264186, 120.1906)	1.19	94.90 ± 4.81	8060.43	151.47	32.30	10.63
AS02	Asago, Japan (35.299821, 134.82888)	1.30	112.58 ± 14.45	1909.16	195.17	0.00	0.00
AS03	Hikaminuma, Japan (35.228251, 135.01375)	8.21	103.07 ± 9.40	3747.08	420.68	0.00	0.00
AS04	Hikaminuma, Japan (35.228251, 135.01375)	1.03	104.72 ± 11.34	14294.28	93.90	26.20	53.09
AS05	Amman, Jordan (32.008767, 35.877349)	6.23	121.30 ± 19.98	128.78	78.83	0.00	0.47
AS06	Amman, Jordan (32.008767, 35.877349)	6.19	99.36 ± 3.88	166.88	158.73	0.00	0.00
AF01	Jos, Nigeria (9.884981, 8.942642)	1.81	114.76 ± 5.34	471.10	143.93	0.00	6.08
AF02	Mapunguwe National Park, South Africa (-22.25, 29.333333)	0.51	100.26 ± 13.29	123.69	0.00	0.00	4.82
AF03	Buea, Cameroon (4.1573, 9.2697)	0.69	117.46 ± 12.77	508.28	14.22	1.58	0.00
AF04	Edea, Cameroon (3.8122, 10.1372)	2.45	104.66 ± 10.22	1494.23	90.59	0.00	0.00
AF05	Mabira Forest Reserve, Uganda (0.4052, 32.9747)	5.18	104.79 ± 12.47	143.70	16.64	0.00	0.00

Table S4: Sample locations, total organic carbon, PFOA percent recovery and summed analyte concentration for soils collected from Australia (AU), South America (SA) and Antarctica (AN).

Sample ID	Location	TOC (%)	Recovery (%)	ΣPFCA (pg/g)	ΣPFSA (pg/g)	ΣFTCA (pg/g)	ΣFTUCA (pg/g)
AU01	Parkville, Australia (-37.76854, 144.65103)	2.72	107.05 ± 18.61	229.63	297.21	0.00	0.00
AU02	Hamilton, New Zealand (-37.78537, 175.2071)	17.06	103.38 ± 31.10	1123.86	196.22	0.00	0.00
AU03	Gatton, Australia (-27.55778, 152.34389)	2.94	100.81 ± 17.05	79.18	79.02	0.00	0.00
AU04	Southern Cross, Australia (-31.48682, 119.19243)	1.79	100.79 ± 5.98	1259.58	43.73	0.00	0.00
SA01	Santiago, Chile (-33.62377, -70.4837)	2.14	107.31 ± 10.98	319.28	35.64	0.00	0.00
SA02	Montevideo, Uruguay (-34.82508, -56.37932)	2.96	120.06 ± 29.93	65.08	25.57	0.00	0.00
SA03	San Bernardo (Chena Peak), Chile (-33.5942, -70.73)	0.98	103.17 ± 6.24	28.71	48.07	0.00	0.00
AN01	Lake Bonney, Antartica (-77.71568, 162.29942)	0.10	100.37 ± 16.31	191.20	7.20	0.00	0.00

348 Table S5: Summary of PFOA and PFOS concentrations in North American sampling sand

349 blanks.

Sample ID	PFOA (pg/g)	PFOS (pg/g)
NA01	55.33 ± 5.05	6.60 ± 0.49
NA02	<LOD	4.61 ± 1.01
NA03	21.19 ± 0.33	6.83 ± 1.91
NA04	16.31 ± 1.66	7.86 ± 2.72
NA05	13.17 ± 1.20	4.95 ± 0.80
NA06	12.71 ± 1.47	7.78 ± 1.55
NA07	16.89 ± 0.55	7.49 ± 2.31
NA08	10.81 ± 1.61	15.34 ± 4.03
NA09	9.90 ± 2.54	6.08 ± 0.64
NA10	20.66 ± 0.92	6.19 ± 2.16
NA11	34.82 ± 3.93	9.97 ± 4.37
NA12	20.06 ± 0.75	7.35 ± 2.50
NA13	11.10 ± 1.40	14.22 ± 1.96
NA14	74.26 ± 2.30	5.56 ± 2.50
NA15	19.78 ± 2.41	7.06 ± 1.39
NA16	19.79 ± 0.98	6.24 ± 1.26
NA17	<LOQ	5.14 ± 0.80
NA18	25.88 ± 2.61	<LOD
NA19	44.77 ± 3.09	3.90 ± 0.66
NA20	<LOQ	4.15 ± 0.74
NA21	72.94 ± 4.71	4.87 ± 1.63
NA22	27.97 ± 1.99	3.82 ± 0.42
NA23	101.08 ± 10.35	3.22 ± 1.22
NA24	19.67 ± 0.49	2.58 ± 0.37
NA25	17.67 ± 2.27	4.64 ± 2.08
NA26	22.90 ± 9.86	40.09 ± 0.36
NA27	<LOQ	6.13 ± 0.55
NA28	12.46 ± 1.48	5.03 ± 1.50
NA29	15.43 ± 1.31	5.80 ± 1.12
NA30	<LOQ	7.14 ± 0.94
NA31	<LOQ	5.59 ± 1.73
NA32	<LOQ	8.80 ± 1.97
NA33	10.58 ± 0.52	3.95 ± 0.77

350

Table S6: Summary of PFOA and PFOS concentrations in European, Asian and African sampling sand blanks.

