SUPPLEMENTARY INFORMATION

PERFLUOROALKANE ACIDS IN HUMAN MILK UNDER THE GLOBAL MONITORING PLAN OF THE STOCKHOLM CONVENTION ON PERSISTENT ORGANIC POLLUTANTS (2008–2019)

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Global Indicators According to World Bank

Table S 1: WBC classification for selected years as GNI per capita, current US$, Atlas method (World Bank Open Data Catalog) according to World Bank (U.S. dollar) (World Bank, No Year-a)

		2008	2012	2017	2018	2019
High income	H	>11,905	>12,615	>12,055	>12,375	>12,535
Upper middle income	UM	3,856-11,905	4,086-12,615	3,896-12,055	3,996-12,375	4,046-12,535
Lower middle income	LM	976-3,855	1,036-4,085	996-3,895	1,026-3,995	1,036-4,045
Low income	L	≤975	≤1,035	≤995	≤1,025	≤1,035

Table S 2: Population density codes based on World Bank visualization (for 2019) (World Bank, No Year-b)

PD_Code	Range: Inhabitants *per* km²
A	<30.6
B	>30.61-<56.9
C	>56.91-<103.7
D	>103.71-<237.3
E	>237.31-<19198.7
F	>19197.71

Chemical Analysis

For the identification and quantification of PFOS isomers, the potassium salt of a technical grade PFOS containing 78.8% perfluorooctane-1-sulfonate, the linear isomer (L-PFOS) and 21.2% of a mixture of branched isomers (br-PFOS) (Wellington Laboratories, Guelph, ON, Canada) was used. For PFOA and PFHxS, only the linear isomers were quantified since no analytical standards existed for the identification and quantification of the branched isomers at the time of the project's conceptualization (US-EPA, 2019, 2020).

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The chemical analysis includes alkaline digestion with sodium hydroxide (NaOH) and liquid-liquid extraction followed by weak anion exchange, solid-phase extraction (Waters Oasis® WAX; SPE-WAX; Waters Corporation Milford, USA) and additional clean-up with Supelclean ENVI-Carb SPE tube. The target PFAA were separated and quantified by using an ultra-performance liquid chromatography electrospray ionization (ESI) operating in the negative ionization mode coupled to a triple-quadrupole mass spectrometer detector (XEVO TQS Waters Corporation, Milford, USA). Aliquots of 10 µL were injected onto a BEH (ethylene bridged hybrid) C_{18} column (1.7 µm, 2.1 mm × 100 mm; Waters Corporation Milford, USA) kept at 50 °C. The mobile phases consisted of ammonium acetate (2 mM) in 80:20 (v:v) water: acetonitrile (A), and ammonium acetate (2 mM) in acetonitrile (B). The flow rate was set at 0.5 mL/min, and the eluent gradient started at 99% A for 0.57 min followed by linear ramp up to 70% B till 10 min, then linear increase for 1 min to 100% B that was held for 1 min, then reverted to initial conditions of 99% A for 1.2 min, and 0.8 min to allow the system to equilibrate prior to next injection. A graphical sketch is shown in Figure S 1.

Laboratory produced Milli-Q water was used throughout the experiments. Acetonitrile (HPLC grade, ≥ 99.8%), methanol (HPLC grade, ≥ 99.8%), sodium hydroxide and ammonia solution (0.91, 25%) were acquired from Fisher Scientific (Waltham, MA, United States), ammonium acetate (≥99.0%), and acetic acid (glacial) from Sigma Aldrich (Darmstadt, Germany).

One procedural blank and one quality control sample (human milk sample obtained from 4th round of the UNEP-coordinated bi-ennial interlaboratory assessment (UNEP et al., 2021)) were included together with the human milk samples by performing the entire run to estimate the background contamination introduced during analytical procedures and evaluate the reproducibility of the analytical method. The limit of quantification (LOQ) was defined as the lowest point in the calibration curve. Recovery was evaluated based on the measured amount of mass-labelled extraction standard spiked before extraction with the injection standard spiked after extraction multiplied by 100 percent.

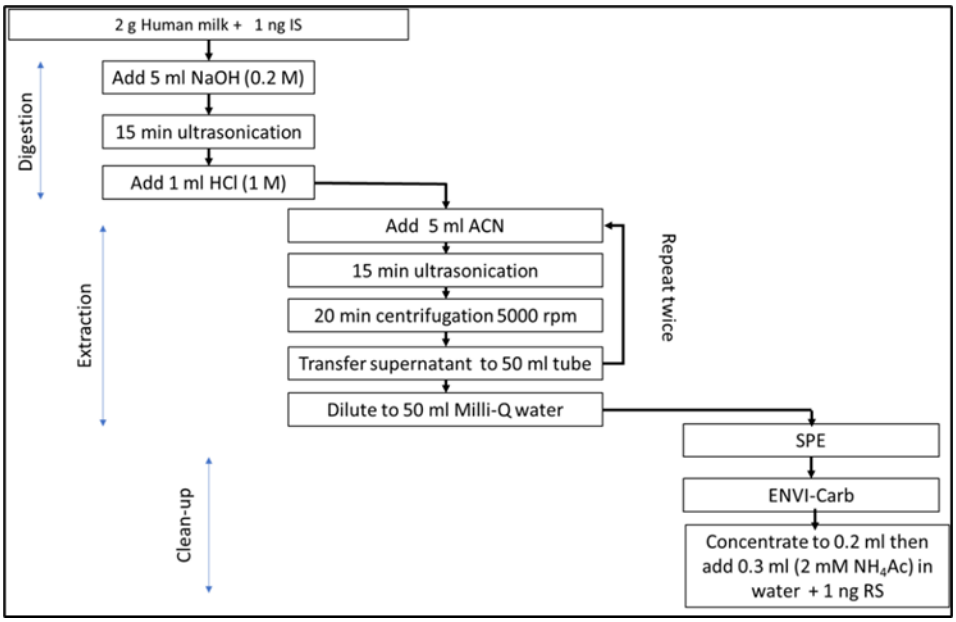

Figure S 1: Schematic diagram for extraction of PFAA from human milk samples

ACN	acetonitrile	NH_4Ac	ammonium acetate
HCl	hydrochloric acid	rpm	revolutions per minute
NaOH	sodium hydroxide	SPE	solid phase extraction

