Supporting Information

Are melamine and its derivatives the alternatives for per- and polyfluoroalkyl substance (PFAS) fabric treatments in infant clothes?

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Chemicals. $^{13}C_3$-Melamine ($^{13}C_3$-MEL), 1,3,5-triallyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione (TTT), 2,4,6-triallyloxy-1,3,5-triazine (TALT), and 2,4-diamino-6-phenyl-1,3,5-triazine (DPT) were obtained from Sigma Aldrich (St. Louis, MO, USA). 2-Amino-4-methoxy-6-methyl-1,3,5-triazine (AMMT), $^{15}N_3$-MEL ($^{15}N_3$-MEL), 2,4,6-*tris*[*bis*(methoxymethyl)amino]-1,3,5-triazine (TBMMAT), 1,3,5-*tris*(3,5-di-tert-butyl-4-hydroxybenzyl)-s-triazine-2,4,6(1H,3H,5H)-trione (TDTBHI), dichloroisocyanuric acid (DICASS), and $^{13}C_3$-cyanuric acid ($^{13}C_3$-CYA) were purchased from Toronto Research Chemicals. MEL, ammeline (AMN), ammelide (AMD), and cyanuric acid (CYA) were purchased from AccuStandard (New Haven, CT, USA). $^{13}C_3$-$^{15}N_3$-Cyanuric acid ($^{13}C_3$-^{15}N-CYA) was obtained from Cambridge Isotope Laboratories (Andover, MA, USA). All target PFAS compounds were obtained from Wellington Laboratories (Guelph, ON, Canada). All individual standards had a purity ≥ 98% and all solvents were Optima grade. The complete list of the target compounds, their abbreviations, and the surrogate and internal standards are listed in Tables S1 and S2.

Instrumental methods for PFAS analyses. The analysis of ionic PFAS was performed on an UPLC-QQQ MS (Agilent 1290 Infinity II UPLC – 6470 QQQ-MS) with negative electrospray ionization (ESI-). The nebulizer, gas flow, gas temperature, capillary voltage, sheath gas temperature and sheath gas flow were set as 25 psi, 10 L/min, 300 °C, 2800 V, 330 °C and 11 L/min, respectively. The mobile phases consisting of 2 mM ammonium acetate in water (*A*) and 2 mM ammonium acetate in methanol (*B*) were used with gradient elution. The gradient was as follows: 0 min, 10% *B*; 0.5 min, 10% *B*; 1 min, 40% *B*; 17.5 min, 100% *B*. The instrument was equilibrated for 3.5 min after every run. The injection volume was 5 µL. UPLC separation was achieved on an Acquity UPLC BEH C18 column (50 mm, 2.1 mm i.d., 1.7 µm thickness, Waters, Milford, MA), and column temperature was 40 °C. Data acquisition was performed using multiple

reaction monitoring (MRM) mode and the optimized MRM transitions are summarized in Table S1.

Neutral PFAS were analyzed on an Agilent 7890 gas chromatograph (GC) coupled to an Agilent 5975C mass spectrometer (MS) in the electron capture positive ionization (PCI) mode. The mass spectrometer ion source, quadrupole temperatures, GC/MS transfer line and the injection temperature, were maintained at 200, 106, 200 and 200 °C, respectively. A splitless injector was used and the injection volume was 2 μL and the carrier gas was helium at a constant flow rate of 1 mL/min. Separation was achieved on a CP-WAX 57 CB (25 m, 250 μm i.d., and 0.2 μm film thickness) fused silica capillary GC column (Agilent J&W). The initial oven temperature was set at 60 °C, held for 3 min, then increased to 85 °C at 25 °C/min, then increased to 190 °C at 3 °C/min, and kept for 8 min. The target compounds were analyzed through selected ion monitoring (SIM) mode and the optimized SIM transitions are summarized in Table S2.

Table S1. The optimized MRM transitions, fragmentors, and collision energies for melamine (MEL)-based compounds and PFAS, surrogate and internal standards analyzed under ESI (+) mode and ESI (-) mode. IS: Internal standard; SS: surrogate standard.