Sample ID	PFOA (pg/g)	PFOS (pg/g)
EU01	19.85 ± 2.56	6.42 ± 0.49
EU02	<LOQ	5.66 ± 1.61
EU03	<LOQ	5.92 ± 1.12
EU04	9.17 ± 1.22	6.26 ± 2.53
EU05	11.65 ± 2.59	7.38 ± 1.11
EU06	8.85 ± 2.02	8.77 ± 0.79
EU07	9.51 ± 0.61	6.13 ± 0.86
EU08	13.12 ± 2.07	4.27 ± 1.19
EU09	9.91 ± 1.55	3.07 ± 0.44
EU10	<LOQ	5.13 ± 1.15
AS01	42.61 ± 1.49	7.55 ± 2.32
AS02	28.22 ± 7.25	<LOD
AS03	ND	ND
AS04	ND	ND
AS05	<LOQ	11.22 ± 3.40
AS06	16.14 ± 2.47	7.72 ± 0.44
AF01	9.35 ± 1.51	9.77 ± 3.79
AF02	37.93 ± 2.59	4.18 ± 1.94
AF03	434.18 ± 31.57	5.83 ± 1.07
AF04	13.26 ± 0.47	7.11 ± 1.75
AF05	104.36 ± 7.36	4.64 ± 2.08

Table S7: Summary of PFOA and PFOS concentrations in Australian, South American and Antarctic sampling sand blanks.

Sample ID	PFOA (pg/g)	PFOS (pg/g)
AU01	10.12 ± 0.93	7.68 ± 2.59
AU02	12.52 ± 1.42	4.62 ± 1.67
AU03	18.00 ± 10.10	19.91 ± 4.06
AU04	21.77 ± 2.15	4.57 ± 1.13
SA01	10.67 ± 0.88	4.62 ± 1.50
SA02	22.15 ± 0.94	8.32 ± 2.62
SA03	10.01 ± 3.07	5.57 ± 1.66
AN01	ND	ND

Table S8: Identification of anomalous sand blanks.

Summary Statistic	All data		Filtered (1)
	PFOA (pg/g)	PFOS (pg/g)	PFOA (pg/g)
Mean	26.51	6.62	16.68
Stan. Dev.	57.98	5.42	16.36
Coef. Var.	2.19	0.82	0.98

(1) With a PFOA coefficient of variation (COV) for all sand blanks exceeding 1 suggesting non-normal distribution, we filtered 3 blanks exceeding mean +1 standard deviation (NA23, AF03 and AF05) which dropped COV < 1. Based on these PFOA data for blanks, field contamination cannot be ruled out for NA23, AF03 and AF05.

Table S9: Summary of PFCA concentrations and statistical treatment of procedural blanks.

Process Blank	PFHxA (pg/g)	PFHpA (pg/g)	PFOA (pg/g)	PFNA (pg/g)	PFDA (pg/g)	PFUnDA (pg/g)	PFDoDA (pg/g)	PFTrDA (pg/g)	PFTeDA (pg/g)
B1A	2.0	4.4	43.3	10.5	8.7	1.8	8.7	4.3	<LOD
B1B	6.1	0.2	25.9	13.3	6.5	2.6	6.4	0.6	<LOD
B1C	0.0	10.7	25.9	6.3	9.2	7.9	4.4	3.9	<LOD
B2A	4.5	11.9	37.7	8.2	6.4	3.7	2.5	<LOD	4.1
B2B	9.0	13.7	70.3	6.4	27.5	10.0	54.2	<LOD	<LOD
B2C	5.4	2.7	60.6	5.1	15.0	4.1	3.7	<LOD	<LOD
B3A	20.0	7.6	60.7	5.5	14.4	3.9	6.3	1.0	<LOD
B3B	11.3	12.1	57.3	11.5	15.7	3.0	2.7	1.4	1.4
B3C	13.2	5.2	51.8	4.3	5.1	1.5	2.5	<LOD	3.4
TB1	132.1	283.4	1040.3	1806.4	872.1	463.8	363.0	81.5	95.5
TB2	24.4	8.8	35.4	19.4	18.2	0.6	17.6	0.1	<LOD
Summary Statistics for All Blanks									
Mean	20.7	32.8	137.2	172.4	90.8	45.7	42.9	8.4	9.5
Stan. Dev.	37.7	83.2	299.9	541.9	259.2	138.7	107.2	24.3	28.6
Coef. Var.	1.82	2.54	2.19	3.14	2.86	3.03	2.50	2.88	3.01
Summary Statistics Omitting Blank TB1									
Mean	9.6	7.7	46.9	9.0	12.7	3.9	10.9	1.1	0.9
Stan. Dev.	7.8	4.5	15.5	4.7	6.9	2.9	15.9	1.6	1.6
Coef. Var.	0.81	0.58	0.33	0.52	0.55	0.75	1.46	1.46	1.77

386 Table S10: Summary of PFSA concentrations and statistical treatment of procedural blanks.

Process Blank	PFHxS (pg/g)	PFOS (pg/g)	PFDS (pg/g)
B1A	<LOD	0.5	<LOD
B1B	<LOD	<LOD	<LOD
B1C	<LOD	0.3	<LOD
B2A	7.7	3.5	<LOD
B2B	6.8	1.8	<LOD
B2C	1.3	0.2	<LOD
B3A	<LOD	0.4	<LOD
B3B	<LOD	0.5	<LOD
B3C	0.1	<LOD	<LOD
TB1	<LOD	1.2	<LOD
TB2	<LOD	0.9	<LOD
Summary of Statistics for All Blanks			
Mean	1.4	0.9	<LOD
Stan. Dev.	2.9	1.0	-
Coef. Var.	2.0	1.2	-

387

388

389 Table S11: Summary of FTCA concentrations and statistical treatment of procedural blanks.

Process Blank	5:3 FTCA (pg/g)	7:3 FTCA (pg/g)	9:3 FTCA (pg/g)	11:3 FTCA (pg/g)	13:3 FTCA (pg/g)	6:2 FTCA (pg/g)	8:2 FTCA (pg/g)	10:2 FTCA (pg/g)	12:2 FTCA (pg/g)	14:2 FTCA (pg/g)
B1A	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B1B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B1C	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B2A	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B2B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B2C	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B3A	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B3B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B3C	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
TB1	<LOD	<LOD	1.3	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
TB2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Summary Statistics for All Blanks										
Mean	<LOD	<LOD	0.1	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Stan. Dev.	-	-	0.4	-	-	-	-	-	-	-
Coef. Var.	-	-	3.3	-	-	-	-	-	-	-

390

391

Table S12: Summary of FTUCA concentrations and statistical treatment of procedural blanks.