Table S 3: Overview classification of the national pools using the development indicators (World Bank, No Year-c) and status of ratification of the 2019 amendments of the Stockholm Convention concerning PFOS and PFOA (R_PFAS)

Sample_ID	ISO3	Year	WBC	PD	R_PFAS	Sample_ID	ISO3	Year	WBC	PD	R_PFAS
CIV (2010)	CIV	2010	LM	C	Yes	BGR (2014)	BGR	2014	UM	C	Yes
CIV (2015)	CIV	2015	LM	C	Yes	CZE (2014)	CZE	2014	H	D	Yes
COD (2009)	COD	2009	L	A	Yes	CZE (2019)	CZE	2019	H	D	Yes
COD (2017)	COD	2017	L	B	Yes	GEO (2009)	GEO	2009	LM	C	Yes
ETH (2019)	ETH	2019	L	C	Yes	GEO (2014)	GEO	2014	LM	C	Yes
GHA (2009)	GHA	2009	L	D	Yes	HRV (2014)	HRV	2014	H	C	Yes
GHA (2019)	GHA	2019	LM	D	Yes	LTU (2009)	LTU	2009	UM	B	Yes
KEN (2009)	KEN	2009	L	C	Yes	LTU (2015)	LTU	2015	H	B	Yes
KEN (2019)	KEN	2019	LM	C	Yes	MDA (2009)	MDA	2009	LM	D	No
MAR (2019)	MAR	2019	LM	C	Yes	MDA (2015)	MDA	2015	LM	D	No
MLI (2009)	MLI	2009	L	A	Yes	ROU (2014)	ROU	2014	UM	C	Yes
MLI (2019)	MLI	2019	L	A	Yes	SVK (2019)	SVK	2019	H	D	Yes
MUS (2009)	MUS	2009	UM	E	No	ARG (2019)	ARG	2019	UM	A	No
MUS (2018)	MUS	2018	UM	E	No	ATG (2008)	ATG	2008	H	D	Yes
NER (2011)	NER	2011	L	A	Yes	ATG (2018)	ATG	2018	H	D	Yes
NER (2015)	NER	2015	L	A	Yes	BRA GR1 (2012)	BRA	2012	UM	A	Yes
NGA (2008)	NGA	2008	LM	D	Yes	BRA GR2 (2012)	BRA	2012	UM	A	Yes
NGA (2019)	NGA	2019	LM	D	Yes	BRA GR3 (2012)	BRA	2012	UM	A	Yes
SEN (2009)	SEN	2009	LM	C	Yes	BRB (2018)	BRB	2018	H	E	Yes
SEN (2018)	SEN	2018	LM	C	Yes	CHL (2008)	CHL	2008	UM	A	Yes
TGO (2010)	TGO	2010	L	D	Yes	CHL (2011)	CHL	2011	UM	A	Yes
TGO (2017)	TGO	2017	L	D	Yes	COL (2019)	COL	2019	UM	B	Yes
TUN (2019)	TUN	2019	LM	C	Yes	ECU (2019)	ECU	2019	UM	C	Yes
TZA (2019)	TZA	2019	L	C	Yes	HTI (2011)	HTI	2011	L	E	No
UGA (2009)	UGA	2009	L	D	Yes	HTI (2015)	HTI	2015	L	E	No
UGA (2018)	UGA	2018	L	D	Yes	JAM (2011)	JAM	2011	UM	E	Yes
ZMB (2019)	ZMB	2019	LM	A	Yes	JAM (2018)	JAM	2018	UM	E	Yes
IDN (2011)	IDN	2011	LM	D	Yes	MEX (2011)	MEX	2011	UM	C	Yes
IND (2009)	IND	2009	LM	E	No	MEX (2017)	MEX	2017	UM	C	Yes
KHM (2019)	KHM	2019	LM	C	Yes	PER (2011)	PER	2011	UM	A	Yes
MNG (2018)	MNG	2018	LM	A	Yes	PER (2019)	PER	2019	UM	A	Yes
SYR (2009)	SYR	2009	LM	D	Yes	URY (2009)	URY	2009	UM	A	Yes
THA (2018)	THA	2018	UM	D	Yes	URY (2019)	URY	2019	H	A	Yes
TJK (2009)	TJK	2009	L	B	Yes	AUT (2016)	AUT	2016	H	C	Yes
VNM (2019)	VNM	2019	LM	E	Yes	BEL (2015)	BEL	2015	H	E	Yes
FJI (2011)	FJI	2011	LM	B	Yes	CHE (2016)	CHE	2016	H	D	Yes
FJI (2019)	FJI	2019	UM	B	Yes	DEU NS (2019)	DEU	2019	H	D	Yes
KIR (2018)	KIR	2018	LM	D	Yes	DEU SH (2019)	DEU	2019	H	D	Yes
MHL (2019)	MHL	2019	UM	E	Yes	IRL (2019)	IRL	2019	H	C	Yes
NIU (2017)	NIU	2017	UM	A	Yes	NLD (2014)	NLD	2014	H	E	Yes
NIU (2019)	NIU	2019	UM	A	Yes	SWE (2019)	SWE	2019	H	A	Yes
PLW (2018)	PLW	2018	H	B	Yes						
SLB (2019)	SLB	2019	LM	A	Yes						
VUT (2018)	VUT	2018	LM	A	No						
WSM (2019)	WSM	2019	UM	C	Yes						

