Supporting Information

Chlorinated Paraffins in Human Milk from Urban Sites in China, Sweden, and Norway

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Methods and Materials

Sample storage

All samples were collected 2-12 weeks after giving birth. The target volume was at least 50 mL from each mother. Samples were subdivided into several portions and stored in the freezer below -18°C in glass bottles until chemical analysis. The Swedish samples were initially stored at -18ºC in plastic bags and bottles. Upon being shipped to the Environmental Specimen Bank at the Swedish Museum of Natural History in Stockholm, samples were then thawed, transferred to pre-washed glass bottles with lids covered with aluminum foil, and stored at -20ºC prior to analysis.

Sample extraction and clean up

The denaturation and extraction method followed Fängström, et al. [1] with some minor modifications. $^{13}C_{10}$-1,5,5,6,6,10-hexachlorodecane (10 ng) was spiked as surrogate standard to the human milk samples (about 5 g) in a test tube (18 mL). The samples were homogenized with a solvent mixture of formic acid (6 M, 1 mL), isopropanol (6 mL) and diethyl ether mixed with n-hexane (6 mL, 1:1 v/v). After centrifugation for 5 minutes at 3000 rpm, the organic phase was transferred to a new tube (18 mL). The aquatic phase was extracted once more with isopropanol (1.5 mL) and diethyl ether/n-hexane (3 mL, 1:1 v/v). The combined organic phase was further washed twice with a sodium chloride solution (4 mL, 0.9%), whereas the aquatic phase was removed to a 15 mL tube. The combined aquatic phase was further extracted with diethyl ether/n-hexane (3 mL, 1:1 v/v) to get the analytes recovered to the organic phase. Finally, the combined organic phase was transferred to a pre-weighted beaker and lipid content was measured gravimetrically.

The lipids were dissolved with n-hexane (4 mL) in a test tube. Sulfuric acid (4 mL) was added to remove the lipids. The upper organic phase was taken out after inverting and centrifugation. The lower acid phase was partitioning once more with 4 mL n-hexane. The combined organic phase was concentrated to about 0.5 mL by a gentle nitrogen flow. A multi-layer column prepared in a Pasteur pipette packaged with activated silica gel (0.1) and silica gel impregnated with sulfuric acid (0.9 g, 2:1 w/w) from bottom to top was applied to further clean up. The column was preconditioned with n-hexane (3 mL), afterwards, the sample was applied to the column and eluted with n-hexane/dichloromethane (15 mL, 1:1 v/v). The collected solvent was evaporated to 1 mL by nitrogen flow. Silica gel column (deactivated with 3% water w/w, 4g, 0.063-0.2 mm Kiselgel 60) was applied to separate CPs from other nonpolar POPs described in detail elsewhere.[2] The column was conditioned by n-hexane (30 mL). The first fraction eluted with n-hexane (28 mL) was discarded to remove nonpolar POPs such as PCBs and HCB. The second fraction containing CPs was collected with n-hexane/diethyl ether (30 mL, 3:1 v/v). Prior to instrumental analysis, Dechlorane 603 (10 ng) was added to samples as volumetric standard. Three empty tubes were prepared as field blanks in China and tested for possible contamination. One procedural blank was analyzed in parallel with each batch of five samples to assess any potential contamination through the laboratory work.

Instrumental analysis of CPs

The recovery of $^{13}C_{10}$-1,5,5,6,6,10-hexachlorodecane was analyzed by a Varian 450 gas chromatograph equipped with an electron capture detector (GC-ECD) and a Varian CP-8400 auto-sampler. Injections (1 µL) were performed on a programmable temperature vaporizing (PTV) injector operating in splitless mode at a temperature of 250 °C. CP-Sil 5CB column (15 m x 0.25 mm i.d. x 0.25µm film thickness; Varian capillary column) was used. Helium and nitrogen were used as carrier gas and make-up gas, respectively. The column oven temperature program was set as: 80 °C hold for 2 min, 15 ºC/min to 300ºC and hold for 8 min. Native CPs in human milk samples were analyzed using direct injection dichloromethane-enhanced APCI-QTOF-MS (QTOF Premier, Waters, UK) with same instrumental settings described in a previous study.[3] We screened chloride adduct ions of 280 CP congener groups from $C_9H_{17}Cl_3$ to $C_{31}H_{52}Cl_{14}$ (C_{9-31}, Cl_{2-14}) in individual samples. C_9 and up to C_{27} CPs were confirmed. Instrumental responses of the CP congener groups represented their profile of each sample.

We compared CP chain length ranges in the samples with those in a set of CP technical products (n = 65) and selected a sub-set of 16 products (Table S3) for quantification in this study, consisting of 5 SCCPs, 6 MCCPs, and 5 LCCPs. The CP congener group profile of each human milk samples was reconstructed by a deconvolution algorithm[4] from the profiles of the selected technical products. Then percentage contributions of the technical products were used to calculate instrumental response factors of SCCPs, MCCPs, and LCCPs in the sample. Here concentrations of C_9 CPs were included in those of SCCPs, as they were considered as impurities of SCCPs.[5] The performance of CP quantification was evaluated by the goodness of fit (R^2) between the reconstructed profile and the native one. $R^2 \geq 0.50$ indicates a valid quantification.[4, 6] Quantification of all samples fulfilled the criterion (Table S4).

Table S1. Theoretical number of positional isomers for polychlorinated *n*-alkanes by assuming no more than one chlorine atom bound to any carbon atom. The calculation is based on Tomy et al. (1997).[7] CP congeners are defined as individual chemicals identified by its exact structure, chirality not applied.

Chain length	$C_nH_{2n+2-m}Cl_m$, m =	Total number of positional congeners
10	1-10	527
11	1-11	1 055
12	1-12	2 079
13	1-13	4 159
14	1-14	8 255
15	1-15	16 511
16	1-16	32 895
17	1-17	65 791
18	1-18	131 327
19	1-19	262 655
20	1-20	524 799
21	1-21	1 049 599
22	1-22	2 098 175
23	1-23	4 196 351
24	1-24	8 390 655
25	1-25	16 781 311
26	1-26	33 558 527
27	1-27	67 117 055
28	1-28	134 225 919
29	1-29	268 451 839
30	1-30	536 887 295

Table S2. Detailed information and CP concentrations in human milk from Shanghai, Jiaxing, Shaoxing (China), Stockholm (Sweden), and Bodø (Norway).

Sample ID	Sampling Date (yyyy)	Age of Mother (year)	Duration [a] (year)	Occupation	Age of child [b] (day)	No. of children [c]	SCCPs (ng g^{-1} fat)	MCCPs (ng g^{-1} fat)	LCCPs (ng g^{-1} fat)
Shanghai-1	2015	30	10	Financial staff	39	1	664	1260	56.9
Shanghai-2	2015	38	8	Staff	47	2	33.4	37.9	15.0
Shanghai-3	2015	28	7	Planning designer	52	2	25.6	66.2	11.8
Shanghai-4	2015	31	9	Editor	34	2	89.2	129	16.9
Shanghai-5	2016	25	6	financial staff	43	1	19.9	50.9	20.8
Shanghai-6	2016	31	31	Architect	74	1	676	192	<LOD
Shanghai-7	2016	32	16	Teacher	54	2	36.6	76.3	1.83
Shanghai-8	2016	36	27	NA	46	1	16.8	73.9	7.44
Shanghai-9	2016	24	24	NA	NA	1	14.8	59.8	9.10
Shanghai-10	2016	27	27	NA	NA	1	38	56.4	3.49
Jiaxing-1	2015	30	10	Financial staff	59	2	16.7	32.3	12.4
Jiaxing-2	2015	28	28	Freelance	34	2	134	153	3.18
Jiaxing-3	2015	39	39	Freelance	49	2	140	70.1	18.4
Jiaxing-4	2015	35	12	Policeman	40	2	28.6	63	6.12
Jiaxing-5	2015	29	29	Bank employee	47	1	467	246	8.01
Jiaxing-6	2015	26	26	NA	40	2	21.9	51.3	8.71
Jiaxing-7	2016	40	40	Freelance	26	2	18.7	48.6	5.23
Jiaxing-8	2016	28	28	Staff	57	1	11.6	35.3	3.14
Jiaxing-9	2016	27	27	Accountant	45	1	620	263	1.53
Jiaxing-10	2016	28	28	Teacher	51	1	642	335	<LOD
Jiaxing-11	2016	30	30	Teacher	38	1	17.9	28.0	<LOD
Jiaxing-12	2016	40	21	NA	43	2	171	547	<LOD
Jiaxing-13	2016	28	28	Staff	29	2	<LOD	16.1	6.79
Shaoxing-1	2010	20-25	>10	NA	NA	1	21.2	53.1	46.9
Shaoxing-2	2010	20-25	>10	NA	NA	1	31.9	55.9	4.89
Shaoxing-3	2010	20-25	>10	NA	NA	1	28.7	89.7	15.7
Shaoxing-4	2010	20-25	>10	NA	NA	1	<LOD	37.5	9.91
Shaoxing-5	2010	20-25	>10	NA	NA	1	81.2	132	7.94
Shaoxing-6	2010	20-25	>10	NA	NA	1	66.1	121	184
Shaoxing-7	2010	20-25	>10	NA	NA	1	37.9	123	12.6
Shaoxing-8	2010	20-25	>10	NA	NA	1	64.7	121	14.6
Shaoxing-9	2010	20-25	>10	NA	NA	1	124	187	22.8
Shaoxing-10	2010	20-25	>10	NA	NA	1	43.5	86.0	120
Shaoxing-11	2010	20-25	>10	NA	NA	1	52.5	125	7.94
Shaoxing-12	2010	20-25	>10	NA	NA	1	17.3	139	8.90
Shaoxing-13	2010	20-25	>10	NA	NA	1	<LOD	81.3	15.3