Abbr.	Compound Name	Precursor ion	Fragmentor (volts)	Product ions (m/z)	Collision energy (volts)
ESI (+)					
MEL	Melamine	127	90	85	25
				68	37
AMN	Ammeline	128	90	86	25
				69	53
DPT	2,4-Diamino-6-phenyl-1,3,5-triazine	188	107	104	25
				77	57
				43	69
TTT	1,3,5-Triallyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione	250.1	45	81.1	9
				41.2	33
TALT	2,4,6-Triallyloxy-1,3,5-triazine	250.1	50	81.1	5
				41.2	33
TBMMAT	2,4,6-*Tris*[*bis*(methoxymethyl)amino]-1,3,5-triazine	391.2	78	359.2	5
				177.1	33
				45.1	73
TDTBHI	*Tris*(3,5-di-tert-butyl-4-hydroxybenzyl) isocyanurate	784.5	185	497.1	10
				219	17
AMMT	2-Amino-4-methoxy-6-methyl-1,3,5-triazine	141.1	98	57.1	21
				43.1	45
DICASS	Dichloroisocyanuric acid	197.9	152	166.1	17
				124.8	21
				93	65
				178.1	13
$^{13}C_3$-MEL (SS)	Melamine-$^{13}C_3$	130	90	87	25
				70	37
$^{15}N_3$-MEL (IS)	Melamine-$^{15}N_3$	130.1	107	69.1	41
				44.1	53
ESI (-)					
AMD	Ammelide	127	90	84	33
				42	73
CYA	Cyanuric Acid	128	90	85	33
				42	73
$^{13}C_3$-$^{15}N_3$-CYA (SS)	Cyanuric acid-$^{13}C_3$-$^{15}N_3$	134	90	89	33
				44	73
$^{13}C_3$-CYA (IS)	Cyanuric acid-$^{13}C_3$	131	90	87	5
				43	20
PFBA	Perfluorobutanoic acid	213.0	64	169	5
				\	\
PFPeA	Perfluoropentanoic acid	263.0	64	218.9	5
				140.8	5
PFHxA	Perfluoro-*n*-hexanoic acid	313.0	73	268.9	5
				119	21
PFHpA	Perfluoro-*n*-heptanoic acid	363.0	78	319	5
				169	17
PFOA	Perfluoro-*n*-octanoic acid	413.1	83	369	5
				169	17
PFNA	Perfluoro-*n*-nonanoic acid	463.1	83	419	5
				218.9	17
PFDA	Perfluoro-*n*-decanoic acid	513.0	93	468.9	5
				269	17

PFUdA	Perfluoro-*n*-undecanoic acid	563.0	102	518.9	5
				268.9	17
PFDoA	Perfluoro-*n*-dodecanoic acid	613.0	102	569	9
				269	21
PFTrDA	Perfluoro-*n*-tridecanoic acid	663.1	107	619	9
				169	29
PFTeDA	Perfluoro-*n*-tetradecanoic acid	713.1	112	668.9	13
				169	29
PFHxDA	Perfluoro-*n*-hexadecanoic acid	813.1	121	768.9	13
				168.9	37
PFPrS	Perfluoro-1-propanesulfonic acid	249.1	140	80	37
				98.9	33
PFBS	Perfluoro-1-butanesulfonic acid	299.0	149	80	37
				98.9	37
PFPeS	Perfluoro-1-pentanesulfonic acid	349.0	175	80	45
				98.9	37
PFHxS	Perfluoro-1-hexanesulfonic acid	399.0	179	80	45
				98.9	41
PFHpS	Perfluoro-1-heptanesulfonic acid	449.0	183	80	49
				98.9	45
PFOS	Perfluoro-1-octanesulfonic acid	499.0	208	80	101
				98.9	49
PFNS	Perfluoro-1-nonanesulfonic acid	549.0	218	80	105
				98.9	49
PFDS	Perfluoro-1-decanesulfonic acid	598.9	232	80	137
				98.9	53
				98.9	29
FOSA	Perfluoro-1-octanesulfonamide	498.0	169	78	37
				48.1	150
MeFOSA	N-methylperfluoro-1-octanesulfonamide	512.0	160	169	29
				218.9	25
EtFOSA	N-ethylperfluoro-1-octanesulfonamide	526.0	165	169	29
				219	29
4:2 FTS	1H,1H,2H,2H-perfluorohexane sulfonic acid (4:2)	327.1	136	306.9	21
				81	33
6:2 FTS	1H,1H,2H,2H-perfluorooctane sulfonic acid (6:2)	427.0	164	406.9	25
				81	41
8:2 FTS	1H,1H,2H,2H-perfluorodecane sulfonic acid (8:2)	527.0	179	506.9	29
				81	41
M3PFBA (SS)	Perfluoro-*n*-[2,3,4-$^{13}C_3$]butanoic acid	216.0	64	172	5
MPFHxA (SS)	Perfluoro-*n*-[1,2-$^{13}C_2$]hexanoic acid	315.1	78	270	5
MPFOA (SS)	Perfluoro-*n*-[1,2,3,4-$^{13}C_4$]octanoic acid	417.1	83	372	5
MPFUdA (SS)	Perfluoro-*n*-[1,2-$^{13}C_2$]undecanoic acid	565.1	97	520	9
M2PFTeDA (SS)	Perfluoro-*n*-[1,2-$^{13}C_2$]tetradecanoic acid	715.1	116	669.9	13
M3PFBS (SS)	Perfluoro-1-[2,3,4-$^{13}C_3$]butanesulfonic acid	302.0	149	80	45
MPFHxS (SS)	Perfluoro-1-hexane[$^{18}O_2$]sulfonic acid	403.0	169	84	49
MPFOS (SS)	Perfluoro-1-[1,2,3,4-$^{13}C_4$]octanesulfonic acid	503.0	198	80	93
d-N-MeFOSA (SS)	N-methyl-d_3-perfluoro-1-octanesulfonamide	515.0	160	169	29
MPFBA (IS)	Perfluoro-*n*-[1,2,3,4-$^{13}C_4$]butanoic acid	217.0	64	172	5
M8PFOA (IS)	Perfluoro-*n*-[$^{13}C_8$]octanoic acid	421.1	83	376	5
M7PFUdA (IS)	Perfluoro-*n*-[1,2,3,4,5,6,7-$^{13}C_7$]undecanoic acid	570.0	97	525	9
M3PFHxS (IS)	Perfluoro-1-[1,2,3-$^{13}C_3$]hexanesulfonic acid	402.0	184	80	45
M8PFOS (IS)	Perfluoro-[$^{13}C_8$]octanesulfonic acid	507.0	203	79.9	97

