SUPPORTING INFORMATION

Perfluorinated Alkyl Acids in Blood Serum from Primiparous Women in Sweden: Serial Sampling during Pregnancy and Nursing, and Temporal Trends 1996-2010

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Table S1. Composition of the pooled serum samples in the temporal trend study.[a]

year (N)	no. of pools	N in each pool	age (years, mean±SD)
1996 (19)	3	6-7	30±5
1997 (62)	3	20-21	28±4
1998 (74)	3	24-25	29±4
1999 (17)	3	5-6	27±3
2000 (20)	2	10	30±4
2001 (9)	1	9	29±5
2002 (31)	3	10-11	30±3
2004 (32)	3	10-11	29±4
2006 (30)	3	10	30±4
2007 (29)	3	9-10	30±5
2008 (30)	3	10	29±5
2009 (30)	3	10	29±4
2010 (30)	3	10	30±5

[a] Blood serum was sampled from primiparous women 3 weeks after delivery.

Table S2. Analytical standard compounds used in the present study and selected instrumental parameters.

Compound	Acronym	Obtained as	Supplier	Triple quadrupole MS/MS ESI negative ions, Capillary 2.5 kV, Source Temp. 120 ºC, Desolv. Temp. 150 ºC, Drying Gas N_2 400 L/h, Nebulizing Gas N_2 20 L/h, Collision Gas Ar 5×10^{-4} mbar			Q-ToF-HRMS ESI negative ions, Capillary 1.0 kV, Source Temp. 100 ºC, Desolv. Temp. 350 ºC, Desolv. Gas N_2 800 L/h	
				Cone voltage [V]	MRM precursor → product mass [m/z]	Collision energy [eV]	Cone voltage [V]	Quantification mass [m/z]
Perfluorobutane sulfonate	PFBS	Potassium salt	Wellington mixture PFS-MXA[b]	52	499 → 80+99	77	50	298.94
Perfluorohexane sulfonate	PFHxS	Sodium salt					50	398.94
Perfluorooctane sulfonate[a]	PFOS	Sodium salt					50	498.93
Perfluorodecane sulfonate	PFDS	Sodium salt					50	598.92
Perfluorooctane sulfonamide	FOSA	-	Wellington[b]				50	497.95
Perfluorohexanoate	PFHxA	Carboxylic acid	Wellington mixture PFC-MXA[b]	20	413 → 369	10	20	268.98
Perfluoroheptanoate	PFHpA	Carboxylic acid					20	318.98
Perfluorooctanoate	PFOA	Carboxylic acid					20	368.98
Perfluorononanoate	PFNA	Carboxylic acid		20	463 → 419	12	20	418.97
Perfluorodecanoate	PFDA	Carboxylic acid					20	468.97
Perfluoroundecanoate	PFUnDA	Carboxylic acid					20	518.97
Perfluorododecanoate	PFDoDA	Carboxylic acid					20	568.96
Perfluorotridecanoate	PFTrDA	Carboxylic acid					20	618.96
Perfluorotetradecanoate	PFTeDA	Carboxylic acid					20	668.96
$^{18}O_2$-Perfluoro-*n*-hexane sulfonate[c]	$^{18}O_2$-PFHxS	Sodium salt	Wellington mixture MPFAC-MXA[b]	52	503 → 80	77	50	402.93
$^{13}C_4$-Perfluoro-*n*-octane sulfonate[c,d]	$^{13}C_4$-PFOS	Sodium salt					50	502.93
$^{13}C_2$-Perfluoro-*n*-hexanoate[c]	$^{13}C_2$-PFHxA	Carboxylic acid		20	417 → 372	10	20	269.99
$^{13}C_4$-Perfluoro-*n*-octanoate[c,d]	$^{13}C_4$-PFOA	Carboxylic acid					20	371.98
$^{13}C_5$-Perfluoro-*n*-nonanoate[c]	$^{13}C_5$-PFNA	Carboxylic acid					20	422.98
$^{13}C_2$-Perfluoro-*n*-decanoate[c]	$^{13}C_2$-PFDA	Carboxylic acid					20	469.98
$^{13}C_2$-Perfluoro-*n*-undecanoate[c]	$^{13}C_2$-PFUnDA	Carboxylic acid					20	519.97
$^{13}C_2$-Perfluoro-*n*-dodecanoate[c]	$^{13}C_2$-PFDoDA	Carboxylic acid					20	569.97
$^{13}C_8$-Perfluoro-*n*-octane sulfonate[e]	$^{13}C_8$-PFOS	Sodium salt	Wellington[b]	52	507 → 80	77	50	506.93
$^{13}C_8$-Perfluoro-*n*-octanoate[e]	$^{13}C_8$-PFOA	Carboxylic acid	Wellington[b]	20	421 → 376	10	20	375.98