Process Blank	5:3 FTUCA (pg/g)	7:3 FTUCA (pg/g)	9:3 FTUCA (pg/g)	11:3 FTUCA (pg/g)	13:3 FTUCA (pg/g)	6:2 FTUCA (pg/g)	8:2 FTUCA (pg/g)	10:2 FTUCA (pg/g)	12:2 FTUCA (pg/g)	14:2 FTUCA (pg/g)
B1A	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.4	<LOD	<LOD
B1B	<LOD	<LOD	<LOD	<LOD	<LOD	1.5	1.3	<LOD	<LOD	<LOD
B1C	<LOD	<LOD	<LOD	<LOD	<LOD	1.4	1.2	<LOD	<LOD	<LOD
B2A	<LOD	<LOD	<LOD	<LOD	<LOD	0.8	<LOD	<LOD	<LOD	<LOD
B2B	<LOD	<LOD	<LOD	<LOD	<LOD	1.4	0.3	1.0	<LOD	<LOD
B2C	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
B3A	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	13.0	<LOD	<LOD	<LOD
B3B	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	12.9	13.6	<LOD	<LOD
B3C	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	12.9	13.7	<LOD	<LOD
TB1	<LOD	<LOD	<LOD	<LOD	<LOD	1.7	12.5	14.6	<LOD	<LOD
TB2	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.5	5.4	<LOD	<LOD
Summary Statistics for All Blanks										
Mean	<LOD	<LOD	<LOD	<LOD	<LOD	0.6	5.2	4.4	<LOD	<LOD
Stan. Dev.	-	-	-	-	-	0.7	6.1	6.3	-	-
Coef. Var.	-	-	-	-	-	1.2	1.2	1.4	-	-

Table S13: Summary of PFCA concentrations in North American samples.

