

SUPPORTING INFORMATION

(22 PAGES, 4 TABLES, 1 FIGURE)

Analysis of a Homologous Series of Perfluorocarboxylates

from American Red Cross Adult Blood Donors, 2000 - 2001 and 2006

**Geary W. Olsen Mark E. Ellefson, David C. Mair, Timothy R. Church,
Corinne L. Goldberg, Ross M. Herron, Zahra Medhdizadehkashi, John B. Nobiletti,
Jorge A. Rios, William K. Reagen, Larry R. Zobel**

Sample Preparation. The 3M Environmental Laboratory prepared and analyzed the samples. Target analytes were extracted from serum or plasma by protein precipitation in acetonitrile (8) via a MultiPROBE II HT EX robotic liquid handling system (PerkinElmer, Wellesley, MA). Using a disposable transfer pipette, 200 μ L of serum/plasma samples were transferred to 1.5 mL centrifuge tubes. Each sample was spiked with 25 μ L of acetonitrile that contained surrogate recovery standards and internal standards. Quality control samples (consisting of 200 μ L of control human plasma or control bovine serum) were spiked with 25 μ L of acetonitrile containing surrogate recovery standards, internal standards, and all target analytes. After spiking, all the samples were placed on the MultiPROBE II and mixed by aspirating and dispensing 100 μ L of the sample three times. Following aspiration, 775 μ L of acetonitrile was added to each sample. Samples were vortex mixed and placed in a centrifuge for 20 minutes at 10,000 rpm. After centrifugation, 60 μ L of an aqueous 5% phosphoric acid solution was added to each autovial and the extracts were transferred to them for analysis. A new surrogate matrix-matched bovine calibration curve and full set of quality control samples and blanks were prepared and analyzed with every batch of samples.

HPLC/MS/MS Analysis. Quantitation of sample extracts was accomplished by high performance liquid chromatography tandem mass spectrometry (HPLC/MS/MS) using an Agilent Series 1200 Liquid Chromatograph system (Agilent Technologies, Santa Clara, CA) attached to an Applied Biosystems MDS Sciex API 5000 triple-quadrupole mass spectrometer equipped with a TurboIon Spray Source (PE Sciex, Concord, Ontario, Canada) maintained at 450 °C in the negative ion mode. The target analytes, internal standards, and surrogate recovery standards were monitored in two separate runs recording one to three transitions for each ion to improve sensitivity (9).

For PFBA, PFPeA, PFHxA, PFHpA, MPFBA, MPFHxA, and MPFOA the analytical column was a PrismTM RP 2x50mm, 5 μ m particle size (Thermo Electron, Franklin, MA). A PrismTM RP 2x50mm, 5 μ m particle size guard column was inserted in the HPLC between the pump and the autosampler to provide separation of any non-specific background fluorochemicals originating from the PTFE instrument components

or the mobile phase. The injection volume was set at 20 μ L with a flow rate of 0.3 mL/min. and a column temperature maintained at 30° C. The aqueous mobile phase consisted of 5mM ammonium acetate in 0.01% acetic acid and methanol was used as the organic solvent phase. The chromatographic elution gradient was programmed to hold at 10% methanol (b) for three minutes, followed by a five minute linear gradient to 95% (b), held at 95% (b) for six minutes, returned to 10% (b) in one minute, and equilibrated at 10% (b) for an additional four minutes.