[a] A technical PFOS standard from Fluka (Buchs, Switzerland) was applied in MS/MS
[b] Wellington, Guelph, ON, Canada
[c] Used as internal (surrogate) standard
[d] Also obtained as individual standard
[e] Used as volumetric standard

Table S3. Linear regression (r^2 values) and residuals for the solvent based calibration curves.[a]

	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA
r^2	0.9991	0.9994	0.9992	0.9997	0.9989	0.9998	0.9986	0.9995	0.9992
residuals	<12%	<14%	<9%	<8%	<15%	<8%	<7%	<13%	<12%
	PFBS	PFHxS	PFOS	PFDS	FOSA				
r2	0.9990	0.9999	0.9998	0.9996	0.9999				
residuals	<15%	<6%	<8%	<11%	<12%				

[a] Five concentrations included from the individual quantification limits to 5 ng/mL.

Table S4. Compound specific method detection (MDLs) and quantification limits (MQLs) of perfluorinated substances in human serum samples as well as mean recovery ± relative standard error (*n*=3) from spiked (2 ng/g) serum and whole blood samples.

	Triple quad. MS/MS		Q-ToF-HRMS		Recovery[a]	
	MDL [ng/g]	MQL [ng/g]	MDL [ng/g]	MQL [ng/g]	Serum [%]	Whole blood [%]
PFHxA			0.05	0.15	80±4	72±6
PFHpA			0.05	0.15	78±2	83±4
PFOA	0.15	0.42	0.035	0.1	84±5[b]	81±3
PFNA	0.05	0.16	0.03	0.1	90±2	89±5
PFDA			0.07	0.2	93±6	87±5
PFUnDA			0.05	0.15	92±3	83±4
PFDoDA			0.1	0.3	90±3	84±4
PFTrDA			0.15	0.45	81±4	75±6
PFTeDA			0.25	0.7	72±5	68±5
PFBS			0.013	0.03	77±4	83±4
PFHxS			0.007	0.02	81±2[b]	82±3
PFOS	0.2	0.57	0.008	0.025	83±4[c]	86±3
PFDS			0.01	0.03	76±5	80±5
FOSA			0.04	0.1	77±3	78±2

[a] Calculated after subtraction of endogenous PFAS concentrations.
[b] Uncertain estimations due to the relatively high endogenous concentrations (PFOA 2.3 ng/g and PFHxS 1.4 ng/g quantified) compared to the spiked concentrations of 2 ng/g.
[c] A spiking level of 20 ng/g was used for PFOS due to the high endogenous concentrations (24.2 ng/g quantified).

Table S5. Mean recovery ± relative standard error for all internal standards in the temporal trend study (*n*=36) and for $^{13}C_4$-PFOA and $^{13}C_4$-PFOS in the serial samples study (*n*=95). The spike concentration was 10 ng/g.

	$^{13}C_2$-PFHxA	$^{13}C_4$-PFOA	$^{13}C_5$-PFNA	$^{13}C_2$-PFDA	$^{13}C_2$-PFUnDA	$^{13}C_2$-PFDoDA	$^{18}O_2$-PFHxS	$^{13}C_4$-PFOS
Recovery [%] (*n*=36)	78±3	84±1	92±1	86±2	88±1	83±2	84±0.6	81±2
Recovery [%] (*n*=95)		84±0.8						80±1

Table S6. Levels of PFAAs and FOSA [ng/g] in pooled samples of blood serum from nursing women 3 weeks after delivery.[a]