Table S2. Instrumental details for the analysis of PFAS analyzed using gas chromatographic mass spectrometry in the positive chemical ionization mode.

Abbreviation	Compound Name	Quantifier	Qualifier
4:2 FTOH	2-Perfluorobutyl ethanol (4:2)	265	227
6:2 FTOH	2-Perfluorohexyl ethanol (6:2)	365	327
8:2 FTOH	2-Perfluorooctyl ethanol (8:2)	465	427
10:2 FTOH	2-Perfluorodecyl ethanol (10:2)	565	527
6:2 FTAcr	1H,1H,2H,2H-perfluorooctyl acrylate	419	399
8:2 FTAcr	1H,1H,2H,2H-Perfluorodecyl acrylate	519	499
10:2FTAcr	1H,1H,2H,2H-Perfluorododecyl acrylate	619	599
6:2 FTMAcr	1H,1H,2H,2H-perfluorooctyl methacrylate	433	413
8:2 FTMAcr	1H,1H,2H,2H-heptadecafluorodecyl methacrylate	533	513
MeFOSE	2-(N-methylperfluoro-1-octanesulfonamido)-ethanol	558	540
EtFOSE	2-(N-ethylperfluoro-1-octanesulfonamido)-ethanol	572	554
M2FOET (SS)	2-Perfluorooctyl-[1,2-$^{13}C_2$]-ethanol(8:2)	467	429
MFOET (IS)	2-Perfluorooctyl-[1,1-$^{2}H_2$]-[1,2-$^{13}C_2$]-ethanol(8:2)	469	431

Table S3. The details of samples analyzed in this study, including the fabric and clothing type, country of manufacture, and production year.