For PFNA, PFDA, PFUnA, PFDoA, MPFOA, MPFDA, and MPFDoA the analytical column was a BetasilTM C18 2.1x100mm, 5 μ m particle size (Thermo Electron, Franklin, MA). A PrismTM RP 2x50mm, 5 μ m particle size guard column was inserted in the HPLC between the pump and the autosampler to provide separation of any non-specific background fluorochemicals originating from the PTFE instrument components or the mobile phase. The injection volume was set at 50 μ L with a flow rate of 0.3 mL/min. and a column temperature maintained at 30° C. An Oasis HLB Online column, 3x20 mm, 25 μ m particle size (Waters Corporation, Milford, MA) was positioned just prior to the analytical column to minimize the solvent effect of a 50 μ L injection volume. The aqueous mobile phase consisted of 2mM ammonium acetate and acetonitrile was used as the organic solvent phase. The chromatographic elution gradient was programmed to hold at 5% acetonitrile (b) for one minute, followed by a ten minute linear gradient to 90% (b), held at 90% (b) for three and one half minutes, returned to 5% (b) in one half minute, and equilibrated at 5% (b) for an additional three minutes.

The ions monitored to quantitate the target analytes are listed in TABLE S1 of the Supporting Information.

TABLE S1. Ions Used for Quantitation, American Red Cross Plasma Samples

Compound	Parent	Product	Dwell (ms)	Declustering	Collision	Collision Cell
				Potential (DP) (V)	Energy (CE) (V)	Exit Potential (CXP) (V)
PFBA	213	169	80	-40	-7	-7
PFPeA	263	219	80	-30	-12	-15
PFHxA	313	119	80	-45	-28	-17
PFHpA	363	319	80	-35	-14	-17
	363	169	80	-35	-24	-17
PFNA	463	419	50	-45	-15	-10
	463	219	50	-40	-15	-10
	463	169	50	-40	-20	-10
PFDA	513	469	50	-40	-16	-9
	513	219	50	-45	-20	-15

TABLE S1. (continued)

Compound	Parent	Product	Dwell (ms)	Declustering	Collision	Collision Cell
				Potential (DP) (V)	Energy (CE) (V)	Exit Potential (CXP) (V)
PFUnA	563	519	50	-45	-20	-16
	563	269	50	-45	-20	-16
PFDoA	613	569	50	-45	-20	-16
	613	219	50	-45	-20	-10
	613	169	50	-45	-20	-10
MPFBA	217	172	80	-45	-20	-11
MPFH _x A	315	119	80	-45	-20	-15
MPFOA	417	372	50	-35	-14	-7
MPFDA	515	470	50	-65	-14	-16
MPFDoA	615	570	50	-40	-15	-16

TABLE S2. Geometric Means (95 % Confidence Intervals) for PFBA, PFHpA, PFOA, PFNA, PFDA, and PFUnA by Age and Sex, American Red Cross Blood Donors, 2000 - 2001 and 2006

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFBA				
2000 - 2001						
all		2.74 (2.51 – 2.99)	2.54 (2.31 – 2.79)	2.66 (2.47 – 2.87)	2.62 (2.38 – 2.89)	2.49 (2.31 – 2.68)
males		2.87 (2.54 – 3.25)	2.60 (2.35 – 2.87)	2.98 (2.36 – 2.98)	2.70 (2.30 – 3.18)	2.53 (2.29 – 2.78)
females		2.61 (2.30 – 2.98)	2.49 (2.12 – 2.92)	2.68 (2.43 – 2.96)	2.53 (2.27 – 2.83)	2.44 (2.16 – 2.75)
2006						
all		0.28 (0.22 – 0.35)	0.32 (0.26 – 0.39)	0.34 (0.28 – 0.42)	0.35 (0.28 – 0.42)	0.37 (0.31 – 0.44)
males		0.32 (0.23 – 0.44)	0.32 (0.24 – 0.42)	0.37 (0.27 – 0.49)	0.32 (0.24 – 0.42)	0.32 (0.25 – 0.42)
females		0.25 (0.18 – 0.34)	0.32 (0.24 – 0.43)	0.32 (0.24 – 0.42)	0.38 (0.28 – 0.51)	0.42 (0.32 – 0.54)

TABLE S2. (continued)

