Polyfluoroalkyl Chemicals in the U.S. Population: Data from the National Health and Nutrition Examination Survey (NHANES) 2003–2004 and Comparisons to NHANES 1999–2000

Supplemental Material

Antonia M. Calafat*, Lee-Yang Wong, Zsuzsanna Kuklenyik, John A. Reidy, Larry L. Needham

Division of Laboratory Sciences, National Center for Environmental Health, Centers for Disease Control and Prevention, Atlanta, Georgia, USA.

The following are available as supplemental material: Tables including the geometric mean and selected percentile concentrations in serum (in μg/L) from the National Health and Nutrition Examination Survey (NHANES) 2003–2004 of 2-(N-methyl-perfluorooctane sulfonamido) acetic acid (Table S1), perfluorodecanoic acid (Table S2), perfluorooctane sulfonamide (Table S3), perfluoroundecanoic acid (Table S4), perfluoroheptanoic acid (Table S5), and 2-(N-ethyl-perfluorooctane sulfonamido) acetic acid (Table S6); table including statistical significance differences between least squares geometric mean concentrations of PFOA, PFOS, PFHxS, and PFNA for various demographic groups (Table S7); and analytical method comparison of NHANES 1999–2000 vs. 2003–2004 (Figures S1 and S2).

Table S1. Selected percentiles of 2-(N-methyl-perfluorooctane sulfonamido) acetic acid (Me-PFOSA-AcOH) concentrations in serum (in µg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	75th percentile	90th percentile	95th percentile	N
ALL	0.7 (<LOD–0.7)	1.0 (0.8–1.1)	1.3 (1.1–1.5)	2094
12–19 years	0.6 (<LOD–0.8)	1.0 (0.9–1.2)	1.4 (1.2–1.8)	640
20–39 years	<LOD	0.9 (0.7–1.0)	1.1 (0.9–1.2)	490
40–59 years	<LOD	1.0 (0.8–1.1)	1.1 (1.0–1.4)	387
≥60 years	0.7 (<LOD–0.7)	1.0 (0.8–1.1)	1.7 (1.2–1.9)	577
Female	0.6 (<LOD–0.7)	0.9 (0.8–1.1)	1.0 (0.9–1.1)	1041
Male	0.6 (<LOD–0.7)	1.1 (0.9–1.2)	1.3 (1.1–1.6)	1053
Mexican-American	<LOD	0.7 (<LOD–0.9)	0.8 (<LOD–1.1)	485
non-Hispanic black	0.7 (<LOD–0.9)	1.1 (0.9–1.2)	1.4 (1.1–1.8)	538
non-Hispanic white	0.6 (<LOD–0.7)	1.0 (0.8–1.1)	1.2 (1.1–1.6)	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, and 50th percentile concentrations were less than the limit of detection (LOD). LOD was 0.6 µg/L. The weighted frequency of detection was 27.5%. N (sample size).

Table S2. Selected percentiles of perfluorodecanoic acid (PFDeA) concentrations in serum (in µg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	75th percentile	90th percentile	95th percentile	N
ALL	0.3 (<LOD–0.4)	0.5 (0.4–0.9)	0.8 (0.5–1.4)	2094
12–19 years	<LOD	0.5 (<LOD–1.0)	0.7 (0.3–1.2)	640
20–39 years	0.3 (<LOD–0.4)	0.6 (0.4–0.8)	0.8 (0.5–1.3)	490
40–59 years	0.4 (<LOD–0.5)	0.6 (0.4–1.1)	1.0 (0.6–2.5)	387
≥60 years	0.3 (<LOD–0.4)	0.5 (0.4–0.8)	1.0 (0.6–1.4)	577
Female	<LOD	0.4 (0.3–0.6)	0.8 (0.5–1.1)	1041
Male	0.4 (<LOD–0.5)	0.7 (0.4–1.3)	1.1 (0.6–2.0)	1053
Mexican-American	<LOD	0.5 (0.4–0.5)	0.5 (0.5–0.6)	485
non-Hispanic black	0.3 (<LOD–0.6)	0.8 (0.4–1.3)	1.0 (0.5–1.8)	538
non-Hispanic white	<LOD	0.5 (0.4–0.9)	0.8 (0.5–1.7)	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, and 50th percentile concentrations were less than the limit of detection (LOD). LOD was 0.3 µg/L. The weighted frequency of detection was 31.3%. N (sample size).