Table S 4: Number of samples (and percentages) according to geographic distribution (as UN region), 5-year periods, income (as WBC), population density (using PD_Code as defined in Table S 2), and ratification of PFOS and PFOA amendments

	Africa (N=27)	Asia (N=18)	CEE (N=12)	GRULAC (N=21)	WEOG (N=8)	Overall (N=86)
Region	27 (31.4%)	18 (20.9%)	12 (14.0%)	21 (24.4%)	8 (9.3%)	86 (100%)
Period						
2005-2009	8	3	3	3	0	17 (19.8%)
2010-2014	3	2	5	8	1	19 (22.1%)
2015-2019	16	13	4	10	7	50 (58.1%)
WBC						
H	0	1	5	4	8	18 (20.9%)
UM	2	6	3	15	0	26 (30.2%)
LM	11	10	4	0	0	25 (29.1%)
L	14	1	0	2	0	17 (19.8%)
Population density						
A	6	5	0	10	1	22 (25.6%)
B	1	4	2	1	0	8 (9.3%)
C	9	2	5	3	1	20 (23.3%)
D	9	4	5	2	2	22 (25.6%)
E	2	3	0	5	4	14 (16.3%)
R_PFAS						
No	2	2	2	3	0	9 (10.5%)
Yes	25	16	10	18	8	77 (89.5%)

* PD_Code ‘A’ refers to least densely populated, ‘E’ to most densely populated countries (see Table S 2)

Detailed Results

Table S 5: Results for PFAA concentrations in all national pools (pg/g f.w.). LOQs are displayed as "<" and in red color.
Results for PFOS, PFOA, and PFHxS from 44 samples collected 2016-2019 have been published previously (Fiedler and Sadia, 2021)

Country	Year	Sample_ID	L_PFOS	br_PFOS	PFOS	PFOA	PFHxS	PFBS	PFDS	PFBA	PFPeA	PFHxA	PFHpA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTDA
Africa																			
Congo DR	2009	COD (2009)	8.25	3.16	11.4	13.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Congo DR	2017	COD (2017)	<6.2	<1.2	<6.2	12.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Côte D'Ivoire	2010	CIV (2010)	26.0	6.32	32.4	16.2	<5.5	<5	<6.2	51	9.92	45.8	6.3	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Côte d'Ivoire	2015	CIV (2015)	37.5	7.07	44.6	17.0	<5.5	<5	<6.2	9	<6.2	12.2	<6.2	<6.2	<6.2	6.6	<6.2	<6.2	<6.2
Ethiopia	2019	ETH (2019)	6.20	2.91	9.1	6.2	<5.5	<5	<6.2	26.6	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Ghana	2009	GHA (2009)	18.1	6.45	24.6	16.6	<5.5	<5	<6.2	12	8.6	34.5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Ghana	2019	GHA (2019)	12.0	3.60	15.6	11.3	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Kenya	2009	KEN (2009)	8.68	3.72	12.4	17.5	<5.5	<5	<6.2	41	9.2	31.9	5.8	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Kenya	2019	KEN (2019)	<6.2	<1.2	<6.2	10.4	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mali	2009	MLI (2009)	18.7	5.15	23.9	20.8	7.56	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mali	2019	MLI (2019)	16.4	5.46	21.9	12.8	<5.5	<5	<6.2	37.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mauritius	2009	MUS (2009)	15.7	7.32	23.0	28.1	<5.5	<5	<6.2	98	<6.2	7.6	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mauritius	2018	MUS (2018)	11.5	1.30	12.8	18.0	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Morocco	2019	MAR (2019)	11.5	2.05	13.6	15.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Niger	2011	NER (2011)	16.7	2.54	19.2	21.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Niger	2015	NER (2015)	13.8	4.90	18.7	15.0	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Nigeria	2008	NGA (2008)	24.2	6.39	30.6	14.4	<5.5	<5	<6.2	45	<6.2	10.1	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Nigeria	2019	NGA (2019)	8.7	1.74	10.4	14.3	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Senegal	2009	SEN (2009)	11.6	3.84	15.4	16.4	<5.5	<5	<6.2	<6.2	12.6	29.5	7.3	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Senegal	2018	SEN (2018)	8.12	2.11	10.2	13.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Tanzania	2019	TZA (2019)	7.50	<1.2	7.5	11.0	<5.5	<5	<6.2	<6.2	<6.2	<6.2	9.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Togo	2010	TGO (2010)	19.4	8.06	27.5	24.5	5.64	<5	<6.2	<6.2	<6.2	10.9	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Togo	2017	TGO (2017)	11.0	1.80	12.8	17.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Tunisia	2019	TUN (2019)	15.7	4.09	19.8	18.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Uganda	2009	UGA (2009)	6.57	2.54	9.1	63.4	<5.5	<5	<6.2	1,155	<6.2	<6.2	<6.2	17.7	<6.2	<6.2	<6.2	<6.2	<6.2
Uganda	2018	UGA (2018)	<6.2	<1.2	<6.2	8.4	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Zambia	2019	ZMB (2019)	<6.2	<1.2	<6.2	8.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Asia-Pacific																			
Cambodia	2019	KHM (2019)	14.6	2.60	17.2	14.6	<5.5	<5	<6.2	33.0	<6.2	<6.2	<6.2	10.7	8.7	<6.2	<6.2	<6.2	<6.2
India	2009	IND (2009)	7.69	4.28	12.0	12.8	<5.5	<5	<6.2	28	<6.2	24.7	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Indonesia	2011	IDN (2011)	17.4	4.84	22.3	20.0	<5.5	<5	<6.2	53	<6.2	34.9	8.9	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mongolia	2018	MNG (2018)	8.2	1.92	10.2	20.1	<5.5	<5	<6.2	8.1	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Syria	2009	SYR (2009)	9.11	4.28	13.4	20.5	<5.5	<5	<6.2	83	<6.2	12.0	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2