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFHpA				
2000 - 2001						
all		0.14 (0.12 – 0.16)	0.12 (0.11 – 0.13)	0.11 (0.10 – 0.13)	0.12 (0.11 – 0.14)	0.14 (0.12 – 0.15)
males		0.12 (0.10 – 0.14)	0.12 (0.10 – 0.14)	0.12 (0.10 – 0.14)	0.13 (0.11 – 0.16)	0.14 (0.12 – 0.17)
females		0.15 (0.13 – 0.19)	0.12 (0.10 – 0.14)	0.11 (0.09 – 0.13)	0.12 (0.10 – 0.14)	0.13 (0.11 – 0.16)
2006						
all		0.11 (0.09 – 0.13)	0.08 (0.07 – 0.09)	0.07 (0.06 – 0.08)	0.09 (0.07 – 0.10)	0.09 (0.08 – 0.10)
males		0.11 (0.09 – 0.14)	0.09 (0.08 – 0.12)	0.08 (0.06 – 0.10)	0.09 (0.07 – 0.11)	0.10 (0.08 – 0.12)
females		0.11 (0.09 – 0.14)	0.07 (0.06 – 0.09)	0.06 (0.05 – 0.08)	0.08 (0.07 – 0.10)	0.08 (0.07 – 0.10)

TABLE S2. (continued)

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFOA				
2000 - 2001						
all		5.08 (4.61 – 5.59)	4.54 (4.09 – 5.04)	4.63 (4.21 – 5.10)	4.65 (4.24 – 5.11)	4.64 (4.16 – 5.18)
males		5.93 (5.29 – 6.65)	4.91 (4.21 – 5.73)	4.78 (4.17 – 5.46)	4.72 (4.17 – 5.35)	4.88 (4.21 – 5.65)
females		4.34 (3.74 (5.04)	4.21 (3.65 -4.85)	4.49 (3.91 – 5.15)	4.57 (3.96 – 5.28)	4.37 (3.69 – 5.17)
2006						
all		3.75 (3.47 – 4.05)	3.78 (3.44 – 4.15)	3.21 (2.91 – 3.54)	3.46 (3.12 – 3.83)	3.07 (2.81 – 3.35)
males		4.41 (4.02 – 4.85)	4.73 (4.24 – 5.280	3.65 (3.19 – 4.180	3.66 (3.21 – 4.18)	3.43 (3.03 – 3.87)
females		3.19 (2.85 – 3.56)	3.02 (2.65 – 3.44)	2.82 (2.46 – 3.23)	3.27 (2.79 – 3.83)	2.75 (2.43 – 3.12)

TABLE S2. (continued)

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFNA				
2000 - 2001						
all		0.53 (0.48 – 0.57)	0.60 (0.55 – 0.66)	0.59 (0.54 – 0.64)	0.57 (0.52 – 0.63)	0.59 (0.53 – 0.64)
males		0.60 (0.53 – 0.67)	0.66 (0.58 – 0.76)	0.64 (0.57 – 0.72)	0.69 (0.54 – 0.69)	0.61 (0.54 – 0.69)
females		0.46 (0.41 – 0.52)	0.54 (0.48 – 0.62)	0.54 (0.47 – 0.62)	0.54 (0.47 – 0.62)	0.56 (0.49 – 0.64)
2006						
all		1.02 (0.93 – 1.11)	1.01 (0.93 – 1.10)	0.93 (0.84 – 1.04)	0.94 (0.86 – 1.04)	0.93 (0.86 – 1.01)
males		1.14 (1.02 – 1.28)	1.18 (1.07 – 1.300)	1.04 (0.90 – 1.20)	0.93 (0.81 – 1.05)	0.95 (0.84 – 1.07)
females		0.91 (0.80 – 1.02)	0.86 (0.76 – 0.98)	0.83 (0.73 – 0.95)	0.96 (0.83 – 1.12)	0.92 (0.81 – 1.03)

TABLE S2. (continued)