Sample ID	PFHxA (pg/g)	PFHpA (pg/g)	PFOA (pg/g)	PFNA (pg/g)	PFDA (pg/g)	PFUnDA (pg/g)	PFDoDA (pg/g)	PFTrDA (pg/g)	PFTeDA (pg/g)
NA01	62.54 ± 11.34	22.66 ± 2.39	157.66 ± 6.78	52.69 ± 9.38	28.66 ± 4.33	25.39 ± 2.33	12.37 ± 2.87	1.95 ± 1.12	3.41 ± 0.73
NA02	997.62 ± 103.57	478.62 ± 36.72	1770.92 ± 195.65	271.47 ± 78.49	226.08 ± 44.52	237.70 ± 21.46	141.92 ± 20.69	26.30 ± 8.17	<LOQ
NA03	60.03 ± 13.90	57.96 ± 5.38	132.13 ± 34.72	46.72 ± 9.32	23.39 ± 5.27	35.51 ± 0.54	<LOQ	1.51 ± 0.57	<LOD
NA04	785.42 ± 169.77	428.41 ± 96.97	1137.30 ± 234.94	164.38 ± 35.33	95.78 ± 39.18	64.34 ± 1.72	61.22 ± 49.22	15.41 ± 7.20	14.61 ± 5.47
NA05	94.37 ± 25.05	112.88 ± 20.43	190.04 ± 9.42	94.41 ± 33.80	23.11 ± 5.99	41.10 ± 2.47	11.26 ± 1.44	<LOQ	<LOD
NA06	160.87 ± 49.14	449.91 ± 106.22	972.06 ± 60.47	337.83 ± 70.32	82.03 ± 18.35	196.39 ± 73.97	30.49 ± 14.02	4.46 ± 1.76	<LOD
NA07	417.50 ± 1.78	149.13 ± 10.33	163.42 ± 23.23	46.56 ± 13.52	11.46 ± 4.25	9.83 ± 4.48	<LOQ	3.19 ± 1.18	1.35 ± 0.64
NA08	111.23 ± 14.91	226.37 ± 10.17	464.42 ± 34.99	95.93 ± 21.40	16.91 ± 4.81	16.75 ± 1.47	<LOQ	5.29 ± 1.20	2.11 ± 1.11
NA09	145.43 ± 10.60	129.38 ± 14.86	349.22 ± 50.74	92.94 ± 16.17	43.42 ± 7.48	25.29 ± 2.51	19.86 ± 3.40	5.32 ± 0.77	<LOQ
NA10	402.03 ± 149.09	276.08 ± 50.78	746.80 ± 101.63	505.10 ± 11.26	106.48 ± 17.25	63.55 ± 25.90	18.15 ± 0.27	19.72 ± 9.83	2.77 ± 2.43
NA11	266.04 ± 14.47	318.22 ± 44.90	605.70 ± 50.46	244.06 ± 68.23	204.40 ± 48.39	586.04 ± 197.87	91.51 ± 19.29	33.32 ± 21.76	11.95 ± 0.82
NA12	69.43 ± 13.35	166.82 ± 44.05	270.18 ± 48.23	113.90 ± 22.60	176.07 ± 10.74	100.96 ± 11.88	131.22 ± 26.23	12.73 ± 2.43	<LOD
NA13	767.89 ± 139.78	1295.53 ± 158.25	1838.17 ± 237.71	354.76 ± 36.39	88.73 ± 53.76	230.87 ± 56.38	28.92 ± 20.64	13.25 ± 5.60	<LOQ
NA14	159.14 ± 10.16	154.95 ± 11.65	562.06 ± 17.70	166.63 ± 27.68	80.54 ± 9.86	92.82 ± 7.92	37.25 ± 12.06	33.23 ± 13.00	19.45 ± 9.18
NA15	72.71 ± 13.23	43.68 ± 18.20	118.64 ± 9.41	20.62 ± 4.20	13.65 ± 2.88	<LOQ	<LOQ	<LOD	<LOD
NA16	69.41 ± 0.95	27.38 ± 2.41	22.10 ± 2.73	14.84 ± 3.98	4.67 ± 0.65	5.28 ± 2.55	<LOD	1.14 ± 0.10	<LOQ
NA17	428.07 ± 91.70	392.25 ± 45.32	1248.92 ± 132.52	758.50 ± 96.82	407.68 ± 62.61	490.03 ± 85.84	362.85 ± 87.15	148.48 ± 35.57	<LOQ
NA18	50.82 ± 12.88	20.57 ± 4.06	59.09 ± 5.95	15.41 ± 4.13	14.69 ± 1.95	9.28 ± 3.81	<LOQ	<LOD	<LOD
NA19	241.33 ± 18.37	95.53 ± 12.75	112.40 ± 42.74	27.85 ± 5.08	10.21 ± 0.82	8.81 ± 0.43	<LOD	<LOD	<LOD
NA20	1986.51 ± 222.58	482.94 ± 68.74	961.35 ± 93.20	79.32 ± 34.38	9.56 ± 1.23	11.70 ± 1.37	<LOQ	<LOD	<LOD
NA21	98.27 ± 12.34	81.60 ± 24.97	430.40 ± 162.73	366.55 ± 194.99	200.05 ± 81.39	274.18 ± 237.77	88.95 ± 72.45	11.71 ± 3.49	5.91 ± 1.06
NA22	32.27 ± 3.21	41.12 ± 10.90	94.04 ± 42.90	47.84 ± 8.13	56.82 ± 23.64	18.88 ± 2.28	21.17 ± 13.27	2.06 ± 0.25	<LOQ
NA23	167.04 ± 13.93	76.70 ± 30.24	187.18 ± 12.30	83.26 ± 16.24	21.73 ± 6.41	38.75 ± 8.78	<LOQ	<LOD	1.34 ± 0.31
NA24	311.67 ± 103.86	345.59 ± 122.98	363.30 ± 49.87	240.66 ± 27.09	51.22 ± 3.98	121.34 ± 26.16	20.74 ± 9.69	<LOD	<LOQ
NA25	287.37 ± 50.77	99.36 ± 8.24	246.53 ± 135.51	71.78 ± 14.11	49.62 ± 15.45	67.52 ± 12.29	27.35 ± 7.98	7.54 ± 5.72	<LOQ
NA26	301.45 ± 7.18	203.75 ± 8.34	201.71 ± 32.39	133.89 ± 22.28	47.58 ± 10.12	44.18 ± 4.73	14.43 ± 1.89	<LOD	<LOD
NA27	181.84 ± 39.43	99.68 ± 31.31	210.44 ± 23.01	101.31 ± 30.20	32.10 ± 4.21	38.99 ± 9.99	10.76 ± 2.02	<LOD	<LOD
NA28	257.12 ± 50.04	230.39 ± 40.84	713.68 ± 136.91	789.07 ± 147.53	820.20 ± 40.21	429.43 ± 39.75	281.07 ± 9.15	38.80 ± 9.16	32.98 ± 2.42
NA29	413.11 ± 35.94	307.46 ± 14.42	973.03 ± 65.71	1002.42 ± 127.57	842.34 ± 89.81	1507.74 ± 132.48	793.43 ± 207.73	136.75 ± 3.22	102.29 ± 2.42
NA30	684.92 ± 88.51	540.56 ± 133.08	836.67 ± 104.37	1061.13 ± 78.24	961.71 ± 42.95	451.47 ± 31.55	266.14 ± 38.95	45.30 ± 8.11	56.77 ± 4.08
NA31	229.05 ± 42.43	275.28 ± 49.94	561.45 ± 105.34	590.57 ± 84.97	880.43 ± 19.43	442.69 ± 30.62	263.85 ± 30.81	82.87 ± 2.44	177.29 ± 8.86
NA32	36.31 ± 5.73	16.91 ± 4.38	136.46 ± 21.55	78.11 ± 7.57	29.83 ± 3.28	27.73 ± 4.47	15.84 ± 2.27	<LOD	<LOD
NA33	375.11 ± 51.98	311.24 ± 58.11	989.98 ± 141.36	90.50 ± 27.25	84.27 ± 10.45	84.69 ± 5.11	13.15 ± 3.90	8.53 ± 2.47	3.47 ± 2.21

412 Table S14: Summary of PFSA concentrations in North American samples.

Sample ID	PFHxS (pg/g)	PFOS (pg/g)	PFDS (pg/g)
NA01	3.78 ± 0.08	303.65 ± 7.77	7.70 ± 0.92
NA02	30.34 ± 0.98	1956.34 ± 133.28	<LOD
NA03	4.30 ± 0.61	112.3 ± 4.57	22.71 ± 2.47
NA04	13.02 ± 1.05	390.74 ± 14.67	<LOD
NA05	6.93 ± 0.21	326.92 ± 11.72	<LOD
NA06	6.10 ± 0.25	116.84 ± 11.30	127.23 ± 12.20
NA07	4.93 ± 0.60	30.05 ± 0.54	<LOD
NA08	6.33 ± 0.70	110.59 ± 6.98	<LOD
NA09	21.43 ± 0.64	574.76 ± 29.46	<LOD
NA10	12.39 ± 0.68	168.93 ± 3.64	<LOD
NA11	15.54 ± 3.05	561.46 ± 96.96	<LOD
NA12	5.66 ± 0.25	657.22 ± 164.56	4.14 ± 2.61
NA13	3.35 ± 0.98	63.04 ± 6.84	<LOD
NA14	34.31 ± 0.32	700.31 ± 26.64	11.69 ± 1.13
NA15	<LOQ	18.09 ± 1.34	<LOD
NA16	4.19 ± 0.12	34.89 ± 2.87	<LOD
NA17	34.31 ± 0.08	1809.01 ± 42.43	<LOD
NA18	<LOQ	71.80 ± 12.81	<LOD
NA19	4.38 ± 0.36	35.83 ± 1.26	<LOD
NA20	7.10 ± 0.56	158.11 ± 3.22	<LOD
NA21	18.96 ± 3.75	684.57 ± 112.69	<LOD
NA22	2.93 ± 0.10	109.99 ± 4.17	<LOD
NA23	5.08 ± 0.91	149.01 ± 5.08	<LOD
NA24	31.33 ± 1.34	350.26 ± 25.83	<LOD
NA25	12.29 ± 3.11	148.93 ± 7.53	12.08 ± 1.51
NA26	8.60 ± 0.06	226.13 ± 9.51	<LOD
NA27	7.54 ± 0.55	182.66 ± 11.94	<LOD
NA28	3.78 ± 0.17	268.22 ± 2.76	2.94 ± 0.68
NA29	<LOD	309.70 ± 32.87	<LOD
NA30	2.02 ± 1.06	302.05 ± 2.25	3.74 ± 0.15
NA31	<LOQ	184.59 ± 3.15	1.23 ± 1.09
NA32	36.51 ± 3.79	689.06 ± 33.69	16.79 ± 0.85
NA33	31.74 ± 1.53	1145.50 ± 69.35	<LOD