Country	Year	Sample_ID	L_PFOS	br_PFOS	PFOS	PFOA	PFHxS	PFBS	PFDS	PFBA	PFPeA	PFHxA	PFHpA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTDA
Tajikistan	2009	TJK (2009)	6.88	1.05	7.9	17.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Thailand	2018	THA (2018)	21.8	3.72	25.5	25.8	7.32	<5	<6.2	27.8	<6.2	<6.2	<6.2	7.3	<6.2	<6.2	<6.2	<6.2	<6.2
Vietnam	2019	VNM (2019)	26.8	5.27	32.1	13.0	<5.5	<5	<6.2	688	<6.2	<6.2	<6.2	9.6	8.1	<6.2	<6.2	<6.2	<6.2
Subgroup of Pacific Islands																			
Fiji	2011	FJI (2011)	18.0	8.18	26.2	35.0	5.77	<5	<6.2	144	<6.2	<6.2	10.5	11.2	<6.2	<6.2	<6.2	<6.2	<6.2
Fiji	2019	FJI (2019)	<6.2	<1.2	<6.2	10.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Kiribati	2018	KIR (2018)	154	57.78	211.9	31.8	111	<5	<6.2	10.8	<6.2	<6.2	<6.2	6.6	<6.2	<6.2	<6.2	<6.2	<6.2
Marshall Isl.	2019	MHL (2019)	12.9	<1.2	12.9	12.9	<5.5	<5	<6.2	7.4	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Niue	2017	NIU (2017)	19.2	2.42	21.6	17.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Niue	2019	NIU (2019)	19.5	5.39	24.9	10.0	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Palau	2018	PLW (2018)	9.49	<1.2	9.5	20.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Samoa	2019	WSM (2019)	26.0	<1.2	26.0	21.5	<5.5	<5	<6.2	7.4	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Solomon Isl.	2019	SLB (2019)	14.0	1.30	15.3	11.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Vanuatu	2018	VUT (2018)	9.18	<1.2	9.2	11.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
CEE																			
Bulgaria	2014	BGR (2014)	27.8	7.07	34.9	31.1	<5.5	<5	<6.2	78	<6.2	9.7	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Croatia	2014	HRV (2014)	23.0	5.52	28.5	36.3	5.95	<5	<6.2	35	<6.2	<6.2	<6.2	7.6	<6.2	<6.2	<6.2	<6.2	<6.2
Czech Rep	2014	CZE (2014)	22.3	12.8	35.2	38.9	<5.5	<5	<6.2	33	<6.2	<6.2	<6.2	6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Czech Rep	2019	CZE (2019)	11.9	2.91	14.8	24.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Georgia	2009	GEO (2009)	26.1	5.21	31.3	20.0	<5.5	<5	<6.2	49	9.5	31.7	<6.2	6.4	<6.2	<6.2	<6.2	<6.2	<6.2
Georgia	2014	GEO (2014)	15.1	4.03	19.2	12.6	<5.5	<5	<6.2	54	<6.2	8.9	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Lithuania	2009	LTU (2009)	24.9	9.05	34.0	30.3	<5.5	<5	<6.2	44	<6.2	5.3	<6.2	6.4	<6.2	<6.2	<6.2	<6.2	<6.2
Lithuania	2015	LTU (2015)	12.2	5.70	17.9	33.9	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	14.8	<6.2	<6.2	<6.2	<6.2	<6.2
Moldova	2009	MDA (2009)	52.8	9.92	62.7	29.2	<5.5	<5	<6.2	29	<6.2	30.2	<6.2	10.7	<6.2	7.2	<6.2	<6.2	<6.2
Moldova	2015	MDA (2015)	44.1	18.3	62.4	48.8	9.61	<5	<6.2	8	<6.2	<6.2	<6.2	6.7	<6.2	<6.2	<6.2	<6.2	<6.2
Romania	2014	ROU (2014)	70.5	12.8	83.3	32.6	12.90	<5	<6.2	58	<6.2	11.1	<6.2	8.0	<6.2	<6.2	<6.2	<6.2	<6.2
Slovakia	2019	SVK (2019)	9.92	2.29	12.2	28.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
GRULAC																			
Antigua Barb.	2008	ATG (2008)	24.2	10.7	35.0	45.8	6.45	<5	<6.2	<6.2	<6.2	51.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Antigua Barb.	2018	ATG (2018)	11.0	2.48	13.5	18.8	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Argentina	2019	ARG (2019)	9.11	2.73	11.8	11.8	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Barbados	2018	BRB (2018)	9.80	2.91	12.7	19.0	<5.5	<5	<6.2	7.6	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Brazil	2012	BRA_GR1(2012)	17.4	8.68	26.1	30.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Brazil	2012	BRA_GR2(2012)	32.3	15.2	47.5	27.4	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Brazil	2012	BRA_GR3(2012)	42.7	22.6	65.3	34.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Chile	2008	CHL (2008)	12.3	6.14	18.4	61.5	<5.5	<5	<6.2	100	39.7	162.9	21.8	17.7	<6.2	7.1	<6.2	<6.2	<6.2
Chile	2011	CHL (2011)	7.94	4.28	12.2	23.4	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Colombia	2019	COL (2019)	<6.2	<1.2	<6.2	15.9	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Ecuador	2019	ECU (2019)	9.36	<1.2	9.4	8.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Haiti	2011	HTI (2011)	26.4	11.7	38.1	19.7	34.78	<5	<6.2	113	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Haiti	2015	HTI (2015)	15.4	5.64	21.1	25.3	7.75	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Jamaica	2011	JAM (2011)	29.5	10.2	39.7	23.8	5.83	<5	<6.2	79	<6.2	8.5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2