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFDA				
2000 - 2001						
all		0.15 (0.14 – 0.16)	0.16 (0.15 – 0.18)	0.17 (0.15 – 0.18)	0.17 (0.15 – 0.18)	0.16 (0.14 – 0.18)
males		0.16 (0.14 – 0.18)	0.18 (0.16 – 0.20)	0.18 (0.16 – 0.20)	0.16 (0.15 – 0.19)	0.17 (0.15 – 0.20)
females		0.14 (0.12 – 0.16)	0.15 (0.13 – 0.18)	0.15 (0.13 – 0.18)	0.17 (0.15 – 0.19)	0.14 (0.12 – 0.17)
2006						
all		0.36 (0.33 – 0.40)	0.35 (0.32 – 0.38)	0.32 (0.29 – 0.35)	0.33 (0.31 – 0.37)	0.32 (0.30 – 0.35)
males		0.37 (0.33 – 0.42)	0.39 (0.35 – 0.43)	0.35 (0.30 – 0.40)	0.33 (0.29 – 0.38)	0.33 (0.30 – 0.37)
females		0.35 (0.30 – 0.41)	0.32 (0.28 – 0.36)	0.30 (0.26 – 0.34)	0.34 (0.29 – 0.39)	0.32 (0.28 – 0.35)

TABLE S2. (continued)

		<u>Age (years)</u>				
		20 – 29	30 – 39	40 – 49	50 – 59	60 – 69
		PFUnA				
2000 - 2001						
all		0.08 (0.07 – 0.09)	0.10 (0.09 – 0.12)	0.11 (0.10 – 0.13)	0.12 (0.11 – 0.14)	0.11 (0.09 – 0.12)
males		0.09 (0.07 – 0.10)	0.10 (0.09 – 0.12)	0.13 (0.11 – 0.15)	0.12 (0.10 – 0.14)	0.12 (0.10 – 0.14)
females		0.07 (0.06 – 0.08)	0.11 (0.09 – 0.13)	0.10 (0.08 – 0.12)	0.12 (0.10 – 0.15)	0.10 (0.08 – 0.12)
2006						
all		0.16 (0.14 – 0.18)	0.18 (0.16 – 0.21)	0.17 (0.15 – 0.19)	0.20 (0.18 – 0.22)	0.20 (0.18 – 0.22)
males		0.16 (0.13 – 0.19)	0.21 (0.18 – 0.25)	0.20 (0.17 – 0.23)	0.20 (0.17 – 0.230)	0.21 (0.16 – 0.24)
females		0.16 (0.14 – 0.20)	0.16 (0.13 – 0.19)	0.16 (0.13 – 0.18)	0.20 (0.17 – 0.20)	0.19 (0.17 – 0.22)

TABLE S3. Spearman's Rank Correlation Coefficients of Natural Logs of Six Perfluorocarboxylates , American Red Cross Adult Blood Donors, by Year

Perfluorocarboxylate/ Year	PFBA	PFHpA	PFOA	PFNA	PFDA	PFUnA
PFBA						
2000 - 2001	1.000	-0.028	0.051	-0.050	-0.017	0.024
2006	1.000	0.086 ^a	0.086 ^a	0.068	0.042	0.007
PFHpA						
2000-2001		1.000	0.408 ^d	0.186 ^d	0.106 ^b	0.015
2006		1.000	0.455 ^d	0.381 ^d	0.357 ^d	0.145 ^c
PFOA						
2000 - 2001			1.000	0.440 ^d	0.354 ^d	0.172 ^d
2006			1.000	0.679 ^d	0.581 ^d	0.307 ^c
PFNA						
2000 - 2001				1.000	0.815 ^d	0.643 ^d
2006				1.000	0.883 ^d	0.633 ^d
PFDA						
2000 - 2001					1.000	0.775 ^d
2006					1.000	0.714 ^d
PFUnA						
2000 - 2001						1.000
2006						1.000

a. $p < 0.05$ b. $p < 0.01$ c. $p < 0.001$ d. $p < 0.0001$

TABLE S4. Measures of Central Tendency and Distribution for Nine Perfluorocarboxylate Concentrations (ng/mL, by Location, American Red Cross Adult Blood Donors, 2000 -2001 and 2006