Table S3. Selected percentiles of perfluorooctane sulfonamide (PFOSA) concentrations in serum (in μg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	90th percentile	95th percentile	N
ALL	0.2 (0.2–0.3)	0.2 (0.2–0.3)	2094
12–19 years	0.3 (0.2–0.3)	0.3 (0.2–0.3)	640
20–39 years	<LOD	0.2 (0.2–0.3)	490
40–59 years	0.3 (0.2–0.3)	0.3 (0.2–0.3)	387
≥60 years	0.3 (0.3–0.4)	0.3 (0.3–0.4)	577
Female	0.3 (0.2–0.3)	0.3 (0.2–0.3)	1041
Male	0.3 (0.2–0.3)	0.3 (0.2–0.3)	1053
Mexican-American	<LOD	0.2 (<LOD–0.2)	485
non-Hispanic black	0.2 (0.2–0.3)	0.2 (0.2–0.3)	538
non-Hispanic white	0.3 (0.2–0.3)	0.3 (0.2–0.3)	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, 50th, and 75th percentile concentrations are less than the limit of detection (LOD). LOD was 0.2 μg/L. The weighted frequency of detection is 22.2%. N (sample size).

Table S4. Selected percentiles of perfluoroundecanoic acid (PFUA) concentrations in serum (in μg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	90th percentile	95th percentile	N
ALL	<LOD	0.6(<LOD–1.1)	2094
12–19 years	<LOD	<LOD	640
20–39 years	<LOD	0.3 (<LOD–0.8)	490
40–59 years	0.3 (<LOD–0.7)	0.7 (<LOD–2.2)	387
≥60 years	<LOD	0.5 (<LOD–1.0)	577
Female	<LOD	0.4 (<LOD–0.6)	1041
Male	0.4 (<LOD–0.9)	0.7 (<LOD–2.0)	1053
Mexican-American	<LOD	<LOD	485
non-Hispanic black	0.5 (<LOD–1.4)	0.8 (0.3–2.9)	538
non-Hispanic white	<LOD	0.5 (<LOD–0.8)	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, 50th, and 75th percentile concentrations are less than the limit of detection (LOD). LOD was 0.3 μg/L. The weighted frequency of detection is 9.7%. N (sample size).

Table S5. Selected percentiles of perfluoroheptanoic acid (PFHpA) concentrations in serum (in µg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	90th percentile	95th percentile	N
ALL	<LOD	0.4 (<LOD–0.5)	2094
12–19 years	0.3 (<LOD–0.5)	0.5 (0.4–0.7)	640
20–39 years	<LOD	0.3 (<LOD–0.4)	490
40–59 years	<LOD	0.3 (<LOD–0.6)	387
≥60 years	<LOD	<LOD	577
Female	<LOD	0.3 (<LOD–0.5)	1041
Male	<LOD	<LOD	1053
Mexican-American	<LOD	0.5 (<LOD–0.9)	485
non-Hispanic black	<LOD	<LOD	538
non-Hispanic white	<LOD	0.3 (<LOD–0.4)	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, 50th, and 75th percentile concentrations are less than the limit of detection (LOD). LOD was 0.3µg/L. The weighted frequency of detection is 6.2%. N (sample size).

Table S6. Selected percentiles of 2-(N-ethyl-perfluorooctane sulfonamido) acetic acid (Et-PFOSA-AcOH) concentrations in serum (in μg/L) for the U.S. population 12 years of age and older: data from the National Health and Nutrition Examination Survey 2003–2004[a]

Group	95th percentile	N
ALL	<LOD	2094
12–19 years	<LOD	640
20–39 years	<LOD	490
40–59 years	<LOD	387
≥60 years	0.4 (<LOD–0.5)	577
Female	<LOD	1041
Male	<LOD	1053
Mexican-American	<LOD	485
non-Hispanic black	0.4 (<LOD–0.5)	538
non-Hispanic white	<LOD	962

[a] The 95% confidence intervals are shown in parentheses. The 10th, 25th, 50th, 75th, and 90th percentile concentrations are less than the limit of detection (LOD). LOD was 0.4 μg/L. The weighted frequency of detection is 3.4%. N (sample size).