Sample ID	Sampling Date (yyyy)	Age of Mother (year)	Duration [a] (year)	Occupation	Age of child [b] (day)	No. of children [c]	SCCPs (ng g^{-1} fat)	MCCPs (ng g^{-1} fat)	LCCPs (ng g^{-1} fat)
Stockholm2016-1	2016	34	>10	NA	NA	1	<LOD	21.1	7.46
Stockholm2016-2	2016	27	>10	NA	NA	1	20.0	19.2	4.53
Stockholm2016-3	2016	31	>10	NA	NA	1	<LOD	21.0	<LOD
Stockholm2016-4	2016	31	>10	NA	NA	1	13.8	<LOD	1.65
Stockholm2016-5	2016	26	>10	NA	NA	1	11.7	27.4	1.70
Stockholm2016-6	2016	26	>10	NA	NA	1	27.8	17.2	9.34
Stockholm2016-7	2016	25	>10	NA	NA	1	<LOD	60	8.32
Stockholm2016-8	2016	27	>10	NA	NA	1	<LOD	29.6	2.80
Stockholm2016-9	2016	27	>10	NA	NA	1	<LOD	19.1	<LOD
Stockholm2011-1	2011	30	>10	NA	NA	1	23.3	71	21.4
Stockholm2011-2	2011	31	>10	NA	NA	1	15.4	26.9	4.43
Stockholm2011-3	2011	31	>10	NA	NA	1	15.8	36.7	4.14
Stockholm2011-4	2011	31	>10	NA	NA	1	14.0	17.3	<LOD
Stockholm2011-5	2011	30	>10	NA	NA	1	12.9	34.7	8.92
Stockholm2011-6	2011	31	>10	NA	NA	1	24.1	77.7	6.58
Stockholm2011-7	2011	30	>10	NA	NA	1	19.0	38.0	1.84
Stockholm2011-8	2011	29	>10	NA	NA	1	18.8	19.2	1.50
Stockholm2011-9	2011	30	>10	NA	NA	1	11.8	53.7	5.26
Stockholm2011-10	2011	31	>10	NA	NA	1	14.4	42.1	3.01
Bodø-1	2014	37	>10	Cashier	77	NA	<LOD	<LOD	<LOD
Bodø-2	2014	27	>10	Store's manager	84	NA	120	88.3	29
Bodø-3	2014	40	3.5	Network Administrator	84	NA	<LOD	52.4	4.35
Bodø-4	2014	32	>10	DNB bank ASA	150	NA	<LOD	16.4	2.46
Bodø-5	2014	30	10	Barber	NA	NA	22.3	28.3	4.95
Bodø-6	2014	39	>10	Accountant	63	NA	<LOD	311	<LOD
Bodø-7	2014	36	>10	Barber	98	NA	24.3	35.5	10.8
Bodø-8	2014	41	>10	Dentist	70	NA	20.4	50.6	4.17

[a] number of years living at the current place of residence;

[b] time of sampling after parturition

[c] number of children.

NA: not available.

LODs of SCCPs, MCCPs, and LCCPs are 11.5, 15.7, and 1.09 ng g^{-1} fat in this study, respectively.

Table S3. CP technical product list involved in the present study.

Category	***Product***	***Manufacturer***	***Country***	***Ingredients****
SCCPs	SCCP 51.5 %Cl	Ehrenstorfer GmbH	Germany	C_{10-13} 51.5% Cl
	SCCP 55.5 %Cl	Ehrenstorfer GmbH	Germany	C_{10-13} 55.5% Cl
	SCCP 63.0 %Cl	Ehrenstorfer GmbH	Germany	C_{10-13} 63.0% Cl
	Witaclor 149	Dynamit Nobel AG	Germany	C_{10-13} 49% Cl
	Hüls 70C	Hüls AG	Germany	C_{10-13} 70% Cl
MCCPs	MP42	Ehrenstorfer GmbH	Germany	C_{14-17} 42.0% Cl
	MP52	Ehrenstorfer GmbH	Germany	C_{14-17} 52.0% Cl
	MP57	Ehrenstorfer GmbH	Germany	C_{14-17} 57.0% Cl
	Cloparin 49st	Caffaro	Italy	C_{14-17} 49% Cl
	Cloparin 50	Caffaro	Italy	C_{14-17} 50% Cl
	CereclorS52	INEOS Chlor Ltd.	UK	C_{14-17} 52% Cl
	YS CP52	unknown	China	C_{14} 52% Cl
LCCPs	Hüls 40N	Hüls AG	Germany	C_{18-26} 40% Cl
	Witaclor 549	Dynamit Nobel AG	Germany	C_{18-25} 49% Cl
	Unichlor 40	Neville Chemical Co.	USA	C_{22-27} 40 %Cl
	LCCP 36.0 %Cl	Ehrenstorfer GmbH	Germany	C_{18-20} 36.0% Cl
	LCCP 49.0 %Cl	Ehrenstorfer GmbH	Germany	C_{18-20} 49.0% Cl

* specifications from the manufacturer.

Table S4. Relative Contributions of CP technical products in pattern reconstruction of individual human milk samples.

Site	R^2	***SCCP 51.5%Cl***	***SCCP 55.5%Cl***	***SCCP 63.0%Cl***	***Witaclor 149***	***Hüls 70C***	***MP42***	***MP52***	***MP57***	***Cloparin 49st***	***Cloparin 50***	***Cereclor S52***	***LCCP 36%Cl***	***LCCP 49%Cl***	***Witaclor 549***	***Hüls 40N***	***Uniclor 40***
Shanghai-1	0.76	7%	9%	0%	18%		24%	16%		23%			2%	0%			
Shanghai-2	0.87		35%	7%		1%	1%	29%		2%	13%		1%	11%	3%		
Shanghai-3	0.89		29%	4%	0%	0%	11%	46%					3%	5%	1%		
Shanghai-4	0.51		30%	8%	1%		3%	43%		3%	5%			7%			
Shanghai-5	0.78	3%	21%	3%	1%	2%		45%			7%			17%			
Shanghai-6	0.57			3%	43%		14%	9%	21%		9%						0%
Shanghai-7	0.87		19%	3%	3%	0%	14%		56%	5%				1%			
Shanghai-8	0.84	22%		2%		0%	3%	60%		1%	5%			6%			
Shanghai-9	0.82	0%	12%	5%	10%	1%	25%		28%	14%			2%	2%			
Shanghai-10	0.78		11%	2%	18%	0%	21%		20%		27%			1%			
Jiaxing-1	0.77	0%	17%	5%	11%		12%	28%			11%		15%	2%			
Jiaxing-2	0.50		12%	4%	22%		41%		18%					3%			
Jiaxing-3	0.68		26%	2%	31%		4%	7%			22%			8%			
Jiaxing-4	0.88	1%	27%	0%	3%	1%	22%	27%			14%			5%			
Jiaxing-5	0.67		11%	2%	51%		13%	16%			6%			1%			
Jiaxing-6	0.89		15%	6%	7%		22%		40%	5%			1%	3%			
Jiaxing-7	0.88	0%	16%	5%	5%	2%	21%		39%	2%	6%			3%			
Jiaxing-8	0.83		18%	0%	6%	1%	7%		34%	0%	32%			2%			
Jiaxing-9	0.68			1%	55%		30%	12%	1%					0%			
Jiaxing-10	0.50		16%	5%	34%		32%		14%								
Jiaxing-11	0.74		22%	5%	14%	5%	27%	16%			10%			1%			
Jiaxing-12	0.91		15%	3%			9%		73%					0%			
Jiaxing-13	0.89	0%	17%	6%	4%	0%	4%		44%	5%	13%			6%		0%	