Country	Year	Sample_ID	L_PFOS	br_PFOS	PFOS	PFOA	PFHxS	PFBS	PFDS	PFBA	PFPeA	PFHxA	PFHpA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTDA
Jamaica	2018	JAM (2018)	15.3	3.10	18.4	16.6	<5.5	<5	<6.2	7.7	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Mexico	2011	MEX (2011)	21.6	6.88	28.5	32.1	<5.5	<5	<6.2	61	<6.2	<6.2	<6.2	8.9	<6.2	<6.2	<6.2	<6.2	<6.2
Mexico	2017	MEX (2017)	6.32	<1.2	6.3	17.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Peru	2011	PER (2011)	6.32	3.04	9.4	15.1	<5.5	<5	<6.2	66	<6.2	7.0	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Peru	2019	PER (2019)	<6.2	<1.2	<6.2	7.8	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Uruguay	2009	URY (2009)	40.5	16.4	56.9	23.8	<5.5	<5	<6.2	42	<6.2	12.4	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Uruguay	2019	URY (2019)	28.5	12.0	40.5	13.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
WEOG																			
Austria	2016	AUT (2016)	15.3	4.46	19.7	33.4	<5.5	<5	<6.2	7.7	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Belgium	2015	BEL (2015)	22.4	6.45	28.9	19.9	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	7.9	<6.2	<6.2	<6.2	<6.2	<6.2
Germany	2019	DEU_NS (2019)	13.3	2.60	15.9	18.3	<5.5	<5	<6.2	10.1	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Germany	2019	DEU_SH (2019)	15.4	4.28	19.7	17.7	<5.5	<5	<6.2	8.9	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Ireland	2019	IRL (2019)	10.5	3.04	13.5	33.4	<5.5	<5	<6.2	7.1	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Netherlands	2014	NLD (2014)	28.8	14.1	43.0	57.8	8.31	<5	<6.2	<6.2	<6.2	<6.2	<6.2	17.5	<6.2	6.2	<6.2	<6.2	<6.2
Sweden	2019	SWE (2019)	20.8	10.4	31.2	37.4	17.4	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
Switzerland	2016	CHE (2016)	36.3	15.1	51.4	35.3	8.49	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2

Table S 6: Compilation of PFAA concentrations in local samples within the three Great regions from Brazil (pg/g f.w.); sampling year 2012. LOQs are displayed as "<"

Sample_ID	L_PFOS	br_PFOS	PFOS	PFOA	PFHxS	PFBS	PFDS	PFBA	PFPeA	PFHxA	PFHpA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTDA
BRA (AC)	17.1	9.73	26.8	23.0	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (AL)	26.4	12.15	38.5	27.8	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (BA)	54.7	24.5	79.2	53.8	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	11.4	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (GO)	28.0	14.8	42.8	38.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	5.9	8.0	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (MA)	38.2	17.3	55.5	20.2	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (MG)	71.8	38.7	110.5	39.3	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	14.3	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (MT)	15.1	8.80	23.9	40.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	5.9	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (PA)	22.1	8.74	30.8	28.1	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	5.6	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (PB)	21.3	9.61	30.9	20.6	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (PE)	29.2	17.4	46.6	23.4	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (RJ)	42.8	21.1	63.9	29.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	5.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (RO)	13.5	7.63	21.1	29.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (RR)	18.1	8.80	26.9	25.5	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (SC)	34.8	15.7	50.6	30.3	<5.5	<5	<6.2	<6.2	<6.2	<6.2	9.9	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2
BRA (SP)	28.0	13.3	41.3	36.7	<5.5	<5	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2	<6.2

Table S 7: Summary statistics according to 5-year periods (national pools, N=86)

	2005-2009 (N=17)	2010-2014 (N=19)	2015-2019 (N=50)	Overall (N=86)
ΣPFOS				
Mean (SD)	24.8 (15.8)	33.6 (17.6)	20.7 (30.5)	24.4 (26.0)
Median [Min, Max]	23.0 [7.94, 62.7]	28.5 [9.36, 83.3]	14.2 [0, 212]	18.9 [0, 212]
PFOA				
Mean (SD)	26.6 (15.7)	28.0 (10.5)	18.3 (8.99)	22.1 (11.7)
Median [Min, Max]	20.5 [12.8, 63.4]	27.4 [12.6, 57.8]	16.8 [6.20, 48.8]	18.6 [6.20, 63.4]
PFHxS				
Mean (SD)	0.824 (2.34)	4.17 (8.33)	3.24 (15.9)	2.96 (12.8)
Median [Min, Max]	0 [0, 7.56]	0 [0, 34.8]	0 [0, 111]	0 [0, 111]

Table S 8: Summary statistics according to ratification of 2019 amendments with respect to PFOS (UNEP, 2019a) and PFOA (UNEP, 2019b)

	No (N=9)	Yes (N=77)	Overall (N=86)
ΣPFOS			
Mean (SD)	28.1 (21.4)	23.9 (26.6)	24.4 (26.0)
Median [Min, Max]	21.1 [9.18, 62.7]	18.7 [0, 212]	18.9 [0, 212]
PFOA			
Mean (SD)	22.8 (11.9)	22.0 (11.7)	22.1 (11.7)
Median [Min, Max]	19.7 [11.2, 48.8]	18.3 [6.20, 63.4]	18.6 [6.20, 63.4]