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415 Table S15: Summary of FTCA concentrations in North American samples.

Sample ID	5:3 FTCA (pg/g)	7:3 FTCA (pg/g)	9:3 FTCA (pg/g)	11:3 FTCA (pg/g)	13:3 FTCA (pg/g)	6:2 FTCA (pg/g)	8:2 FTCA (pg/g)	10:2 FTCA (pg/g)	12:2 FTCA (pg/g)	14:2 FTCA (pg/g)
NA01	<LOQ	0.55 ± 0.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA02	20.16 ± 5.83	23.84 ± 2.96	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA04	1.64 ± 0.70	4.37 ± 0.78	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA05	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA06	<LOD	1.75 ± 0.90	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA07	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA08	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA09	<LOD	1.72 ± 0.67	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA10	<LOQ	0.71 ± 0.56	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA11	<LOD	MDL	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA12	<LOD	1.89 ± 0.76	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA13	8.86 ± 4.23	32.07 ± 3.52	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA14	<LOQ	1.17 ± 1.08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA16	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA17	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA18	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA20	3.65 ± 0.91	9.91 ± 1.68	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA21	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA22	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA23	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA24	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA25	<LOQ	1.02 ± 0.74	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA26	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA28	4.64 ± 1.14	7.58 ± 1.32	5.11 ± 1.69	1.51 ± 0.23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA29	11.34 ± 1.93	18.23 ± 2.88	14.61 ± 6.49	4.33 ± 00.33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA30	4.53 ± 0.92	15.03 ± 1.73	10.50 ± 1.83	3.43 ± 0.80	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA31	4.11 ± 2.60	12.21 ± 3.91	6.10 ± 1.45	3.55 ± 0.27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA32	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA33	11.43 ± 5.33	24.81 ± 9.53	1.01 ± 0.51	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

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Table S16: Summary of FTUCA concentrations in North American samples.

Sample ID	5:3 FTUCA (pg/g)	7:3 FTUCA (pg/g)	9:3 FTUCA (pg/g)	11:3 FTUCA (pg/g)	13:3 FTUCA (pg/g)	6:2 FTUCA (pg/g)	8:2 FTUCA (pg/g)	10:2 FTUCA (pg/g)	12:2 FTUCA (pg/g)	14:2 FTUCA (pg/g)
NA01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
NA02	<LOD	<LOD	<LOD	<LOD	<LOD	95.03 ± 17.57	10.52 ± 3.96	<LOD	<LOQ	<LOD
NA03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA04	<LOD	<LOD	<LOD	<LOD	<LOD	2.42 ± 2.07	<LOD	<LOD	<LOD	<LOD
NA05	<LOD	<LOD	<LOD	<LOD	<LOD	0.27 ± 0.12	<LOD	<LOD	<LOD	<LOD
NA06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
NA11	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA12	<LOD	<LOD	<LOD	<LOD	<LOD	1.44 ± 1.42	<LOD	<LOD	<LOD	<LOD
NA13	0.40 ± 0.35	0.62 ± 0.37	<LOD	<LOD	<LOD	32.98 ± 9.93	17.78 ± 2.63	1.44 ± 0.88	<LOD	<LOD
NA14	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA16	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA17	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA18	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA19	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA20	<LOD	<LOD	<LOD	<LOD	<LOD	4.04 ± 1.67	4.64 ± 1.36	<LOD	<LOD	<LOD
NA21	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.74 ± 1.67	<LOD	<LOD	<LOD
NA22	<LOQ	<LOD	<LOD	<LOD	<LOD	0.61 ± 0.35	<LOD	<LOD	<LOD	<LOD
NA23	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA24	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOQ	<LOD	<LOD	<LOD
NA25	<LOD	<LOD	<LOD	<LOD	<LOD	20.28 ± 7.47	<LOD	<LOD	<LOD	<LOD
NA26	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
NA28	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA29	0.40 ± 0.29	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA30	<LOD	0.51 ± 0.43	<LOD	<LOD	<LOD	0.89 ± 0.47	<LOD	<LOD	<LOD	<LOD
NA31	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD
NA32	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
NA33	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD

Table S17: Summary of PFCA concentrations in European, Asian and African samples.