Site	***R^2***	***SCCP 51.5%Cl***	***SCCP 55.5%Cl***	***SCCP 63.0%Cl***	***Witaclor 149***	***Hüls 70C***	***MP42***	***MP52***	***MP57***	***Cloparin 49st***	***Cloparin 50***	***Cereclor S52***	***LCCP 36%Cl***	***LCCP 49%Cl***	***Witaclor 549***	***Hüls 40N***	***Uniclor 40***
Shaoxing-1	0.86		19%	4%	1%	2%	2%	16%			26%			28%	2%		
Shaoxing-2	0.88		29%	7%	1%			55%						9%			
Shaoxing-3	0.89	13%	5%	4%	0%	1%		62%		1%			0%	9%	4%	1%	
Shaoxing-4	0.84	4%	20%	4%	8%	2%	4%	36%		9%	2%		3%	8%			
Shaoxing-5	0.90		19%		18%	1%	1%	57%					1%	2%			
Shaoxing-6	0.72		18%	1%	1%	1%		19%			14%			43%	0%	3%	
Shaoxing-7	0.91	20%	4%	2%	0%	1%	15%	36%		1%	15%		0%	6%		0%	
Shaoxing-8	0.81	30%	3%	2%	1%	2%	9%	44%			3%			6%			
Shaoxing-9	0.82	21%	10%	1%		0%	16%	3%	32%	1%	5%	7%	0%	3%			
Shaoxing-10	0.86	6%	7%	4%	0%	0%		33%		1%	3%		0%	7%	23%	5%	8%
Shaoxing-11	0.94	14%	9%	5%	1%	1%		65%		1%			0%	3%		1%	
Shaoxing-12	0.98	3%	9%	1%	0%	0%	19%	62%					1%	3%	2%		
Shaoxing-13	0.92	3%	14%		0%	1%		49%			21%			12%			
Stockholm2016-1	0.77	1%	24%	3%	4%	2%	32%	12%		8%			2%	11%	1%		
Stockholm2016-2	0.77		6%	7%	30%	5%	14%	24%		7%	0%		5%	3%			
Stockholm2016-3	0.83	19%		8%	1%	4%	39%	18%			8%			3%			
Stockholm2016-4	0.78		11%	4%	16%	7%	19%	11%		8%	10%		1%	8%	4%		
Stockholm2016-5	0.89		13%	5%	15%	4%	24%	22%			14%			3%			
Stockholm2016-6	0.69		9%	6%	32%	5%	15%	15%		6%				12%			
Stockholm2016-7	0.75	13%	5%	4%	0%	1%	51%	12%		1%	6%			8%		0%	
Stockholm2016-8	0.85		19%	1%	1%	3%	16%	47%		1%	8%		3%	2%			
Stockholm2016-9	0.65	20%	12%	7%	1%	2%	34%	9%		1%	13%		0%	0%			
Stockholm2011-1	0.76	0%	12%	1%	4%	5%	48%	8%			7%			3%	12%	0%	
Stockholm2011-2	0.86		9%	4%	13%	6%	13%	39%			8%		1%	5%			

Site	***R^2***	***SCCP 51.5%Cl***	***SCCP 55.5%Cl***	***SCCP 63.0%Cl***	***Witaclor 149***	***Hüls 70C***	***MP42***	***MP52***	***MP57***	***Cloparin 49st***	***Cloparin 50***	***Cereclor S52***	***LCCP 36%Cl***	***LCCP 49%Cl***	***Witaclor 549***	***Hüls 40N***	***Uniclor 40***
Stockholm2011-3	0.67		19%		8%	2%	46%	20%						5%			
Stockholm2011-4	0.78		16%	2%	13%	5%	52%	8%						3%			
Stockholm2011-5	0.85	10%	4%	5%	1%	5%	58%			0%	5%		7%	5%			
Stockholm2011-6	0.80	0%	22%	1%	2%	2%	60%	5%		1%	2%		3%	3%			
Stockholm2011-7	0.72		17%	3%	16%	6%	47%				9%			3%			
Stockholm2011-8	0.76		18%	4%	21%	7%	30%	7%			9%			4%			
Stockholm2011-9	0.83		17%	2%	1%	2%	51%	13%		1%	6%		2%	5%			
Stockholm2011-10	0.81	15%	7%	6%	0%	1%	58%	7%		1%				0%	4%	1%	
Bodø-1	0.69	0%	21%	3%	12%	12%	25%	0%		5%	20%		1%	1%			
Bodø-2	0.69	43%	6%		1%	1%	27%	9%		2%					7%	2%	
Bodø-3	0.91	10%	6%	3%	0%	1%	69%	5%		0%			0%	5%			
Bodø-4	0.71		21%	3%	12%	5%	33%	11%			10%		2%	4%			
Bodø-5	0.79	0%	18%	5%	7%	8%	24%	16%			15%		5%	2%			
Bodø-6	0.94		0%	0%	1%		0%		98%	0%							
Bodø-7	0.74		8%	6%	20%	3%	31%	12%		6%			4%	5%	4%		
Bodø-8	0.89	0%	8%	3%	16%	4%	20%	16%		7%	21%		2%	4%			