Multiple Participation

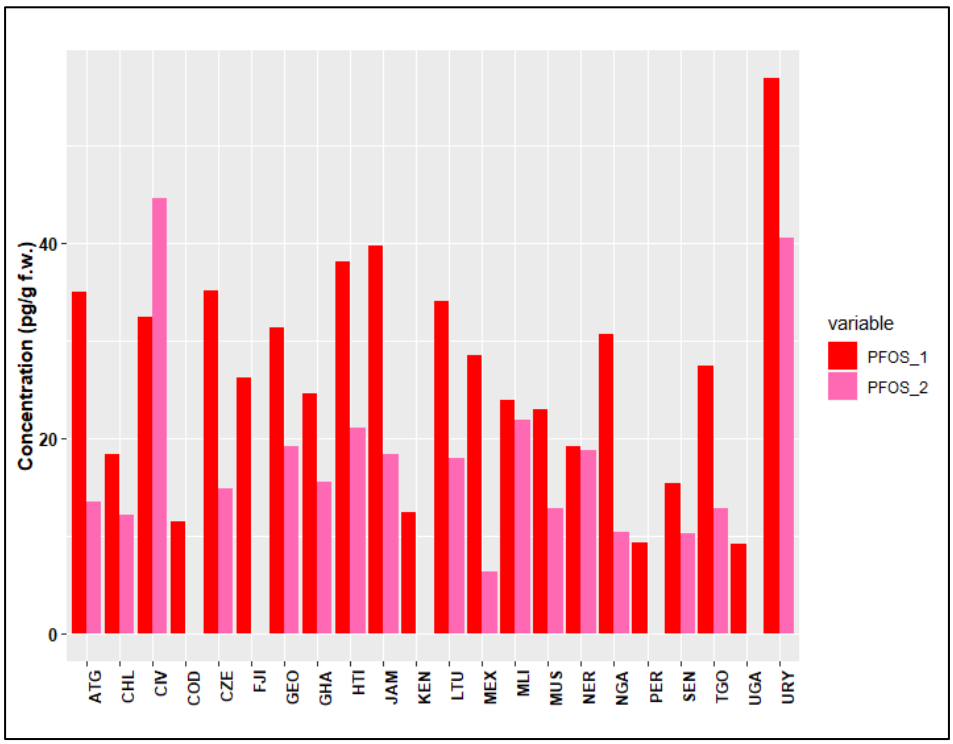

Figure S 2: Barplots for ΣPFOS for countries with two national pools. Older measurement result in strong color (PFOS_1) and second, more recent, measurement in pale color (PFOS_2)

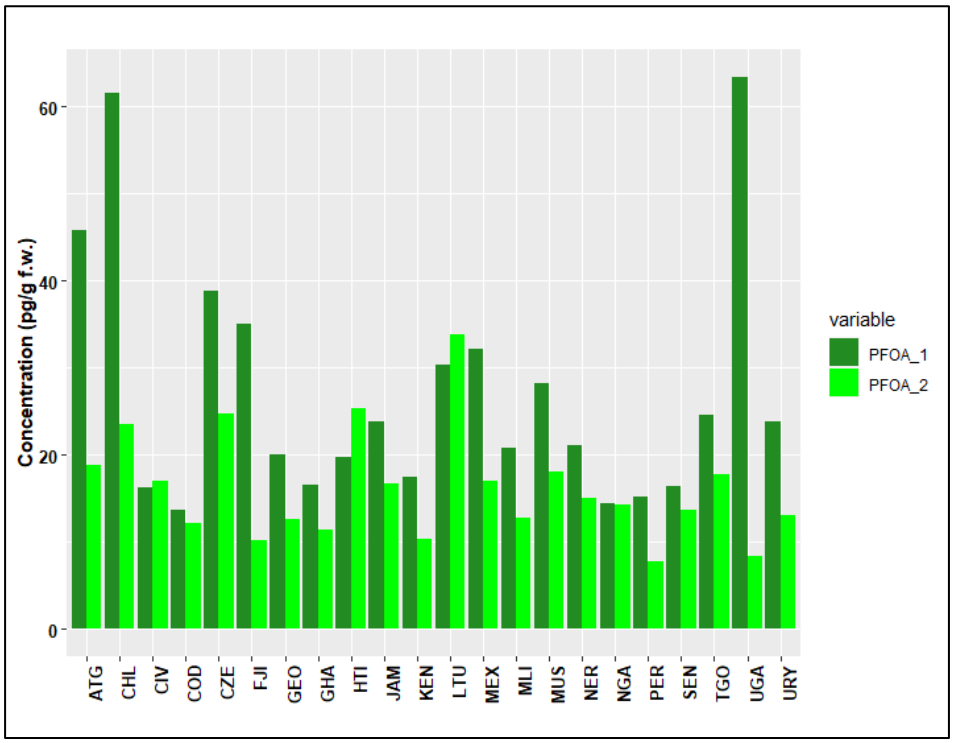

Figure S 3: Barplots for PFOA for countries with two national pools. First measurement result in strong color (PFOA_1) and second, more recent, measurement in pale color (PFOA_2)

Brazil Samples

Table S 9: Overview Brazil samples

	GR1 (N=6)	GR2 (N=6)	GR3 (N=6)	Overall (N=18)
Great region (GR)1	6	6	6	6 (100%)
Type				
GR	1	1	1	3 (16.7%)
Local	5	5	5	15 (83.3%)

Table S 10: Brazil samples: Mean (with SD), median, minimum and maximum values (pg/g f.w.)