Fabric (composition)	Clothing type	Country	Production year
Conventional cotton (*n* = 19)			
cotton (100)	Coat	Thailand	2018
cotton (100)	Dress	Thailand	2018
cotton (100)	Onesie	Vietnam	2019
cotton (100)	Onesie	India	2017
cotton (100)	Onesie	India	2018
cotton (100)	Onesie	India	2018
cotton (100)	Onesie	India	2019
cotton (100)	Onesie	India	2017
cotton (100)	Onesie	India	2018
cotton (100)	Onesie	Cambodia	2018
cotton (100)	Onesie	Cambodia	2019
cotton (100)	Onesie	Cambodia	2017
cotton (100)	Onesie	Cambodia	2018
cotton (100)	Onesie	Cambodia	2018
cotton (100)	Onesie	Cambodia	2019
cotton (100)	Pants	Vietnam	2017
cotton/other fiber (97/3)	Shirt	Vietnam	2018
cotton (100)	Shirt	Indonesia	2018
cotton (100)	Shirt	Thailand	2019
Organic cotton (*n* = 10)			
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
organic cotton (100)	Diaper	United States	Not available
Cotton/polyester (*n* = 20)			
cotton/polyester (60/40)	Dress	Vietnam	2018
cotton/polyester (60/40)	Onesie	Vietnam	2018
cotton/polyester/spandex (65/34/11)	Onesie	Indonesia	2018
cotton/polyester (60/40)	Onesie	China	2019
cotton/polyester (60/40)	Onesie	Vietnam	2017
cotton/polyester/spandex (57/38/5)	Onesie	Vietnam	2018
cotton/polyester/spandex (58/38/4)	Pants	China	2018
cotton/polyester (60/40)	Pants	China	2018
cotton/polyester/spandex (57/38/5)	Pants	China	2019
cotton/polyester (60/40)	Pants	China	2017
cotton/polyester/spandex (57/38/5)	Pants	China	2018
cotton/polyester (60/40)	Shirt	China	2018
cotton/polyester (60/40)	Shirt	Indonesia	2019
cotton/polyester (60/40)	Shirt	Indonesia	2017
cotton/polyester (60/40)	Shirt	Nicaragua	2018

Fabric (composition)	Clothing type	Country	Production year
cotton/polyester (60/40)	Shirt	Nicaragua	2018
cotton/polyester (60/40)	Shirt	Nicaragua	2018
cotton/polyester (60/40)	Shirt	Nicaragua	2019
cotton/polyester (60/40)	Shirt	Nicaragua	2017
cotton/polyester (60/40)	Shirt	Indonesia	2018
Cotton/spandex (*n* = 18)			
cotton/spandex (95/5)	Onesie	Vietnam	2018
cotton/spandex (95/5)	Pants	Vietnam	2019
cotton/spandex (97/3)	Pants	Salvador	2017
cotton/spandex (97/3)	Pants	Salvador	2018
cotton/spandex (97/3)	Pants	Salvador	2018
cotton/polyester (60/40)	Shirt	Salvador	2018
cotton/spandex (95/5)	Socks	China	2019
cotton/spandex (95/5)	Socks	China	2017
cotton/spandex (95/5)	Socks	China	2018
cotton/spandex (95/5)	Socks	China	2018
cotton/spandex (95/5)	Socks	China	2018
cotton/spandex (95/5)	Socks	China	2019
cotton/spandex (95/5)	Socks	China	2017
cotton/spandex (95/5)	Socks	China	2018
cotton/spandex (95/5)	Socks	China	2018
cotton/spandex (95/5)	Socks	China	2019
cotton/spandex (95/5)	Socks	China	2017
cotton/spandex (95/5)	Socks	China	2018
Polyester (*n* = 11)			
polyester (100)	Diaper	China	2018
polyester (100)	Diaper	China	2019
polyester (100)	Diaper	China	2018
polyester (100)	Diaper	China	2019
polyester (100)	Diaper	China	2017
polyester (100)	Diaper	China	2018
polyester (100)	Diaper	China	2018
polyester (100)	Diaper	China	2018
polyester (100)	Diaper	China	2019
polyester (100)	Diaper	China	2017
polyester/rayon/spandex (62/33/5)	Onesie	China	2018
Nylon (*n* = 8)			
nylon (100)	Coat	Vietnam	2018
nylon (100)	Coat	China	2019
nylon/spandex (80/20)	Onesie	China	2017
nylon/spandex (80/20)	Onesie	China	2018
nylon/spandex (80/20)	Onesie	China	2018
nylon/spandex (80/20)	Onesie	China	2019
nylon/spandex (80/20)	Pants	China	2017
nylon/spandex (80/20)	Pants	China	2018

Table S4. Average recoveries of MEL and PFAS surrogate standards (Tables S1 and S2) in different fabrics (mean ± standard error, %).