Table S5. Test statistics of Kruskal Wallis test grouped by sampling sites

Variable	n	Group	Chi-Square	Asymp. Sig.	Variable	n	Group	Chi-Square	Asymp. Sig.
C9H18Cl2	63	6	0.000	1.000	C13H20Cl8	63	6	20.954	0.001
C9H17Cl3	63	6	5.713	0.335	C13H19Cl9	63	6	1.960	0.855
C9H16Cl4	63	6	29.358	0.000	C13H18Cl10	63	6	0.926	0.968
C9H15Cl5	63	6	37.739	0.000	C13H17Cl11	63	6	11.876	0.037
C9H14Cl6	63	6	19.131	0.002	C13H16Cl12	63	6	4.098	0.535
C9H13Cl7	63	6	9.516	0.090	C13H15Cl13	63	6	0.000	1.000
C9H12Cl8	63	6	15.326	0.009	C13H14Cl14	63	6	0.000	1.000
C9H11Cl9	63	6	5.581	0.349	C14H28Cl2	63	6	5.395	0.370
C10H19Cl3	63	6	5.867	0.319	C14H27Cl3	63	6	32.074	0.000
C10H18Cl4	63	6	20.874	0.001	C14H26Cl4	63	6	35.749	0.000
C10H17Cl5	63	6	29.952	0.000	C14H25Cl5	63	6	17.953	0.003
C10H16Cl6	63	6	29.665	0.000	C14H24Cl6	63	6	11.656	0.040
C10H15Cl7	63	6	20.173	0.001	C14H23Cl7	63	6	20.008	0.001
C10H14Cl8	63	6	7.191	0.207	C14H22Cl8	63	6	18.250	0.003
C10H13Cl9	63	6	12.523	0.028	C14H21Cl9	63	6	8.412	0.135
C10H12Cl10	63	6	8.689	0.122	C14H20Cl10	63	6	0.771	0.979
C10H11Cl11	63	6	14.639	0.012	C14H19Cl11	63	6	9.475	0.092
C10H10Cl12	63	6	5.874	0.319	C14H18Cl12	63	6	5.146	0.398
C11H21Cl3	63	6	6.433	0.266	C14H17Cl13	63	6	0.000	1.000
C11H20Cl4	63	6	15.205	0.010	C14H16Cl14	63	6	0.000	1.000
C11H19Cl5	63	6	16.935	0.005	C15H30Cl2	63	6	5.022	0.413
C11H18Cl6	63	6	9.015	0.108	C15H29Cl3	63	6	31.925	0.000
C11H17Cl7	63	6	9.380	0.095	C15H28Cl4	63	6	35.439	0.000
C11H16Cl8	63	6	27.270	0.000	C15H27Cl5	63	6	30.044	0.000
C11H15Cl9	63	6	16.512	0.006	C15H26Cl6	63	6	5.784	0.328
C11H14Cl10	63	6	28.552	0.000	C15H25Cl7	63	6	7.797	0.168
C11H13Cl11	63	6	6.655	0.248	C15H24Cl8	63	6	7.308	0.199
C11H12Cl12	63	6	1.341	0.931	C15H23Cl9	63	6	1.312	0.934
C12H24Cl2	63	6	10.936	0.053	C15H22Cl10	63	6	1.693	0.890
C12H23Cl3	63	6	2.176	0.824	C15H21Cl11	63	6	5.566	0.351
C12H22Cl4	63	6	6.856	0.232	C15H20Cl12	63	6	5.540	0.354
C12H21Cl5	63	6	6.190	0.288	C15H19Cl13	63	6	0.000	1.000
C12H20Cl6	63	6	2.448	0.784	C15H18Cl14	63	6	0.000	1.000
C12H19Cl7	63	6	3.448	0.631	C16H32Cl2	63	6	0.000	1.000
C12H18Cl8	63	6	2.449	0.784	C16H31Cl3	63	6	16.401	0.006
C12H17Cl9	63	6	3.632	0.604	C16H30Cl4	63	6	31.864	0.000
C12H16Cl10	63	6	10.064	0.073	C16H29Cl5	63	6	25.621	0.000
C12H15Cl11	63	6	3.936	0.559	C16H28Cl6	63	6	5.787	0.327
C12H14Cl12	63	6	6.970	0.223	C16H27Cl7	63	6	6.023	0.304
C12H13Cl13	63	6	0.000	1.000	C16H26Cl8	63	6	4.859	0.433
C12H12Cl14	63	6	0.000	1.000	C16H25Cl9	63	6	10.717	0.057
C13H26Cl2	63	6	8.453	0.133	C16H24Cl10	63	6	0.755	0.980
C13H25Cl3	63	6	5.843	0.322	C16H23Cl11	63	6	7.377	0.194
C13H24Cl4	63	6	9.835	0.080	C16H22Cl12	63	6	0.889	0.971
C13H23Cl5	63	6	5.896	0.317	C16H21Cl13	63	6	0.000	1.000
C13H22Cl6	63	6	11.402	0.044	C16H20Cl14	63	6	0.000	1.000
C13H21Cl7	63	6	20.075	0.001	C17H34Cl2	63	6	0.000	1.000

Table S5. (Cont.) Test statistics of Kruskal Wallis Test grouped by sampling sites

Variable	n	Group	Chi-Square	Asymp. Sig.	Variable	n	Group	Chi-Square	Asymp. Sig.
C17H33Cl3	63	6	6.698	0.244	C20H31Cl11	63	6	6.267	0.281
C17H32Cl4	63	6	17.194	0.004	C20H30Cl12	63	6	4.595	0.467
C17H31Cl5	63	6	16.034	0.007	C20H29Cl13	63	6	0.000	1.000
C17H30Cl6	63	6	7.506	0.186	C20H28Cl14	63	6	0.000	1.000
C17H29Cl7	63	6	2.656	0.753	C21H42Cl2	63	6	0.000	1.000
C17H28Cl8	63	6	3.752	0.586	C21H41Cl3	63	6	9.574	0.088
C17H27Cl9	63	6	8.487	0.131	C21H40Cl4	63	6	7.555	0.183
C17H26Cl10	63	6	2.888	0.717	C21H39Cl5	63	6	4.445	0.487
C17H25Cl11	63	6	1.515	0.911	C21H38Cl6	63	6	7.976	0.158
C17H24Cl12	63	6	5.935	0.313	C21H37Cl7	63	6	2.672	0.750
C17H23Cl13	63	6	0.000	1.000	C21H36Cl8	63	6	2.558	0.768
C17H22Cl14	63	6	0.000	1.000	C21H35Cl9	63	6	6.550	0.256
C18H36Cl2	63	6	0.000	1.000	C21H34Cl10	63	6	2.879	0.719
C18H35Cl3	63	6	2.708	0.745	C21H33Cl11	63	6	12.207	0.032
C18H34Cl4	63	6	3.314	0.652	C21H32Cl12	63	6	5.743	0.332
C18H33Cl5	63	6	8.847	0.115	C21H31Cl13	63	6	0.000	1.000
C18H32Cl6	63	6	12.606	0.027	C21H30Cl14	63	6	0.000	1.000
C18H31Cl7	63	6	9.694	0.084	C22H44Cl2	63	6	3.846	0.572
C18H30Cl8	63	6	6.147	0.292	C22H43Cl3	63	6	4.143	0.529
C18H29Cl9	63	6	3.138	0.679	C22H42Cl4	63	6	4.516	0.478
C18H28Cl10	63	6	5.837	0.322	C22H41Cl5	63	6	5.743	0.332
C18H27Cl11	63	6	2.992	0.701	C22H40Cl6	63	6	5.673	0.339
C18H26Cl12	63	6	6.061	0.300	C22H39Cl7	63	6	5.743	0.332
C18H25Cl13	63	6	0.000	1.000	C22H38Cl8	63	6	5.673	0.339
C18H24Cl14	63	6	0.000	1.000	C22H37Cl9	63	6	5.673	0.339
C19H38Cl2	63	6	0.000	1.000	C22H36Cl10	63	6	3.656	0.600
C19H37Cl3	63	6	2.594	0.762	C22H35Cl11	63	6	3.609	0.607
C19H36Cl4	63	6	7.863	0.164	C22H34Cl12	63	6	3.609	0.607
C19H35Cl5	63	6	6.478	0.262	C22H33Cl13	63	6	0.000	1.000
C19H34Cl6	63	6	8.537	0.129	C22H32Cl14	63	6	0.000	1.000
C19H33Cl7	63	6	12.985	0.024	C23H46Cl2	63	6	3.846	0.572
C19H32Cl8	63	6	7.590	0.180	C23H45Cl3	63	6	0.000	1.000
C19H31Cl9	63	6	3.403	0.638	C23H44Cl4	63	6	3.846	0.572
C19H30Cl10	63	6	3.287	0.656	C23H43Cl5	63	6	3.609	0.607
C19H29Cl11	63	6	5.069	0.407	C23H42Cl6	63	6	7.816	0.167
C19H28Cl12	63	6	4.624	0.463	C23H41Cl7	63	6	5.821	0.324
C19H27Cl13	63	6	0.000	1.000	C23H40Cl8	63	6	7.816	0.167
C19H26Cl14	63	6	0.000	1.000	C23H39Cl9	63	6	5.743	0.332
C20H40Cl2	63	6	0.000	1.000	C23H38Cl10	63	6	3.656	0.600
C20H39Cl3	63	6	1.651	0.895	C23H37Cl11	63	6	3.846	0.572
C20H38Cl4	63	6	6.531	0.258	C23H36Cl12	63	6	3.846	0.572
C20H37Cl5	63	6	7.529	0.184	C23H35Cl13	63	6	0.000	1.000
C20H36Cl6	63	6	6.710	0.243	C23H34Cl14	63	6	0.000	1.000
C20H35Cl7	63	6	11.314	0.045	C24H48Cl2	63	6	5.300	0.380
C20H34Cl8	63	6	5.255	0.386	C24H47Cl3	63	6	3.656	0.600
C20H33Cl9	63	6	3.644	0.602	C24H46Cl4	63	6	3.609	0.607
C20H32Cl10	63	6	4.896	0.429	C24H45Cl5	63	6	7.816	0.167

Table S5. (Cont.) Test statistics of Kruskal Wallis Test grouped by sampling sites