Table S7. Observed statistical significance values for differences between least square geometric mean concentrations of PFOA, PFOS, PFHxS, and PFNA for various demographic groups

Group	PFOA	PFOS	PFHxS	PFNA
Female vs Male	.	0.00	.	0.00
Sex effect: Age P25	0.00	.	0.00	.
Sex effect: Age P50	0.00	.	0.00	.
Sex effect: Age P75	0.02	.	0.01	.
Sex effect: Age P90	0.52	.	0.21	.
MA vs. NHB	.	.	0.01	.
MA vs. NHW	.	.	0.00	.
NHW vs. NHB	.	.	0.49	.
MA vs. NHB: Female	0.29	.	.	.
MA vs. NHB: Male	0.06	.	.	.
MA vs. NHW: Female	0.00	.	.	.
MA vs. NHW: Male	0.00	.	.	.
NHW vs. NHB: Male	0.09	.	.	.
NHW vs. NHB: female	0.00	.	.	.
MA vs. NHB: Age P25	.	0.00	.	0.03
MA vs. NHB: Age P50	.	0.00	.	0.00
MA vs. NHB: Age P75	.	0.00	.	0.00
MA vs. NHB: Age P90	.	0.00	.	0.00
MA vs. NHW: Age P25	.	0.00	.	0.00

MA vs. NHW: Age P50	.	0.00	.	0.00
MA vs. NHW: Age P75	.	0.00	.	0.01
MA vs. NHW: Age P90	.	0.00	.	0.02
NHB vs. NHW: Age P25	.	0.80	.	0.54
NHB vs. NHW: Age P50	.	0.20	.	0.05
NHB vs. NHW: Age P75	.	0.02	.	0.01
NHB vs. NHW: Age P90	.	0.01	.	0.00
Smoker effect: Age P25	.	.	.	0.04
Smoker effect: Age P50	.	.	.	0.78
Smoker effect: Age P75	.	.	.	0.31
Smoker effect: Age P90	.	.	.	0.14
<HS vs. HS	.	.	.	0.02
<HS vs. >HS	.	.	.	0.01
=HS vs. >HS	.	.	.	0.09
<HS vs. HS: Age P25	0.00	.	.	.
<HS vs. HS: Age P50	0.10	.	.	.
<HS vs. HS: Age P75	0.85	.	.	.
<HS vs. HS: Age P90	0.33	.	.	.
<HS vs. >HS: Age P25	0.02	.	.	.
<HS vs. >HS: Age P50	0.06	.	.	.
<HS vs. >HS: Age P75	0.23	.	.	.
<HS vs. >HS: Age P90	0.44	.	.	.

>HS vs. HS: Age P25	0.15	.	.	.
>HS vs. HS: Age P50	0.54	.	.	.
>HS vs. HS: Age P75	0.10	.	.	.
>HS vs. HS: Age P90	0.05	.	.	.

Age P25 (25^{th} percentile of age=26 years), age P50 (50^{th} percentile of age=41 years), age P75 (75^{th} percentile of age=55 years), and age P90 (90^{th} percentile of age=70 years). MA (Mexican-American), NHB (non-Hispanic black), NHW (non-Hispanic white). <HS (less than high school diploma), HS (high school diploma), >HS (beyond high school), SMK (smoker), NonSMK (nonsmoker).