Sample ID	PFHxA (pg/g)	PFHpA (pg/g)	PFOA (pg/g)	PFNA (pg/g)	PFDA (pg/g)	PFUnDA (pg/g)	PFDoDA (pg/g)	PFTrDA (pg/g)	PFTeDA (pg/g)
EU01	142.76 ± 14.12	200.96 ± 14.40	1086.42 ± 222.73	216.94 ± 38.45	579.78 ± 360.33	84.54 ± 4.82	103.98 ± 15.91	9.77 ± 4.04	18.94 ± 6.12
EU02	51.66 ± 7.15	40.24 ± 13.26	142.47 ± 22.99	15.81 ± 5.92	19.70 ± 6.10	10.24 ± 2.60	13.77 ± 8.00	1.34 ± 0.54	4.94 ± 3.48
EU03	248.44 ± 37.33	377.62 ± 78.62	2674.04 ± 185.08	179.41 ± 15.73	70.05 ± 17.54	61.04 ± 16.47	32.38 ± 12.80	<LOQ	<LOD
EU04	188.33 ± 34.07	309.60 ± 64.57	792.22 ± 123.17	216.63 ± 15.95	84.02 ± 20.97	58.60 ± 4.60	33.66 ± 3.14	<LOQ	<LOD
EU05	91.98 ± 5.77	163.32 ± 26.82	558.07 ± 21.72	114.32 ± 13.36	104.32 ± 4.99	39.73 ± 3.00	34.10 ± 2.25	<LOD	<LOD
EU06	24.38 ± 2.57	23.92 ± 9.27	20.38 ± 2.89	9.13 ± 1.08	<LOD	<LOD	<LOD	<LOD	<LOD
EU07	50.46 ± 9.76	73.94 ± 25.77	62.29 ± 5.24	98.55 ± 17.76	12.87 ± 1.84	19.78 ± 1.21	9.65 ± 2.35	<LOD	<LOD
EU08	21.68 ± 6.48	9.67 ± 2.52	42.57 ± 6.71	25.80 ± 8.05	8.76 ± 1.18	7.18 ± 0.91	9.84 ± 0.57	<LOD	<LOD
EU09	19.78 ± 6.35	8.32 ± 4.76	15.17 ± 5.18	<LOQ	7.56 ± 2.38	3.72 ± 0.54	<LOD	<LOD	<LOD
EU10	52.86 ± 8.49	47.54 ± 14.50	120.07 ± 5.62	13.03 ± 2.92	7.29 ± 3.59	8.45 ± 7.44	<LOQ	2.85 ± 1.84	<LOD
AS01	892.50 ± 161.66	3748.43 ± 907.20	1492.37 ± 169.75	667.84 ± 21.73	148.53 ± 42.89	1005.62 ± 140.21	59.06 ± 8.29	29.95 ± 3.93	16.13 ± 2.09
AS02	269.34 ± 28.48	331.72 ± 33.28	708.25 ± 28.84	297.53 ± 39.62	89.84 ± 13.13	156.71 ± 19.17	42.43 ± 15.84	13.34 ± 8.64	<LOQ
AS03	265.48 ± 9.73	323.64 ± 67.83	1798.66 ± 106.43	720.56 ± 169.59	224.50 ± 24.09	232.65 ± 30.31	100.77 ± 47.71	80.82 ± 57.93	<LOQ
AS04	7544.43 ± 187.72	2607.62 ± 190.24	3436.01 ± 158.24	362.91 ± 38.97	128.63 ± 28.75	145.21 ± 15.02	47.88 ± 12.86	21.59 ± 3.84	<LOQ
AS05	34.62 ± 1.17	39.29 ± 5.28	37.44 ± 9.68	12.86 ± 3.98	<LOQ	4.57 ± 0.38	<LOD	<LOD	<LOD
AS06	25.04 ± 4.23	34.74 ± 12.19	57.33 ± 9.40	18.77 ± 3.27	13.00 ± 0.54	9.81 ± 1.79	<LOQ	4.22 ± 2.32	3.97 ± 1.14
AF01	83.43 ± 9.73	154.82 ± 41.28	128.03 ± 40.96	40.12 ± 3.51	29.08 ± 6.98	5.93 ± 0.42	25.79 ± 6.50	2.37 ± 0.77	1.53 ± 0.17
AF02	85.75 ± 9.33	16.73 ± 4.66	14.22 ± 3.07	3.37 ± 1.5	2.58 ± 1.42	1.04 ± 0.56	<LOD	<LOD	<LOD
AF03	219.30 ± 19.56	74.58 ± 17.67	160.11 ± 24.00	11.96 ± 1.90	5.87 ± 1.88	33.88 ± 8.26	<LOD	2.58 ± 0.64	<LOD
AF04	873.80 ± 36.92	313.48 ± 18.25	242.28 ± 35.56	18.59 ± 5.12	37.85 ± 2.73	8.23 ± 2.99	<LOQ	<LOQ	<LOD
AF05	17.14 ± 3.11	81.88 ± 19.30	23.19 ± 2.40	6.08 ± 1.23	5.46 ± 3.00	9.95 ± 3.04	<LOQ	<LOD	<LOQ

436 Table S18: Summary of PFSA concentrations in European, Asian and African samples.

Sample ID	PFHxS (pg/g)	PFOS (pg/g)	PFDS (pg/g)
EU01	99.73 ± 3.78	3130.08 ± 51.64	38.71 ± 2.44
EU02	<LOQ	20.34 ± 0.64	<LOD
EU03	43.19 ± 3.00	719.18 ± 18.73	<LOD
EU04	45.14 ± 2.59	2159.28 ± 88.72	53.86 ± 4.42
EU05	22.66 ± 0.49	1579.96 ± 8.51	<LOD
EU06	<LOQ	7.06 ± 1.04	<LOD
EU07	4.35 ± 0.14	83.65 ± 4.05	<LOD
EU08	3.27 ± 0.58	36.34 ± 3.26	<LOD
EU09	<LOD	<LOQ	<LOD
EU10	<LOQ	29.69 ± 1.53	3.59 ± 0.26
AS01	5.82 ± 0.27	145.65 ± 9.97	<LOD
AS02	8.76 ± 0.22	184.19 ± 7.91	2.22 ± 0.49
AS03	14.63 ± 1.94	406.05 ± 28.64	<LOD
AS04	6.84 ± 1.03	87.06 ± 3.21	<LOD
AS05	2.95 ± 0.56	74.07 ± 2.61	1.81 ± 0.49
AS06	4.83 ± 0.76	153.90 ± 5.17	<LOQ
AF01	8.91 ± 0.27	135.02 ± 3.86	<LOD
AF02	<LOQ	72.34 ± 1.00	<LOD
AF03	<LOD	14.22 ± 0.75	<LOD
AF04	<LOQ	90.59 ± 3.42	<LOD
AF05	<LOQ	16.64 ± 0.84	<LOD

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Table S19: Summary of FTCA concentrations in European, Asian and African samples.