Variable	n	Group	Chi-Square	Asymp. Sig.	Variable	n	Group	Chi-Square	Asymp. Sig.
C24H44Cl6	63	6	3.846	0.572	C26H43Cl11	63	6	0.000	1.000
C24H43Cl7	63	6	5.743	0.332	C26H42Cl12	63	6	3.846	0.572
C24H42Cl8	63	6	7.816	0.167	C26H41Cl13	63	6	0.000	1.000
C24H41Cl9	63	6	7.816	0.167	C26H40Cl14	63	6	0.000	1.000
C24H40Cl10	63	6	7.816	0.167	C26H39Cl15	63	6	0.000	1.000
C24H39Cl11	63	6	3.846	0.572	C27H54Cl2	63	6	5.146	0.398
C24H38Cl12	63	6	7.816	0.167	C27H53Cl3	63	6	2.952	0.707
C24H37Cl13	63	6	0.000	1.000	C27H52Cl4	63	6	3.846	0.572
C24H36Cl14	63	6	0.000	1.000	C27H51Cl5	63	6	3.846	0.572
C25H50Cl2	63	6	0.000	1.000	C27H50Cl6	63	6	3.846	0.572
C25H49Cl3	63	6	0.000	1.000	C27H49Cl7	63	6	3.846	0.572
C25H48Cl4	63	6	3.846	0.572	C27H48Cl8	63	6	3.846	0.572
C25H47Cl5	63	6	0.000	1.000	C27H47Cl9	63	6	3.846	0.572
C25H46Cl6	63	6	7.816	0.167	C27H46Cl10	63	6	3.846	0.572
C25H45Cl7	63	6	2.442	0.785	C27H45Cl11	63	6	3.846	0.572
C25H44Cl8	63	6	7.816	0.167	C27H44Cl12	63	6	3.846	0.572
C25H43Cl9	63	6	7.816	0.167	C27H43Cl13	63	6	0.000	1.000
C25H42Cl10	63	6	7.816	0.167	C27H52Cl4	63	6	3.846	0.572
C25H41Cl11	63	6	3.846	0.572	C27H51Cl5	63	6	3.846	0.572
C25H40Cl12	63	6	7.816	0.167	C27H50Cl6	63	6	3.846	0.572
C25H39Cl13	63	6	0.000	1.000	C27H49Cl7	63	6	3.846	0.572
C25H38Cl14	63	6	0.000	1.000	C27H48Cl8	63	6	3.846	0.572
C26H52Cl2	63	6	3.846	0.572	C27H47Cl9	63	6	3.846	0.572
C26H51Cl3	63	6	0.000	1.000	C27H46Cl10	63	6	3.846	0.572
C26H50Cl4	63	6	3.846	0.572	C27H45Cl11	63	6	3.846	0.572
C26H49Cl5	63	6	3.846	0.572	C27H44Cl12	63	6	3.846	0.572
C26H48Cl6	63	6	0.000	1.000	C27H43Cl13	63	6	0.000	1.000
C26H47Cl7	63	6	3.846	0.572	C27H42Cl14	63	6	0.000	1.000
C26H46Cl8	63	6	0.000	1.000	C27H41Cl15	63	6	0.000	1.000
C26H45Cl9	63	6	3.846	0.572	C27H40Cl16	63	6	0.000	1.000
C26H44Cl10	63	6	0.000	1.000					

Table S6. Estimated daily intake [EDI, ng (kg BW)$^{-1}$ day^{-1}] of CPs via breast-feeding of infants, and margin of exposure (MOE, dimensionless).[7]

		China n=36	Sweden n=20	Norway n=8
Mean concentration (ng g fat^{-1})				
SCCPs		124	12.8	23.4
MCCPs		146	33.3	72.8
LCCPs		19.1	4.89	6.97
EDI (ng (kg BW)$^{-1}$ day^{-1})				
SCCPs	average consumption	570	58.7	107
	high consumption	856	88.0	161
MCCPs	average consumption	669	153	334
	high consumption	1000	229	501
LCCPs	average consumption	87.7	22.4	32.0
	high consumption	132	33.6	48.0
MOE (EU)				
SCCPs	average consumption	4030	39200	21400
	high consumption	2690	26100	14300
MCCPs	average consumption	53800	236000	108000
	high consumption	35900	157000	71800
MOE (ECCC)				
MCCPs	average consumption	8970	39300	18000
	high consumption	5980	26200	12000
LCCPs	average consumption	810000	3170000	2220000
	high consumption	540000	2110000	1480000

The EDI was estimated based on the calculation method given in EFSA (2019), and the $BMDL_{10}$ value for SCCPs and MCCPs proposed by EFSA is 2.3 and 36 mg (kg BW)$^{-1}$ day^{-1}, respectively.[7] The LOEL value for MCCPs and LCCPs, respectively, proposed by Environment and Climate Change Canada, is 6 and 71 mg (kg BW)$^{-1}$ day^{-1}.[8]

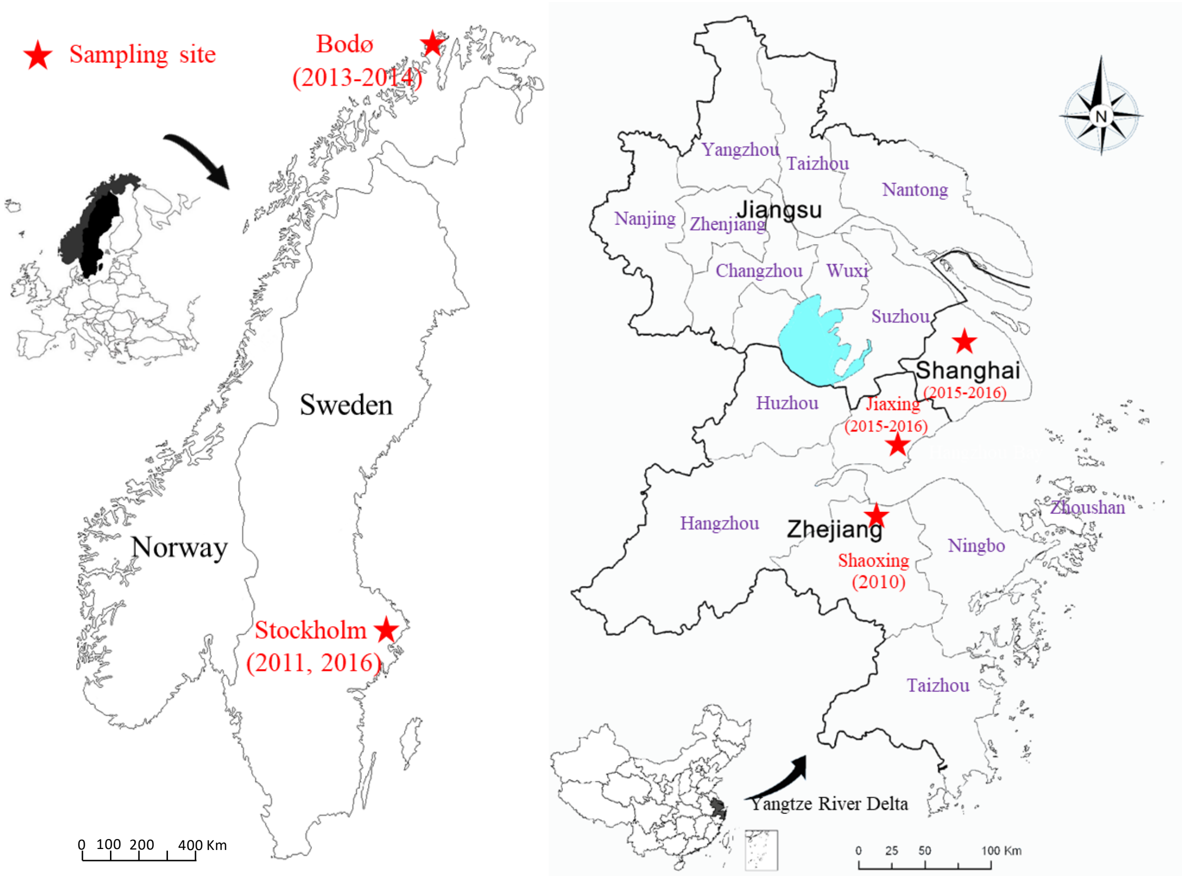

Figure S1. Map of sampling sites and sampling years. Left) map of Norway and Sweden, samples were collected in Bodø in 2014 and in Stockholm in 2011 and 2016. Right) map of the Yangtze River Delta, China, samples were collected in Shanghai in 2015-2016, Jiaxing in 2015-2016, and Shaoxing in 2010.