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFBA						
Range						
2000-01	0.7 – 7.1	1.7 – 7.8	LLOQ – 10.8	0.2 – 5.9	1.3- 15.3	1.5 – 16.7
2006	LLOQ ^a – 3.2	LLOQ – 2.0	0.1 – 1.7	LLOQ – 3.4	LLOQ – 1.7	LLOQ – 1.3
IQR						
2000-01	2.0 – 2.9	2.4 – 3.6	1.4 – 3.2	1.6 – 2.9	2.5 – 4.6	2.8 – 4.7
2006	0.2 – 0.8	0.3 – 0.8	0.3 – 0.6	LLOQ – 1.0	0.3 – 0.6	0.2 – 0.5
90 th Percentile						
2000-01	3.6	4.0	4.0	3.8	5.2	6.8
2006	1.2	1.1	1.0	1.9	0.8	0.6
Median						
2000-01	2.4	3.0	2.2	2.2	3.4	3.8
2006	0.5	0.5	0.3	0.1	0.4	0.3
Geometric Mean and 95% CI						
2000-01	3.57 3.24 – 3.92	2.13 1.98 – 2.29	2.29 2.08 – 2.52	2.40 2.24 – 2.56	3.06 2.85 – 3.28	2.48 2.20 – 2.81
2006	0.44 0.36 – 0.55	0.50 0.44 – 0.58	0.36 0.31 – 0.42	0.15 0.11 – 0.20	0.48 0.42 – 0.54	0.23 0.19 – 0.28

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFPeA						
Range						
2000-01	LLOQ – 1.6	LLOQ – 0.3	LLOQ – 0.5	LLOQ – 0.3	LLOQ – 0.6	LLOQ – 0.4
2006	LLOQ – 1.0	LLOQ – 0.2	LLOQ – 0.1	LLOQ – 0.2	LLOQ – 0.5	LLOQ – 0.7
IQR						
2000-01	LLOQ – 0.1	LLOQ – LLOQ	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.2
2006	LLOQ – 0.1	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – 0.1	LLOQ - LLOQ
90 th Percentile						
2000-01	0.2	0.1	0.2	0.2	0.2	0.3
2006	0.2	LLOQ	LLOQ	0.1	0.2	LLOQ
Median						
2000-01	LLOQ	LLOQ	LLOQ	0.1	LLOQ	0.1
2006	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
Geometric Mean and 95% CI						
2000-01	N/A ^b	N/A	N/A	N/A	N/A	N/A
2006	N/A	N/A	N/A	N/A	N/A	N/A

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFHxA						
Range						
2000-01	LLOQ – 6.0	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.2	LLOQ – 0.2
2006	LLOQ – 1.5	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.3	LLOQ - 0.1	LLOQ – 1.5
IQR						
2000-01	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – 0.2	LLOQ - LLOQ
2006	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ - LLOQ
90 th Percentile						
2000-01	LLOQ	LLOQ	LLOQ	LLOQ	0.2	LLOQ
2006	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
Median						
2000-01	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
2006	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
Geometric Mean and 95% CI						
2000-01	N/A	N/A	N/A	N/A	N/A	N/A
2006	N/A	N/A	N/A	N/A	N/A	N/A