Sample ID	5:3 FTCA (pg/g)	7:3 FTCA (pg/g)	9:3 FTCA (pg/g)	11:3 FTCA (pg/g)	13:3 FTCA (pg/g)	6:2 FTCA (pg/g)	8:2 FTCA (pg/g)	10:2 FTCA (pg/g)	12:2 FTCA (pg/g)	14:2 FTCA (pg/g)
EU01	0.74 ± 0.65	1.62 ± 1.02	1.05 ± 0.32	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU02	<LOQ	0.92 ± 0.15	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU04	<LOD	1.22 ± 0.27	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU05	1.57 ± 0.17	2.26 ± 0.69	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU09	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU10	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS01	5.30 ± 2.81	25.79 ± 4.20	0.72 ± 0.07	0.49 ± 0.16	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS02	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS04	6.55 ± 2.88	19.65 ± 0.43	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS06	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF03	1.58 ± 0.58	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF04	<LOQ	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

458 Table S20: Summary of FTUCA concentrations in European, Asian and African samples.

Sample ID	5:3 FTUCA (pg/g)	7:3 FTUCA (pg/g)	9:3 FTUCA (pg/g)	11:3 FTUCA (pg/g)	13:3 FTUCA (pg/g)	6:2 FTUCA (pg/g)	8:2 FTUCA (pg/g)	10:2 FTUCA (pg/g)	12:2 FTUCA (pg/g)	14:2 FTUCA (pg/g)
EU01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD
EU04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU06	<LOQ	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD
EU07	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
EU08	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
EU10	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AS01	<LOD	<LOQ	<LOD	<LOD	<LOD	0.74 ± 0.39	5.83 ± 0.01	4.06 ± 0.45	<LOD	<LOD
AS02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
AS03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
AS04	0.50 ± 0.43	2.79 ± 0.77	<LOD	<LOD	<LOD	1.26 ± 0.56	42.91 ± 8.94	5.63 ± 2.79	<LOD	<LOD
AS05	<LOD	<LOD	<LOD	<LOD	<LOD	0.47 ± 0.38	<LOD	<LOD	<LOD	<LOD
AS06	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AF01	3.11 ± 1.85	<LOD	<LOD	<LOD	<LOD	0.47 ± 0.15	2.50 ± 0.54	<LOD	<LOD	<LOD
AF02	<LOD	<LOD	<LOD	<LOD	<LOD	4.82 ± 0.25	<LOD	<LOD	<LOD	<LOD
AF03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
AF04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
AF05	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

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461 Table S21: Summary of PFCA concentrations in Australian, South American and Antarctic

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Sample ID	PFHxA (pg/g)	PFHpA (pg/g)	PFOA (pg/g)	PFNA (pg/g)	PFDA (pg/g)	PFUnDA (pg/g)	PFDoDA (pg/g)	PFTrDA (pg/g)	PFTeDA (pg/g)
AU01	39.59 ± 3.82	46.34 ± 12.80	89.02 ± 21.75	18.31 ± 14.91	25.31 ± 21.89	11.06 ± 2.50	<LOD	<LOD	<LOD
AU02	214.64 ± 11.91	95.05 ± 32.15	490.50 ± 161.59	46.79 ± 16.53	238.96 ± 122.12	37.92 ± 17.49	<LOQ	<LOD	<LOQ
AU03	27.35 ± 6.45	6.13 ± 2.33	27.61 ± 18.23	<LOQ	15.66 ± 9.54	2.43 ± 0.35	<LOQ	<LOQ	<LOQ
AU04	17.83 ± 2.85	1155.93 ± 236.89	39.59 ± 9.01	8.33 ± 4.67	9.13 ± 10.84	28.77 ± 7.06	<LOD	<LOD	<LOD
SA01	154.22 ± 31.43	75.85 ± 8.55	67.33 ± 19.96	12.14 ± 2.31	5.61 ± 0.08	4.13 ± 1.39	<LOD	<LOQ	<LOD
SA02	15.89 ± 5.64	<LOQ	43.94 ± 15.33	5.25 ± 3.07	<LOQ	<LOQ	<LOQ	<LOD	<LOD
SA03	4.47 ± 0.10	<LOQ	19.96 ± 3.67	4.28 ± 2.01	<LOQ	<LOQ	<LOD	<LOQ	<LOD
AN01	98.22 ± 60.28	44.84 ± 16.26	48.14 ± 12.74	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

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Table S22: Summary of PFSA concentrations in Australian, South American and Antarctic samples.

Sample ID	PFHxS (pg/g)	PFOS (pg/g)	PFDS (pg/g)
AU01	39.57 ± 1.44	257.64 ± 35.27	<LOD
AU02	21.04 ± 1.50	175.18 ± 4.03	<LOD
AU03	4.90 ± 0.36	74.12 ± 4.71	<LOD
AU04	<LOD	43.73 ± 5.07	<LOD
SA01	<LOD	35.64 ± 1.21	<LOD
SA02	<LOD	25.57 ± 1.80	<LOD
SA03	<LOD	48.07 ± 1.74	<LOD
AN01	<LOD	6.64 ± 0.89	0.56 ± 0.14

Table S23: Summary of FTCA concentrations in Australian, South American and Antarctic samples.