Year	PFBS	PFHxS	PFOS	PFDS	FOSA	PFHpA	PFOA	PFNA	PFDA	PFUnDA
1996	<0.013	1.61	22.7	0.137	0.793	*0.075*	2.18	0.468	*0.159*	0.189
1996	*0.014*	2.44	27.3	0.042	0.572	*0.078*	2.92	0.386	*0.186*	0.212
1996	*0.021*	2.24	23.3	0.260	0.507	*0.084*	2.69	0.502	0.211	0.163
1997	*0.028*	2.30	24.8	0.089	0.610	*0.100*	3.07	0.375	0.231	0.228
1997	<0.013	1.55	20.3	0.153	0.602	*0.114*	2.26	0.258	0.221	0.154
1997	<0.013	1.63	20.7	0.148	0.441	*0.080*	2.54	0.439	0.234	0.261
1998	<0.013	1.16	20.2	*0.021*	0.477	*0.073*	2.22	0.393	*0.198*	0.168
1998	<0.013	1.99	23.1	0.166	0.410	*0.140*	2.66	0.438	0.226	0.223
1998	*0.019*	2.19	21.0	0.044	0.514	*0.084*	2.35	0.411	0.209	0.202
1999	<0.013	2.00	20.0	*0.020*	0.512	*0.061*	2.38	0.311	*0.136*	0.153
1999	<0.013	2.95	21.5	0.046	0.584	*0.056*	3.11	0.404	*0.152*	0.262
1999	*0.013*	1.80	23.0	*0.022*	0.572	*0.118*	2.49	0.387	*0.173*	*0.144*
2000	*0.018*	2.40	18.7	0.048	0.363	*0.063*	2.65	0.353	*0.173*	0.198
2000	<0.013	3.03	22.0	0.052	0.441	*0.094*	2.50	0.384	*0.172*	0.201
2001	*0.017*	1.96	28.1	0.057	0.572	*0.110*	3.05	0.609	0.264	0.320
2002	*0.024*	2.25	17.0	0.064	0.185	*0.079*	2.17	0.353	*0.182*	0.228
2002	*0.016*	3.04	18.7	0.037	0.282	*0.132*	2.59	0.469	0.223	0.272
2002	<0.013	2.87	23.2	0.048	0.301	*0.087*	2.98	0.493	0.267	0.274
2004	*0.028*	2.16	16.0	0.037	*0.073*	*0.102*	2.12	0.497	0.261	0.303
2004	*0.025*	3.82	16.6	0.051	0.205	*0.097*	2.12	0.474	0.295	0.299
2004	<0.013	1.85	13.6	0.052	0.105	*0.107*	2.15	0.431	0.285	0.180
2006	0.067	3.87	16.5	0.043	0.101	*0.089*	2.11	0.573	0.277	0.240
2006	0.042	5.26	12.2	*0.026*	<0.040	*0.093*	1.89	0.432	0.245	0.175
2006	0.032	3.24	10.7	*0.025*	<0.040	*0.080*	1.70	0.417	0.253	0.241
2007	0.036	4.48	15.1	0.057	*0.071*	*0.090*	2.41	0.755	0.289	0.245
2007	0.033	4.68	18.3	*0.022*	*0.078*	*0.065*	2.42	0.553	0.287	0.225
2007	*0.025*	3.20	8.80	*0.023*	*0.056*	*0.064*	1.36	0.503	0.200	0.185
2008	0.051	5.05	11.1	*0.021*	<0.040	*0.114*	2.01	0.798	*0.190*	0.208
2008	0.060	3.92	9.25	*0.024*	*0.049*	*0.058*	1.69	0.499	0.275	0.254
2008	0.064	4.12	10.4	0.039	<0.040	*0.082*	2.58	0.866	0.386	0.262
2009	0.082	4.00	7.14	*0.023*	<0.040	*0.056*	1.54	0.450	0.352	0.281
2009	0.054	4.58	8.68	*0.019*	<0.040	*0.086*	2.40	0.783	0.329	0.247
2009	0.091	5.95	8.89	0.035	<0.040	*0.107*	1.80	0.722	0.288	0.285
2010	0.070	5.83	7.61	*0.025*	<0.040	*0.112*	1.96	0.857	0.421	0.311
2010	0.092	5.63	5.11	*0.011*	<0.040	*0.073*	1.71	0.701	0.299	0.232
2010	0.101	7.95	7.62	*0.014*	<0.040	*0.082*	1.39	0.589	0.276	0.188

[a] PFAAs with levels below the method detection limit (MDL) in all samples: PFHxA (<0.05 ng/g serum), PFDoDA (<0.1 ng/g), PFTrDA (<0.15 ng/g) and PFTeDA (<0.25 ng/g). Data in italics are above MDL but below the method quantification limit. For information about number of individual samples in each pool see Table S1.