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFHpA						
Range						
2000-01	LLOQ – 0.9	LLOQ – 2.0	LLOQ – 0.5	LLOQ – 1.3	LLOQ – 1.3	LLOQ – 0.5
2006	LLOQ – 1.0	LLOQ – 1.5	LLOQ – 1.5	LLOQ – 0.5	LLOQ – 1.0	LLOQ – 0.6
IQR						
2000-01	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.2
2006	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.1
90 th Percentile						
2000-01	0.3	0.5	0.4	0.4	0.3	0.2
2006	0.3	0.4	0.3	0.3	0.2	0.2
Median						
2000-01	LLOQ	0.1	0.1	0.1	0.1	0.1
2006	LLOQ	0.1	0.1	LLOQ	LLOQ	LLOQ
Geometric Mean and 95% CI						
2000-01	0.11 0.10 – 0.12	0.15 0.12 – 0.18	0.13 0.12 – 0.15	0.15 0.13 – 0.17	0.11 0.10 – 0.13	0.11 0.10 – 0.12
2006	0.10 0.08 – 0.11	0.13 0.11 – 0.15	0.09 0.08 – 0.11	0.08 0.07 – 0.09	0.07 0.06 – 0.08	0.07 0.06 – 0.08

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFOA						
Range						
2000-01	1.5 – 13.9	1.5 – 29.0	1.5 – 52.3	1.5 – 34.1	1.4 – 20.0	1.5 – 16.7
2006	0.8 – 9.4	1.6 – 19.7	1.3 – 11.2	1.4 – 8.8	LLOQ – 7.2	0.8 – 28.1
IQR						
2000-01	4.1 – 7.3	4.5 – 8.6	3.3 – 5.9	3.0 – 6.6	3.3 – 5.7	2.8 – 4.7
2006	3.1 – 5.1	4.9 – 6.8	2.3 – 11.2	2.9 – 4.8	2.1 – 3.5	2.6 – 3.8
90 th Percentile						
2000-01	9.7	13.3	7.7	9.2	10.3	6.8
2006	6.5	9.1	6.0	6.1	5.4	4.7
Median						
2000-01	5.5	6.3	4.7	4.6	4.6	3.8
2006	4.1	4.9	3.3	3.9	2.8	2.6
Geometric Mean and 95% CI						
2000-01	5.38 4.96 – 5.84	6.36 5.71 – 7.08	4.45 3.99 – 4.96	4.36 3.87 – 4.90	4.60 4.13 – 5.13	3.71 3.37 – 4.09
2006	3.91 3.59 – 4.26	5.33 4.58 – 5.54	3.23 2.96 – 3.53	3.63 3.33 – 3.96	2.79 2.54 – 3.06	2.59 2.32 – 2.88

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFNA						
Range						
2000-01	0.1 – 1.6	0.3 – 2.2	0.3 – 2.7	0.1 – 2.4	0.2 – 1.2	0.1 – 1.3
2006	0.4 – 3.3	0.6 – 3.4	0.4 – 5.1	0.1 – 1.8	0.3 – 2.2	0.3 – 1.8
IQR						
2000-01	0.5 – 0.8	LLOQ – 1.2	0.5 – 1.1	0.4 – 0.7	0.4 – 0.6	0.3 – 0.5
2006	0.8 – 1.2	1.4 – 2.1	0.8 – 1.4	0.7 – 1.1	0.5 – 0.9	0.5 – 0.9
90 th Percentile						
2000-01	1.0	1.5	1.5	0.9	0.9	0.6
2006	1.7	2.5	1.8	1.3	1.5	1.2
Median						
2000-01	0.6	0.9	0.8	0.5	0.5	0.4
2006	1.0	1.7	1.1	0.8	0.7	0.7
Geometric Mean and 95% CI						
2000-01	0.61 0.57 – 0.66	0.89 0.82 – 0.96	0.76 0.69 – 0.84	0.52 0.48 – 0.57	0.46 0.42 – 0.50	0.37 0.34 – 0.41
2006	1.00 0.92 – 1.09	1.64 1.54 – 1.75	1.10 1.01 – 1.21	0.87 0.80 – 0.93	0.73 0.67 – 0.80	0.71 0.65 – 0.76