	GR1		GR2		GR3		Overall	
	GR (N=1)	L (N=5)	GR (N=1)	L (N=5)	GR (N=1)	L (N=5)	GR (N=3)	L (N=15)
PFOS								
Mean (SD)	26.1 (NA)	25.9 (3.63)	47.5 (NA)	50.1 (18.7)	65.3 (NA)	61.8 (28.6)	46.3 (19.6)	46.0 (24.0)
Median [Min, Max]	26.1 [26.1, 26.1]	26.8 [21.1, 30.8]	47.5 [47.5, 47.5]	46.6 [30.9, 79.2]	65.3 [65.3, 65.3]	50.6 [41.3, 110]	47.5 [26.1, 65.3]	41.3 [21.1, 110]
PFOA								
Mean (SD)	30.6 (NA)	29.3 (6.74)	27.4 (NA)	29.2 (14.1)	34.2 (NA)	34.9 (4.63)	30.8 (3.41)	31.1 (9.14)
Median [Min, Max]	30.6 [30.6, 30.6]	28.1 [23.0, 40.5]	27.4 [27.4, 27.4]	23.4 [20.2, 53.8]	34.2 [34.2, 34.2]	36.7 [29.5, 39.3]	30.6 [27.4, 34.2]	29.5 [20.2, 53.8]
L_PFOS								
Mean (SD)	17.4 (NA)	17.2 (3.27)	32.3 (NA)	34.0 (13.1)	42.7 (NA)	41.1 (18.2)	30.8 (12.7)	30.7 (16.0)
Median [Min, Max]	17.4 [17.4, 17.4]	17.1 [13.5, 22.1]	32.3 [32.3, 32.3]	29.2 [21.3, 54.7]	42.7 [42.7, 42.7]	34.8 [28.0, 71.8]	32.3 [17.4, 42.7]	28.0 [13.5, 71.8]
br_PFOS								
Mean (SD)	8.68 (NA)	8.74 (0.748)	15.2 (NA)	16.2 (5.73)	22.6 (NA)	20.7 (10.5)	15.5 (6.98)	15.2 (8.18)
Median [Min, Max]	8.68 [8.68, 8.68]	8.80 [7.63, 9.73]	15.2 [15.2, 15.2]	17.3 [9.61, 24.5]	22.6 [22.6, 22.6]	15.7 [13.3, 38.7]	15.2 [8.68, 22.6]	13.3 [7.63, 38.7]

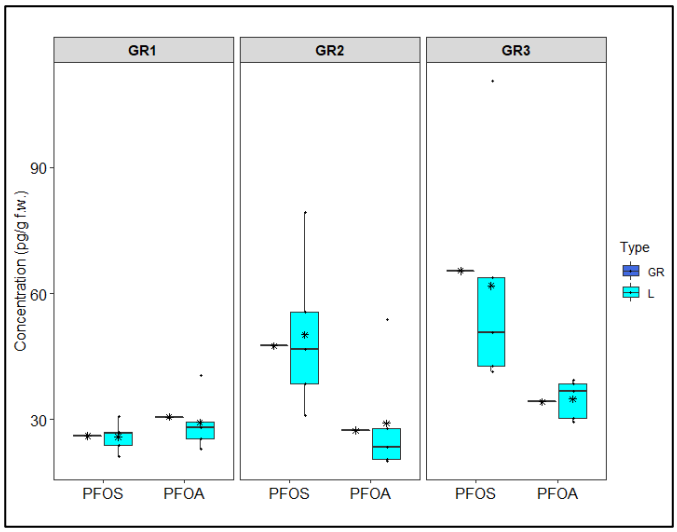

Figure S 4: Box plots, comparison of PFOS and PFOA concentrations in regional *vs*. local samples. Concentrations in pg/g f.w.

The whiskers represent the minimum and maximum concentrations without the outliers. The lower border of the box represents the first quartile (25%), the line inside the box the median and the upper border is the third quartile (75%). The asterisk marks the mean value. The dots outside the whiskers are outliers, which were defined as all concentrations greater or smaller the interquartile range multiplied by 1.5

Lifestyle Factors

Table S 11: Summary statistics according to income using World Bank data
WBC groups: H = high, UM = upper-middle, LM = lower-middle, L = low

	H (N=18)	UM (N=26)	LM (N=25)	L (N=17)	Overall (N=86)
ΣPFOS					
Mean (SD)	24.6 (12.5)	25.0 (20.4)	29.5 (41.2)	15.6 (10.1)	24.4 (26.0)
Median [Min, Max]	19.7 [9.49, 51.4]	22.3 [0, 83.3]	17.2 [0, 212]	12.8 [0, 38.1]	18.9 [0, 212]
PFOA					
Mean (SD)	29.6 (11.6)	22.6 (11.4)	18.3 (9.12)	19.0 (12.6)	22.1 (11.7)
Median [Min, Max]	31.0 [13.1, 57.8]	22.4 [7.81, 61.5]	15.7 [8.62, 48.8]	17.5 [6.20, 63.4]	18.6 [6.20, 63.4]
PFHxS					
Mean (SD)	2.59 (4.86)	1.00 (3.02)	5.06 (22.2)	3.28 (8.57)	2.96 (12.8)
Median [Min, Max]	0 [0, 17.4]	0 [0, 12.9]	0 [0, 111]	0 [0, 34.8]	0 [0, 111]
L_PFOS					
Mean (SD)	18.1 (8.01)	19.2 (15.5)	22.9 (30.1)	11.8 (7.18)	18.6 (19.1)
Median [Min, Max]	15.3 [9.49, 36.3]	16.6 [0, 70.5]	14.6 [0, 154]	11.0 [0, 26.4]	15.2 [0, 154]
br_PFOS					
Mean (SD)	6.54 (4.69)	5.70 (5.91)	6.60 (11.3)	3.83 (3.11)	5.77 (7.34)
Median [Min, Max]	4.99 [0, 15.1]	4.00 [0, 22.6]	4.09 [0, 57.8]	3.16 [0, 11.7]	4.19 [0, 57.8]

Table S 12: Summary statistics according to population density using World Bank data