	cotton	organic cotton	cotton/polyester	polyester	cotton/spandex	Nylon
C_{13}-MEL	113 ± 2.2	90 ± 5.4	114 ± 3.1	100 ± 4.5	104 ± 3.1	99 ± 4.1
C_{13}-N_{15}-CYA	115 ± 2.4	96 ± 4.2	111 ± 3.5	97 ± 4.0	111 ± 3.5	87 ± 5.3
M3PFBA	90 ± 3.8	102 ± 6.3	86 ± 2.3	96 ± 4.0	84 ± 2.3	105 ± 3.8
M3PFBS	68 ± 1.4	78 ± 1.9	69 ± 1.7	65 ± 1.9	67 ± 1.7	74 ± 1.7
MPFHxA	74 ± 2.3	73 ± 2.3	77 ± 2.0	69 ± 1.6	68 ± 2.0	69 ± 1.7
MPFHxS	68 ± 2.6	84 ± 0.9	83 ± 1.8	82 ± 1.1	71 ± 1.8	88 ± 1.6
MPFOA	99 ± 2.9	93 ± 1.0	101 ± 2.8	92 ± 1.1	88 ± 2.8	94 ± 0.8
MPFOS	95 ± 2.6	101 ± 1.2	95 ± 2.4	98 ± 1.4	88 ± 2.4	100 ± 1.3
MPFUdA	79 ± 2.1	90 ± 0.9	78 ± 1.5	84 ± 0.9	75 ± 1.5	88 ± 1.2
dMeFOSA	83 ± 3.0	53 ± 0.6	81 ± 5.0	19 ± 8.8	58 ± 5.0	62 ± 3.4
M2PFTeDA	115 ± 5.7	81 ± 1.0	110 ± 6.5	110 ± 3.3	91 ± 6.5	86 ± 2.6
M2FOET	98 ± 4.6	85 ± 3.5	90 ± 5.1	87± 5.7	84 ± 5.1	87 ± 4.2

Table S5. Median concentrations of MEL derivatives and other emerging chemicals in textiles reported in previous publications.

Textile type	Compounds	Median	Reference
Infant clothing	MEL	35.7	This study
	AMN	1,530	
	AMD	28.2	
	CYA	77.6	
	TBMMAT	0.451	
Raw textiles and infant clothing	MEL	53	Zhu et al., 2020[1]
	AMN	3.42	
	AMD	4.04	
	CYA	43.5	
New and used clothing	Bisphenol-A	26.9	Wang et al., 2019[2]
	Bisphenol-S	7.38	
Pantyhose	Bisphenol-S	1,430	Li and Kannan, 2018[3]
Infant cotton clothing	Di-(2-ethylhexyl) phthalate	2,740	Li et al., 2019[4]
	Dibutyl phthalate	1,510	
	Di-iso-butyl phthalate	690	
Raw textiles and infant clothing	Benzothiazoles	51.1	Liu et al., 2017[5]

Table S6. Median concentrations of MEL-based compounds and PFAS in different types of clothes (ng/g); n.d.: not detected.

	MEL	CYA	AMD	AMN	TBMMAT	ΣMEL	ΣIonic PFAS	ΣNeutral PFAS	ΣPFAS
Outerwear (*n* = 3)	38200	471	453	76,000	126	70,800	3.31	93.8	4.53
Diapers (*n* = 20)	14.0	5,440	42.1	128	0.084	4,570	3.87	17.3	3.93
Dresses (*n* = 2)	26.2	336	11.1	n.d.	0.254	374	2.7	n.d.	2.7
Onesies (*n* = 24)	58.8	48.8	8.29	2,120	0.561	166	3.35	85.1	3.4
Pants (*n* = 12)	69.0	10.1	18.1	1,550	0.402	78.2	3.46	140	3.61
Shirts (*n* = 13)	8.60	7.17	3.40	221	0.162	8.83	3.42	n.d.	3.42
Socks (*n* = 12)	7.45	340	77.0	262	n.d.	23.9	2.71	10.4	3.41

Table S7. Median concentrations of MEL-based compounds and PFAS in clothes manufactured in different countries (ng/g). n.d.: not detected.

	MEL	CYA	AMN	AMD	TBMMAT	ΣMEL	Σ Ionic PFAS	ΣNeutral PFAS	ΣPFAS
Cambodia (*n* = 6)	35.9	96.1	11.8	n.d.	0.616	57.0	3.05	n.d.	3.05
China (*n* = 38)	58.0	86.8	134	5,550	0.276	36.9	3.40	27.1	3.78
India (*n* = 6)	421	51.2	1.47	708	1.88	790	3.46	n.d.	3.46
Indonesia (*n* = 5)	43.7	26.7	n.d.	221	0.367	87.1	4.06	n.d.	4.06
Nicaragua (*n* = 5)	5.72	4.48	3.40	n.d.	n.d.	7.57	2.79	n.d.	2.79
Salvador (*n* = 4)	56.0	19.3	n.d.	n.d.	n.d.	59.0	3.50	n.d.	3.50
Thailand (*n* = 2)	44.9	255	3.46	n.d.	0.254	304	2.10	n.d.	2.10
United States (*n* = 10)	20.8	5,990	42.1	n.d.	n.d.	6,120	4.02	n.d.	4.02
Vietnam (*n* = 10)	36.7	28.9	17.1	61,100	0.597	52.1	3.45	93.8	3.54