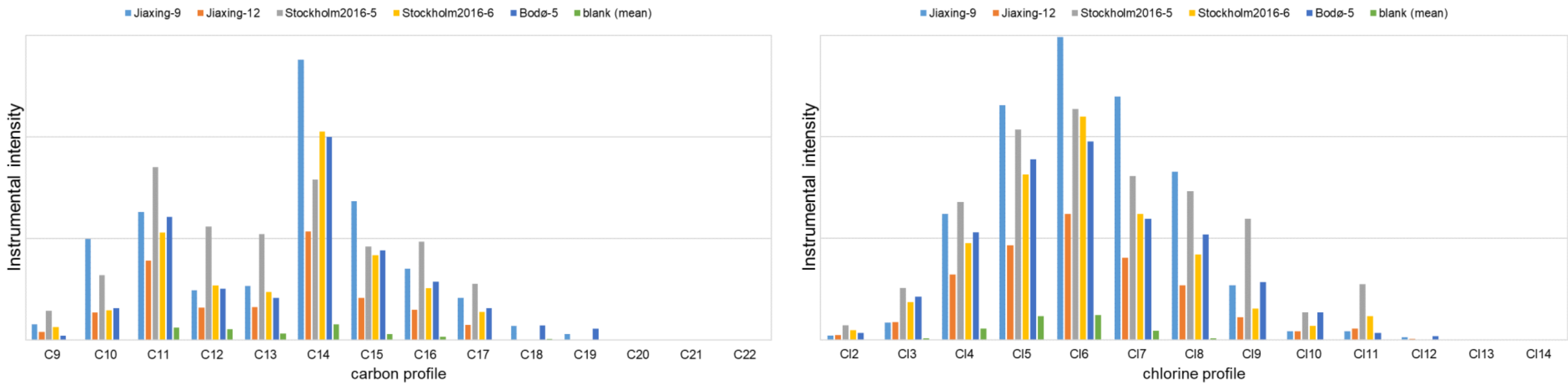

Figure S2. Comparisons of carbon and chlorine profiles between blank (mean) and samples with CP concentrations between LOD and LOQ. There are five samples, consisting of two Chinese, two Swedish, and one Norwegian samples. The instrumental intensities of C- and Cl-homologues in the five samples are higher than in the mean blank by an average factor of 10 for both C_{11-14} and Cl_{4-7} homologues.

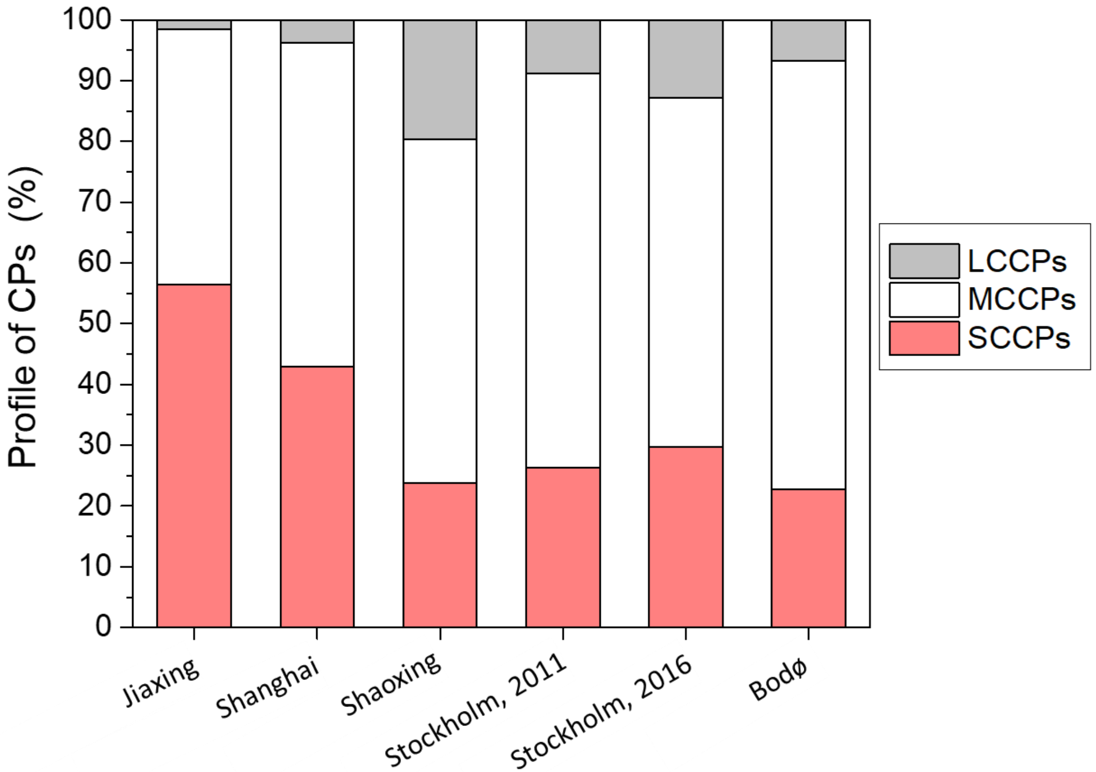

Figure S3. Composition profiles of SCCPs, MCCPs, and LCCPs in human milk from Jiaxing, Shanghai, and Shaoxing in China, Stockholm in Sweden, and Bodø in Norway.

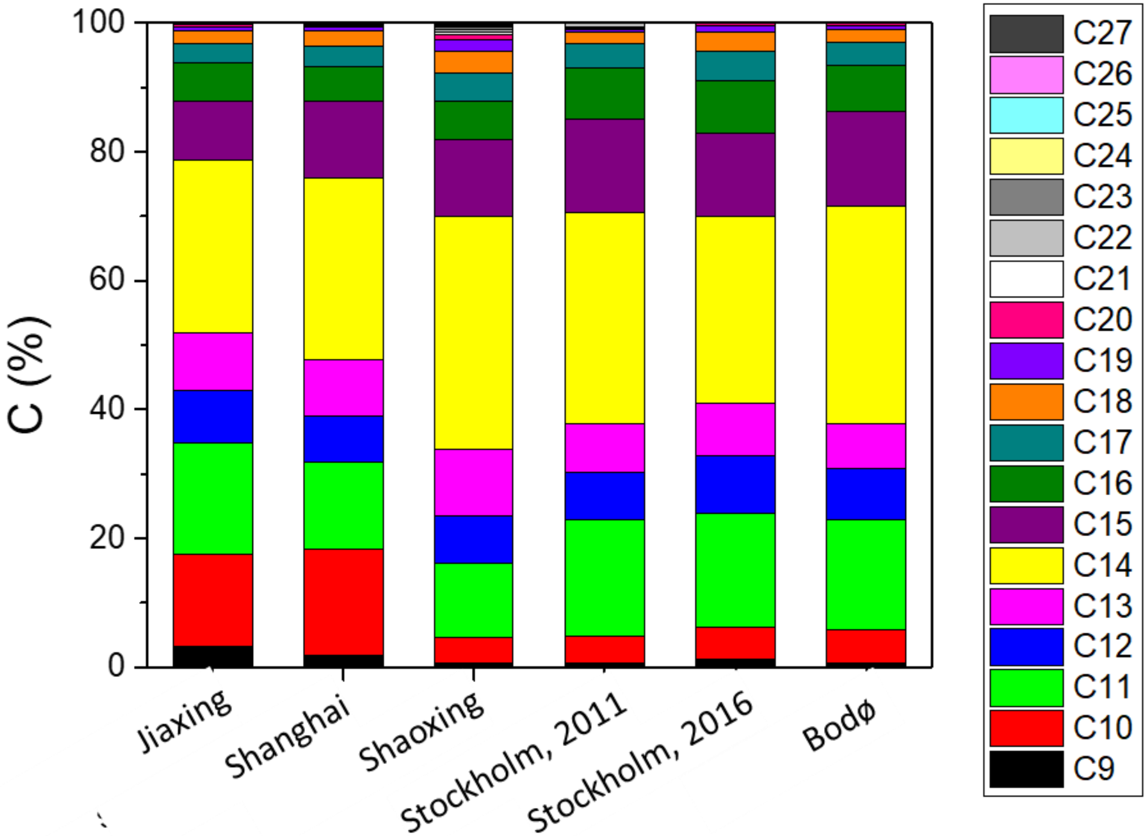

Figure S4. Compositional profiles of carbon chain homologs among the CPs assessed showing the occurrence of $C_{9\text{-}27}$.