Analytical method comparison (NHANES 1999–2000 vs. 2003–2004)

Results from the split analysis of 124 samples are shown in Figures S1 and S2. The two methods showed good agreement. Results were similar for all other analytes (data not shown). Since the LODs for the NHANES 1999-2000 method were slightly lower, the observed decrease in the frequency of detection for the sulfonamides (Me-PFOSA-AcOH, Et-PFOSA-AcOH, and PFOSA) in NHANES 2003–2004, compared to NHANES 1999–2000, must be interpreted with caution. For PFOS and PFHxS, detected in practically all of the samples analyzed for both surveys, and for PFOA and PFNA, for which the LODs were the same regardless of the survey year, analytical sensitivity was less critical than for the amides.

References

Kuklenyik Z, Needham LL, Calafat AM. 2005. Measurement of 18 perfluorinated organic acids and amides in human serum using on-line solid-phase extraction. Anal Chem 77:6085-6091.

Kuklenyik Z, Reich JA, Tully JS, Needham LL, Calafat AM. 2004. Automated solid-phase extraction and measurement of perfluorinated organic acids and amides in human serum and milk. Environ Sci Technol 38:3698-3704.

Figure S1: Correlation plot between the concentrations of 124 NHANES 2003-2004 samples analyzed with the off-line SPE-HPLC-MS/MS approach (offline-SPE) (Kuklenyik et al. 2004) and the online SPE-HPLC-MS/MS approach (online-SPE) (Kuklenyik et al. 2005).

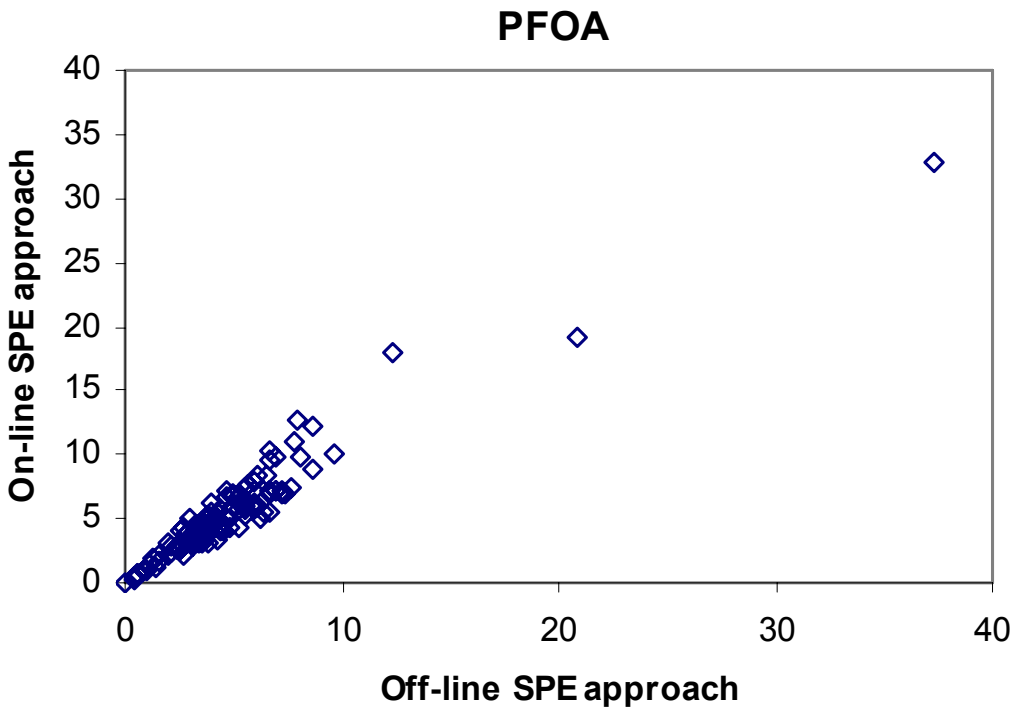

Figure S2. Repeat analysis of 124 samples for PFOA using the off-line SPE-HPLC-MS/MS (Kuklenyik et al. 2004) and the online SPE-HPLC-MS/MS (Kuklenyik et al. 2005) approaches. The Bland-Altman plot (difference in concentrations vs average of concentrations using both methods) illustrates the random variation around zero on the y-axis across the entire concentration range on the x-axis.