Table S8. Estimated daily intakes (EDIs; pg/kg bw/day) of MEL and CYA via dermal absorption from clothing (based on median concentrations), breastfeeding, consumption of infant formula, and ingestion of dust for 0-1, 1-3, 3-6, 6-12 months old infants.

		0-1 mo.	1-3 mo.	3-6 mo.	6-12 mo.
Absorption from non-nylon clothes	MEL	0.0138	0.0127	0.0117	0.0112
	CYA	0.0292	0.0269	0.0247	0.0236
Absorption from nylon clothes	MEL	8.59	7.91	7.26	6.94
	CYA	0.0851	0.0784	0.072	0.0688
Breastfeeding	MEL	30.6	28.6	22.4	16.9
	CYA	161	150	118	88.8
Consumption of infant formula	MEL	60.8	67.3	52.5	28.2
	CYA	473	523	409	219
Ingestion of dust	MEL	34.3	34.3	34.3	34.3
	CYA	8.29	8.29	8.29	8.29

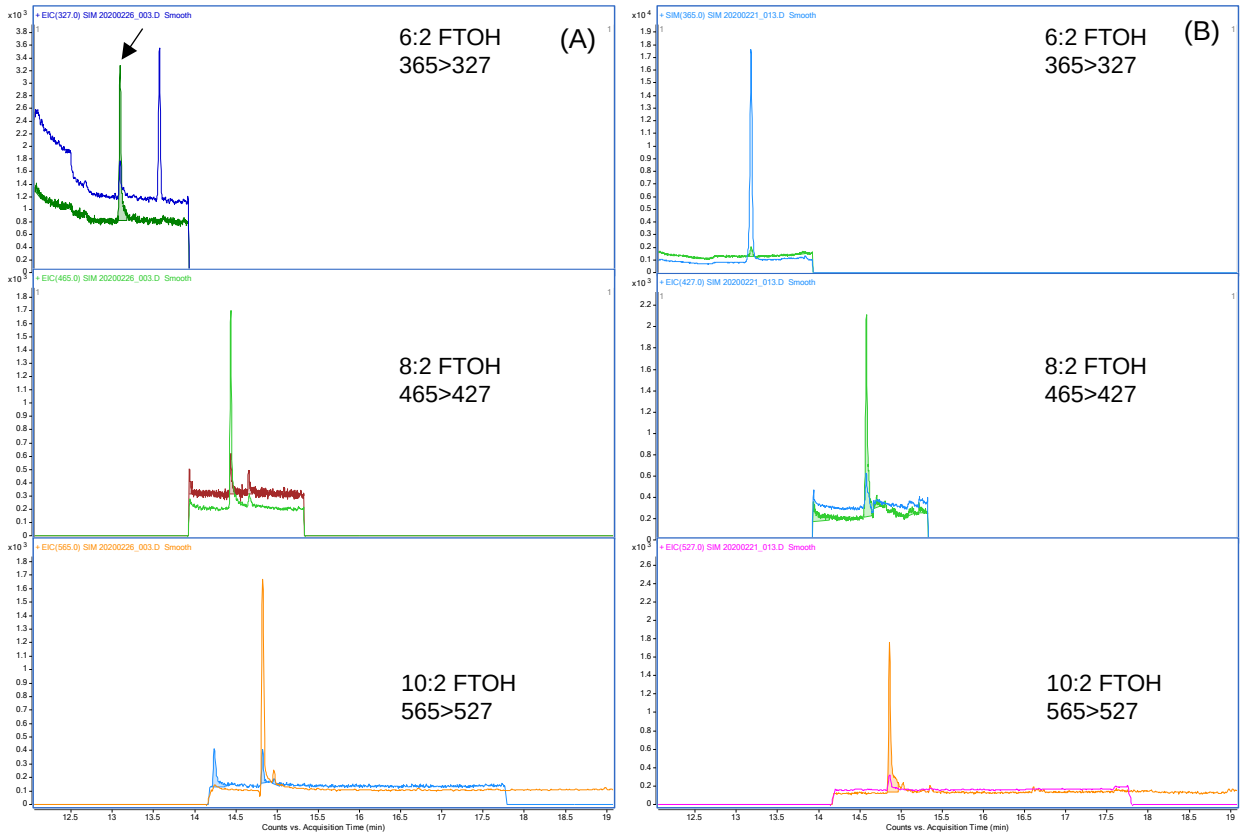

Figure S1. Selected chromatograms of FTOHs in the authentic standard (A, 20 ppb) and in a sample (B).