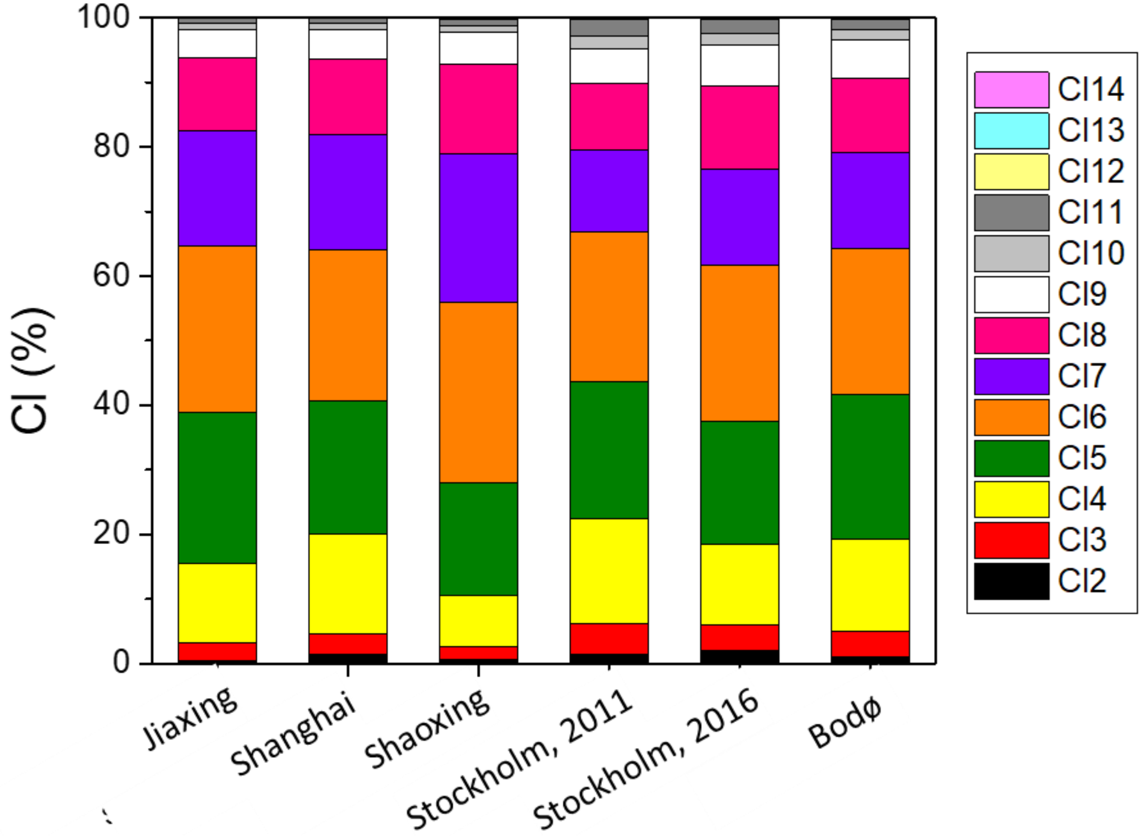

Figure S5 Compositional profiles of the chlorine homolog series of CPs assessed showing Cl_{2-14} in mothers' milk from Jiaxing, Shanghai, Shaoxing, Stockholm, and Bodø.

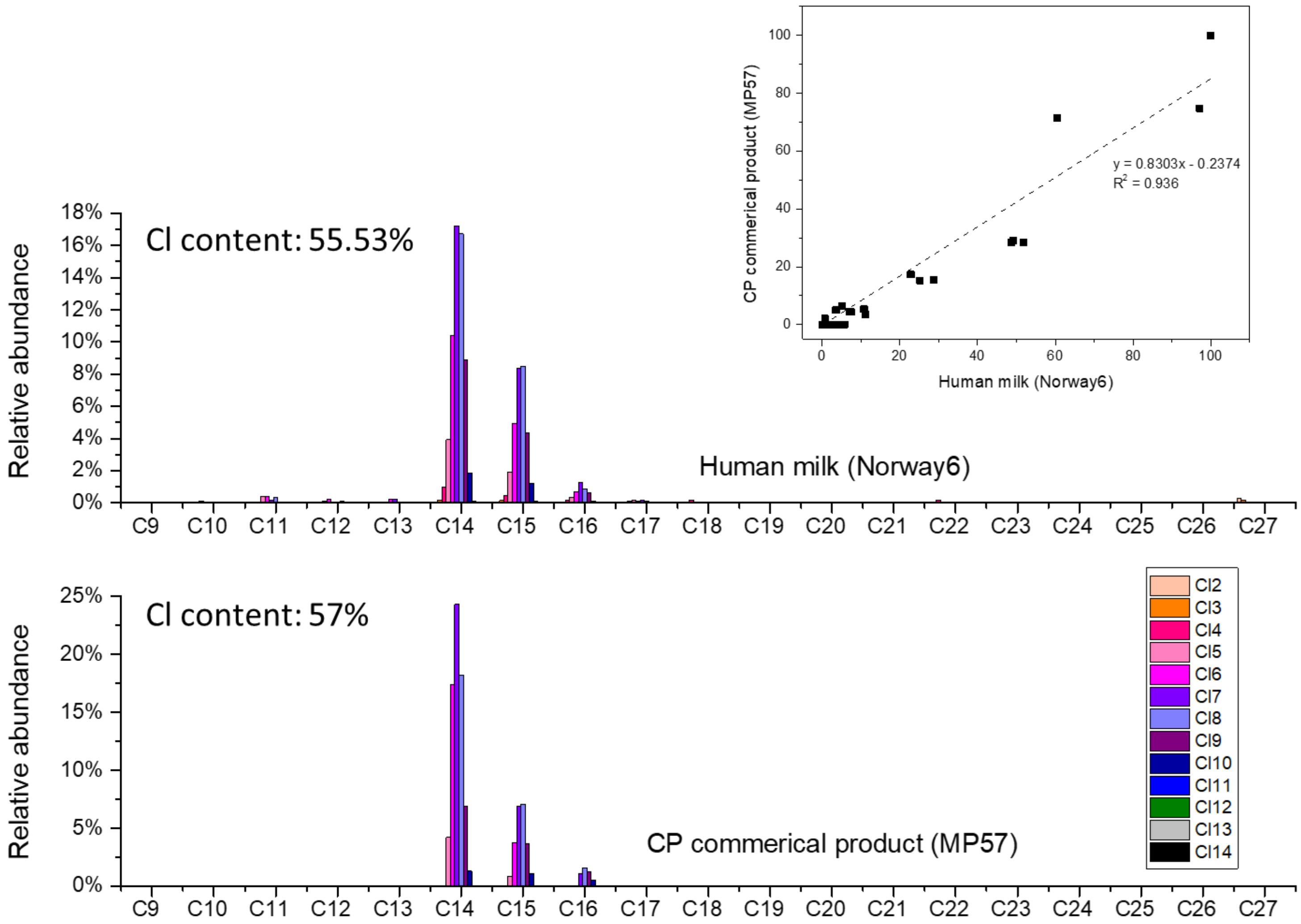

Figure S6. Congener group patterns of chlorinated paraffins (CPs) in one human milk sample (Bodø-6) from Norway and one CP commercial product (MP57) from the European market. The diagram in the upper right corner showed a significant positive correlation (R^2=0.94, p<0.01) between Bodø-6 and MP57.