Sample ID	5:3 FTCA (pg/g)	7:3 FTCA (pg/g)	9:3 FTCA (pg/g)	11:3 FTCA (pg/g)	13:3 FTCA (pg/g)	6:2 FTCA (pg/g)	8:2 FTCA (pg/g)	10:2 FTCA (pg/g)	12:2 FTCA (pg/g)	14:2 FTCA (pg/g)
AU01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AU02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AU03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AU04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
SA01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
SA02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
SA03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AN01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD

Table S24: Summary of FTUCA concentrations in Australian, South American and Antarctic samples.

Sample ID	5:3 FTUCA (pg/g)	7:3 FTUCA (pg/g)	9:3 FTUCA (pg/g)	11:3 FTUCA (pg/g)	13:3 FTUCA (pg/g)	6:2 FTUCA (pg/g)	8:2 FTUCA (pg/g)	10:2 FTUCA (pg/g)	12:2 FTUCA (pg/g)	14:2 FTUCA (pg/g)
AU01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AU02	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AU03	<LOD	<LOD	<LOD	<LOD	<LOD	0.54 ± 0.20	<LOD	<LOD	<LOD	<LOD
AU04	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
SA01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOQ	<LOD	<LOD	<LOD	<LOD
SA02	<LOD	<LOD	<LOD	<LOD	<LOD	LOQ	<LOD	<LOD	<LOD	<LOD
SA03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
AN01	<LOD	<LOD	<LOD	<LOD	<LOD	0.66 ± 0.27	<LOD	<LOD	<LOD	<LOD

Table S25: Qualitative percentage of linear and branched PFOA and PFOS isomers in North American samples.

Sample ID	Linear PFOA (%)	Branched PFOA (%)	Linear PFOS (%)	Branched PFOS (%)
NA01	73.4	26.6	89.9	10.1
NA02	75.0	25.0	73.2	26.8
NA03	49.5	50.5	75.0	25.0
NA04	76.5	23.5	81.9	18.1
NA05	68.2	31.8	82.8	17.2
NA06	86.6	13.4	83.2	16.8
NA07	55.4	44.6	73.7	26.3
NA08	72.3	27.7	78.2	21.8
NA09	100.0	0.0	75.2	24.8
NA10	100.0	0.0	80.2	19.8
NA11	67.5	32.5	84.7	15.3
NA12	97.6	2.4	78.2	21.8
NA13	83.9	16.1	81.3	18.7
NA14	76.8	23.2	79.9	20.1
NA15	59.7	40.3	81.9	18.1
NA16	42.6	57.4	70.1	29.9
NA17	52.2	47.8	78.5	21.5
NA18	23.5	76.5	75.0	25.0
NA19	50.6	49.4	62.8	37.2
NA20	ND	ND	ND	ND
NA21	61.0	39.0	91.9	8.1
NA22	77.5	22.5	81.0	19.0
NA23	71.0	29.0	81.3	18.7
NA24	71.4	28.6	25.6	74.4
NA25	46.2	53.8	85.0	15.0
NA26	83.5	16.5	84.7	15.3
NA27	60.2	39.8	82.9	17.1
NA28	77.2	22.8	82.3	17.7
NA29	100.0	0.0	93.4	6.6
NA30	80.5	19.5	87.0	13.0
NA31	78.5	21.5	87.5	12.5
NA32	26.0	74.0	74.3	25.7
NA33	100.0	0.0	94.7	5.3

Table S26: Qualitative percentage of linear and branched PFOA and PFOS isomers in European, Asian and African samples.

Sample ID	Linear PFOA (%)	Branched PFOA (%)	Linear PFOS (%)	Branched PFOS (%)
EU01	100.0	0.0	76.4	23.6
EU02	44.3	55.7	63.5	36.5
EU03	84.9	15.1	68.5	31.5
EU04	100.0	0.0	82.6	17.4
EU05	100.0	0.0	84.6	15.4
EU06	40.6	59.4	53.0	47.0
EU07	51.8	48.2	68.9	31.1
EU08	69.6	30.4	70.3	29.7
EU09	56.6	43.4	ND	ND
EU10	57.1	42.9	69.6	30.4
AS01	100.0	0.0	76.5	23.5
AS02	93.5	6.5	88.9	11.1
AS03	69.9	30.1	74.4	25.6
AS04	87.7	12.3	75.0	25.0
AS05	52.2	47.8	75.9	24.1
AS06	75.2	24.8	73.9	26.1
AF01	73.8	26.2	74.9	25.1
AF02	31.6	68.4	69.1	30.9
AF03	70.7	29.3	82.7	17.3
AF04	74.7	25.3	71.7	28.3
AF05	37.9	62.1	72.3	27.7

Table S27: Qualitative percentage of linear and branched PFOA and PFOS isomers in Australian, South American and Antarctic samples.

Sample ID	Linear PFOA (%)	Branched PFOA (%)	Linear PFOS (%)	Branched PFOS (%)
AU01	53.5	46.5	74.8	25.2
AU02	ND	ND	64.4	35.6
AU03	66.2	33.8	83.1	16.9
AU04	53.8	46.2	78.7	21.3
SA01	47.7	52.3	83.2	16.8
SA02	51.7	48.3	66.1	33.9
SA03	36.9	63.1	76.1	23.9
AN01	71.5	28.5	69.7	30.3

510 Table S28: PFCA and PFSA factor loading values.

Analyte	Factor 1	Factor 2
PFHxA	0.255006	0.571649
PFHpA	0.388052	0.503163
PFOA	0.556528	0.692889
PFNA	0.923993	-0.021150
PFDA	0.874563	-0.191004
PFUnDA	0.875754	-0.150681
PFDoDA	0.898755	-0.273528
PFTrDA	0.875904	-0.172598
PFTeDA	0.703120	-0.422746
PFHxS	0.170856	0.614007
PFOS	0.315844	0.537564
PFDS	0.036601	0.299286

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