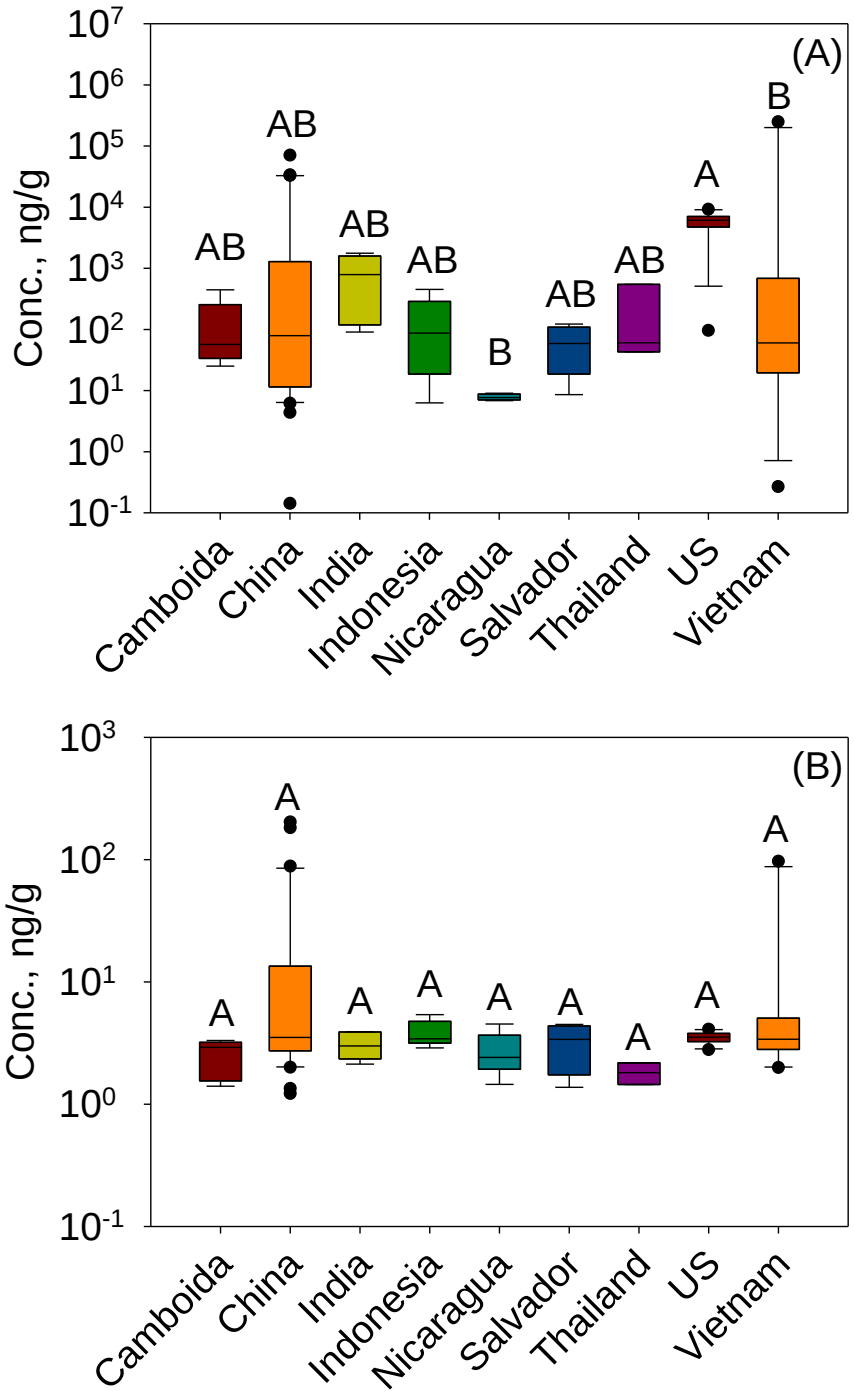

Figure S2. Concentrations of ∑MEL (A) and ∑PFAS (B) in clothes manufactured in different countries (ng/g). Concentrations are shown as boxplots representing the 25th and 75th percentiles; black lines represent the median; and the whiskers represent the 10th and 90th percentiles. The letters represent the results of the analysis of variance (ANOVA); boxes sharing the same letters are not significantly different at $p < 0.05$.