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFDA						
Range						
2000-01	LLOQ – 0.6	LLOQ – 1.2	LLOQ – 0.5	LLOQ – 0.9	LLOQ – 0.3	LLOQ – 0.4
2006	0.2 – 0.9	0.2 – 5.3	0.1 – 1.3	LLOQ – 1.1	0.1 – 1.3	0.1 – 0.8
IQR						
2000-01	0.1 – 0.2	0.2 – 0.4	0.1 – 0.3	0.1 – 0.2	0.1 – 0.2	0.1 – 0.2
2006	0.3 – 0.4	0.4 – 0.7	0.3 – 0.4	0.2 – 0.4	0.2 – 0.3	0.2 – 0.4
90 th Percentile						
2000-01	0.3	0.5	0.4	0.1	0.3	0.2
2006	0.5	0.9	0.6	0.5	0.4	0.5
Median						
2000-01	0.2	0.2	0.2	0.3	0.1	0.1
2006	0.3	0.6	0.3	0.3	0.2	0.3
Geometric Mean and 95% CI						
2000-01	0.18 0.17 – 0.20	0.25 0.23 – 0.28	0.18 0.17 – 0.20	0.14 0.12 – 0.15	0.14 0.13 – 0.15	0.12 0.11 – 0.13
2006	0.33 0.31 – 0.36	0.56 0.52 – 0.61	0.36 0.33 – 0.39	0.32 0.29 – 0.35	0.24 0.22 – 0.26	0.29 0.27 – 0.31

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFUA						
Range						
2000-01	LLOQ – 0.6	LLOQ – 1.2	LLOQ – 0.7	LLOQ – 1.1	LLOQ – 0.3	LLOQ – 0.3
2006	LLOQ – 0.9	LLOQ – 3.2	LLOQ – 1,,2	LLOQ – 1.0	LLOQ – 0.4	LLOQ – 0.9
IQR						
2000-01	0.1 – 0.2	LLOQ – 0.2	LLOQ – 0.2	LLOQ – 0.1	LLOQ – 0.1	LLOQ – 0.1
2006	0.1 – 0.3	0.2 – 0.3	0.1 – 0.3	0.1 – 0.3	0.1 – 0.2	0.1 – 0.2
90 th Percentile						
2000-01	0.3	0.3	0.3	0.2	0.2	0.2
2006	0.4	0.5	0.4	0.4	0.3	0.3
Median						
2000-01	0.2	0.1	0.1	0.1	0.1	0.1
2006	0.2	0.2	0.2	0.2	0.1	0.2
Geometric Mean and 95% CI						
2000-01	0.15 0.13 – 0.17	0.13 0.11 – 0.15	0.13 0.11 – 0.15	0.08 0.07 – 0.09	0.08 0.07 – 0.09	0.08 0.07 – 0.09
2006	0.22 0.19 – 0.24	0.24 0.21 -0.27	0.22 0.19 – 0.25	0.18 0.16 – 0.20	0.12 0.11 – 0.13	0.15 0.13 – 0.17

TABLE S4. (continued)

Fluorochemical	Boston	Charlotte	Hagerstown	Los Angeles	Mpls-St. Paul	Portland
PFDoA						
Range						
2000-01	LLOQ – 0.1	LLOQ – 0.3	LLOQ – LLOQ	LLOQ – 0.1	LLOQ – LLOQ	LLOQ - LLOQ
2006	LLOQ – 0.1	LLOQ – 0.4	LLOQ – 0.2	LLOQ – 0.1	LLOQ – LLOQ	LLOQ – 0.1
IQR						
2000-01	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ - LLOQ
2006	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ – LLOQ	LLOQ - LLOQ
90 th Percentile						
2000-01	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
2006	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
Median						
2000-01	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
2006	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ	LLOQ
Geometric Mean and 95% CI						
2000-01	N/A	N/A	N/A	N/A	N/A	N/A
2006	N/A	N/A	N/A	N/A	N/A	N/A

a. LLOQ in this particular table (S3) refers to less than the lower limit of quantitation.

b. Not applicable to calculate geometric mean because of preponderance of LLOQ values.

FIGURE S1. Comparison of age- and sex-adjusted central tendency distributions of PFUnA concentrations (ng/mL).