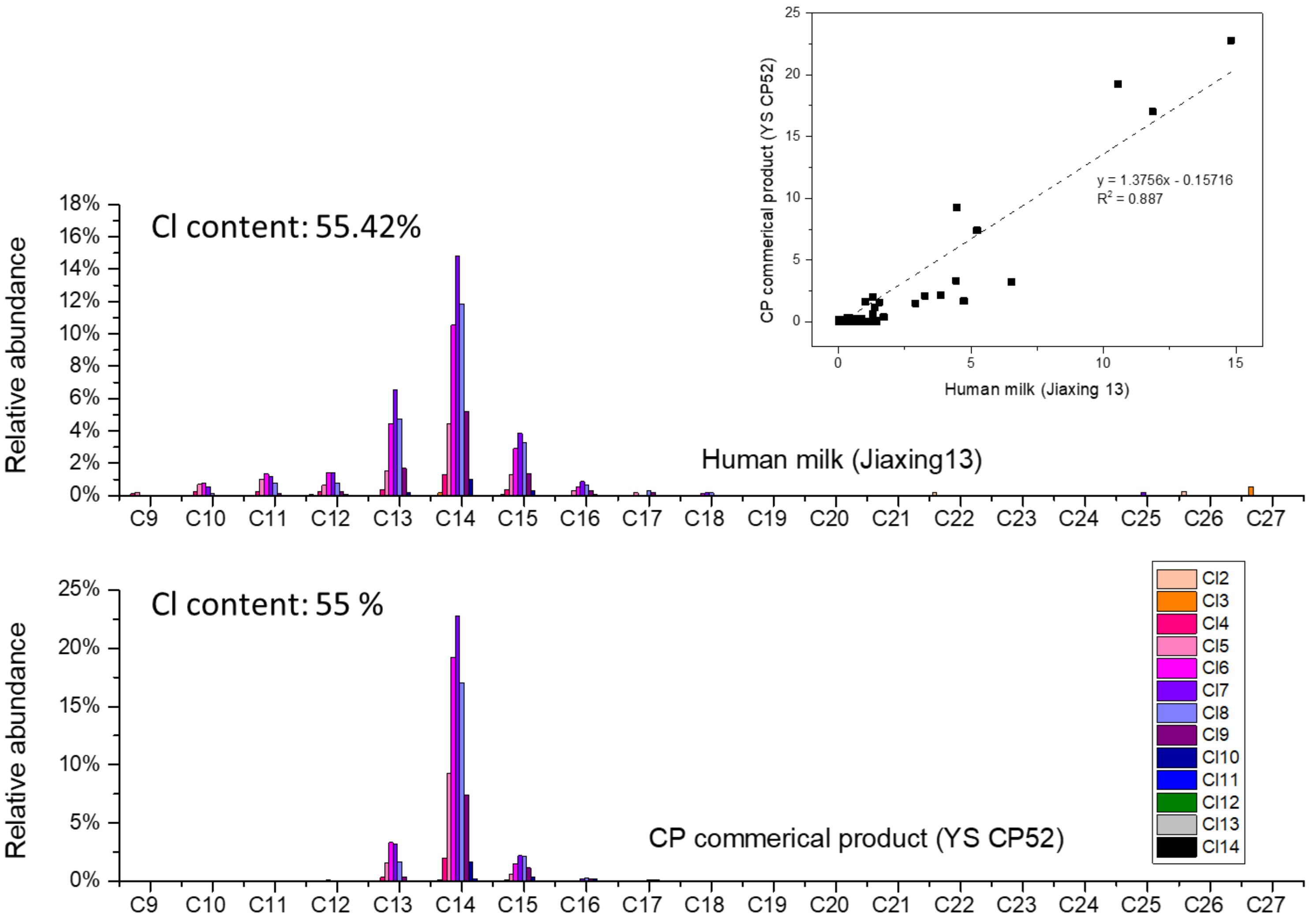

Figure S7. Congener group patterns of chlorinated paraffins (CPs) in one human milk sample (Jiaxing-12) from Jiaxing, China and one CP commercial product (YS CP52) from the Chinese market. The diagram in upper right corner showed a significant positive correlation (R^2=0.89, p<0.01) between Jiaxing-12 and YS CP52).

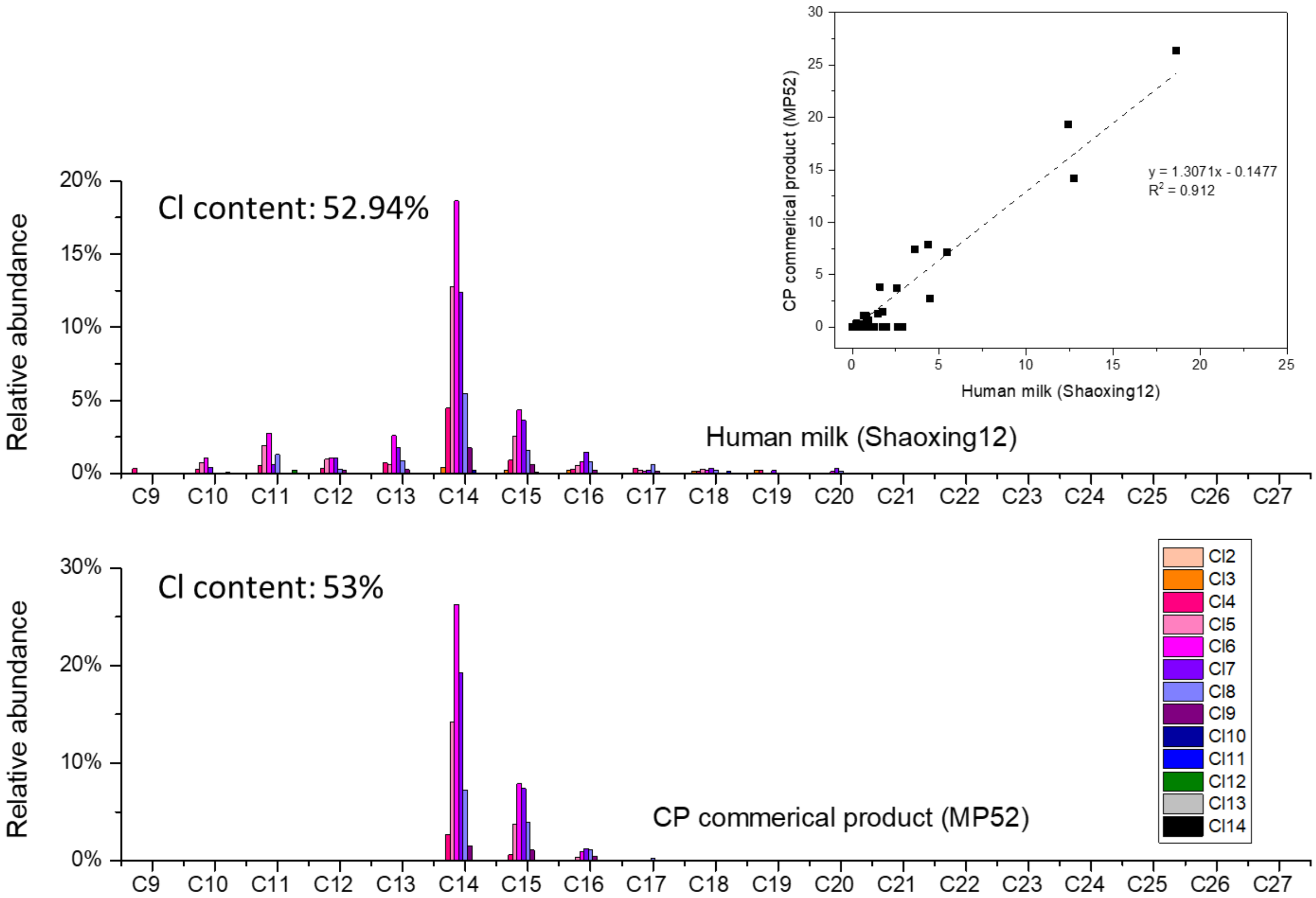

Figure S8. Congener group patterns of chlorinated paraffins (CPs) in one human milk sample (Shaoxing-12) from Shaoxing, China and one CP commercial product (MP52) from the European market. The diagram in the upper right corner showed a significant positive correlation (R^2=0.91, p<0.01) Shaoxing-12 and MP52.

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