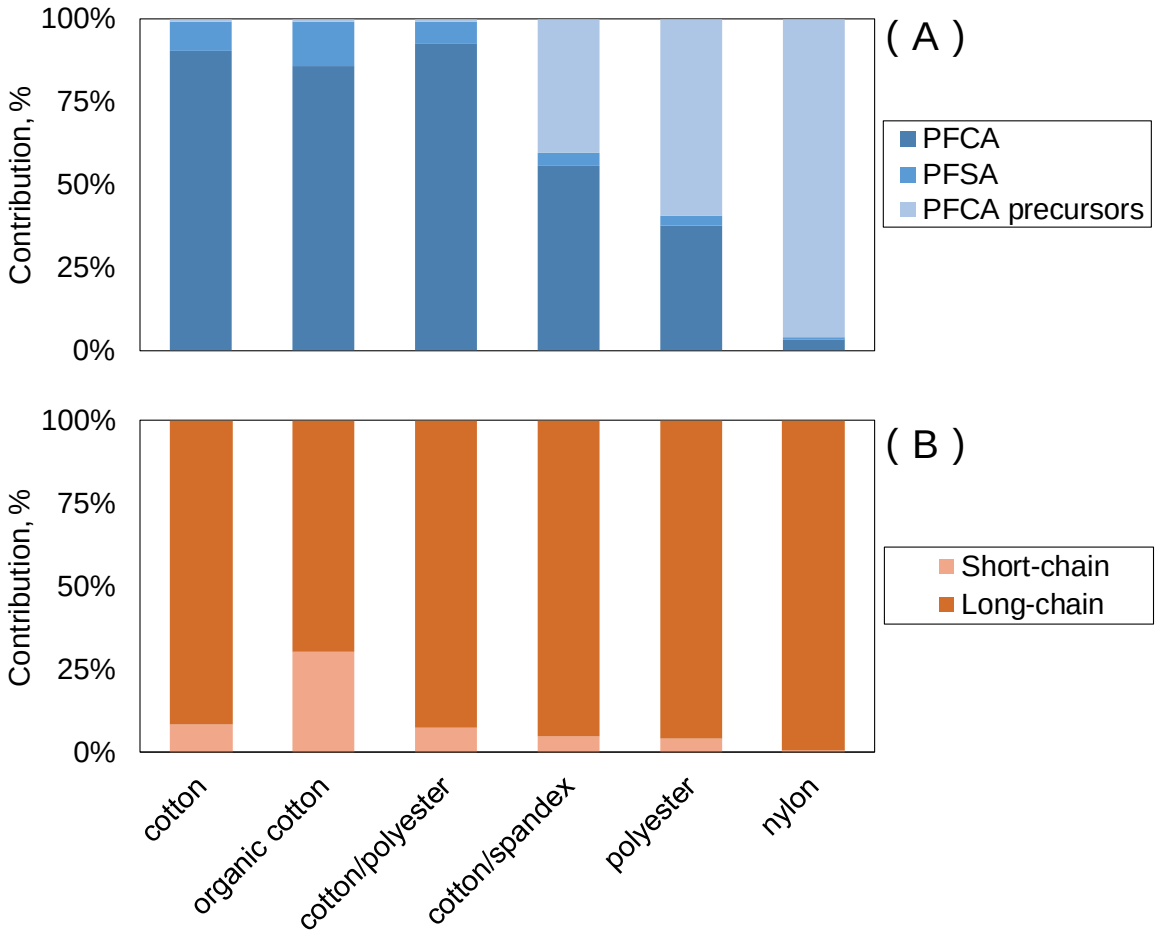

Figure S3. Composition of PFAS measured in different fabrics: (A) shows the contributions of PFCAs, PFSAs, and PFCA precursors (6:2 FTOH, 8:2 FTOH, and 10:2 FTOH) to ∑PFAS concentrations; (B) shows the contributions of short- (< C7) and long-chain PFAS (⩾ C8) to ∑PFAS concentrations.

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