	A (N=22)	B (N=8)	C (N=22)	D (N=22)	E (N=12)	Overall (N=86)
ΣPFOS						
Mean (SD)	22.5 (16.9)	11.9 (13.0)	21.9 (17.5)	33.0 (43.2)	24.5 (11.5)	24.4 (26.0)
Median [Min, Max]	19.0 [0, 65.3]	8.71 [0, 34.0]	18.2 [0, 83.3]	21.0 [0, 212]	22.0 [12.0, 43.0]	18.9 [0, 212]
PFOA						
Mean (SD)	20.4 (12.4)	21.9 (9.79)	19.8 (9.11)	26.1 (13.5)	22.2 (12.2)	22.1 (11.7)
Median [Min, Max]	16.1 [7.81, 61.5]	19.0 [10.2, 35.0]	17.0 [6.20, 36.3]	22.5 [8.37, 63.4]	19.3 [12.8, 57.8]	18.6 [6.20, 63.4]
PFHxS						
Mean (SD)	1.14 (3.98)	0.721 (2.04)	0.857 (2.97)	6.76 (23.5)	4.72 (10.0)	2.96 (12.8)
Median [Min, Max]	0 [0, 17.4]	0 [0, 5.77]	0 [0, 12.9]	0 [0, 111]	0 [0, 34.8]	0 [0, 111]
L_PFOS						
Mean (SD)	16.5 (11.3)	8.94 (9.21)	18.1 (14.8)	24.7 (31.4)	18.5 (7.84)	18.6 (19.1)
Median [Min, Max]	15.2 [0, 42.7]	8.18 [0, 24.9]	14.8 [0, 70.5]	16.4 [0, 154]	15.6 [7.69, 29.5]	15.2 [0, 154]
br_PFOS						
Mean (SD)	6.08 (5.92)	3.00 (3.97)	3.80 (3.12)	8.30 (12.0)	6.02 (4.23)	5.77 (7.34)
Median [Min, Max]	4.59 [0, 22.6]	0.527 [0, 9.05]	3.78 [0, 12.8]	4.28 [0, 57.8]	5.46 [0, 14.1]	4.19 [0, 57.8]

MULTIVARIATE ANALYSIS

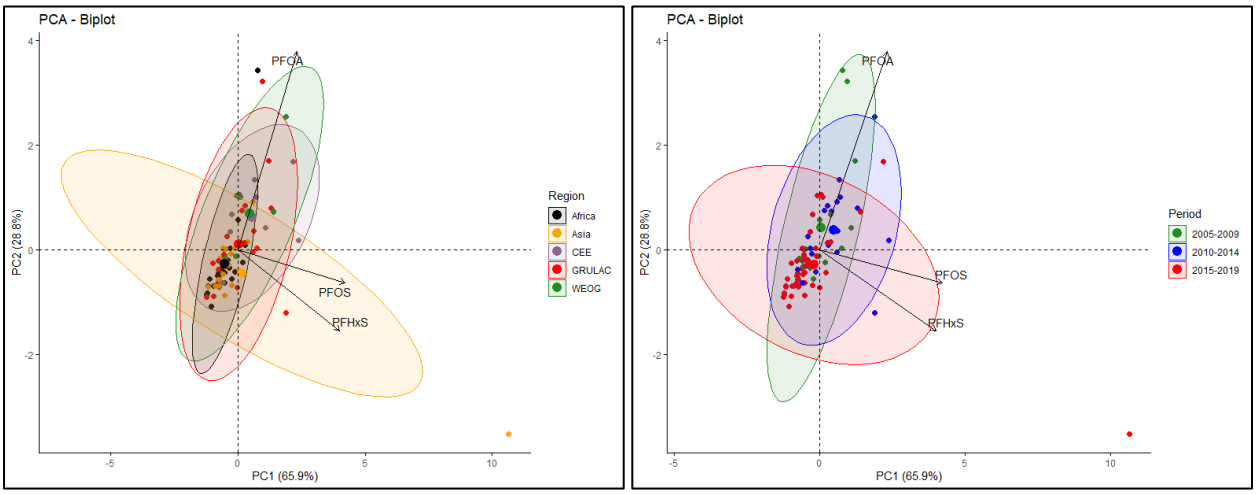

Figure S 5: PCAs of the 86 national pools assessing PFOS, PFOA and PFHxS with ellipses around the regions (left) and the periods (right)

REFERENCES

Fiedler, H., and Sadia, M. (2021). Regional occurrence of perfluoroalkane substances in human milk for the global monitoring plan under the Stockholm Convention on Persistent Organic Pollutants during 2016–2019. Chemosphere *277*, 130287.

UNEP (2019a). Decision SC-9/4: Perfluorooctane sulfonic acid, its salts and perfluorooctane sulfonyl fluoride. In SC-9/4, United Nations Environment Programme (UNEP), ed. (Geneva, Switzerland: Conference of the Parties to the Stockholm Convention on Persistent Organic Pollutants).

UNEP (2019b). Decision SC-9/12: Listing of perfluorooctanoic acid, its salts and PFOA-related compounds. In SC-9/12 (Geneva, Switzerland: UN Environment, Conference of the Parties to the Stockholm Convention on Perssisent Organic Pollutants,).

UNEP, Fiedler, H., Van der Veen, I., and de Boer, J. (2021). Bi-ennial Global Interlaboratory Assessment on Persistent Organic Pollutants – Fourth Round 2018/2019, United Nations Environment Programme (UNEP), ed. (Geneva, Switzerland), pp. 145.

US-EPA (2019). Method 533: Determination of per- and polyfluoroalkyl substances in drinking water by isotope dilution anion exchange solid phase extraction and liquid chromatographt/tandem mass spectrometry. In Office of Water (MS-140) (Cincinnati, OH (USA): EPA Document No. 815-B-19-020).

US-EPA (2020). Method 537.1 Determination of selected per- and polyfluorinated alkyl substances in drinking water by solid phase extraction and liquid chromatography/tandem mass spectrometry (LC/MS/MS). Version 2.0 March 2020. In EPA Document #: EPA/600/R-20/006 (CINCINNATI, OHIO 45268: Center for Environmental Solutions & Emergency Response, Office of Research and Development, U. S. Environmental Protection Agency).

World Bank (No Year-a). GNI per capita, Atlas method (current US$).

World Bank (No Year-b). Population density (people per sq. km of land area), World Bank, ed.

World Bank (No Year-c). World Development Indicators. In Data catalog.