Supporting Information for:

Temporal shifts in poly- and perfluoroalkyl substances (PFASs) in North Atlantic pilot whales indicate large contribution of atmospheric precursors

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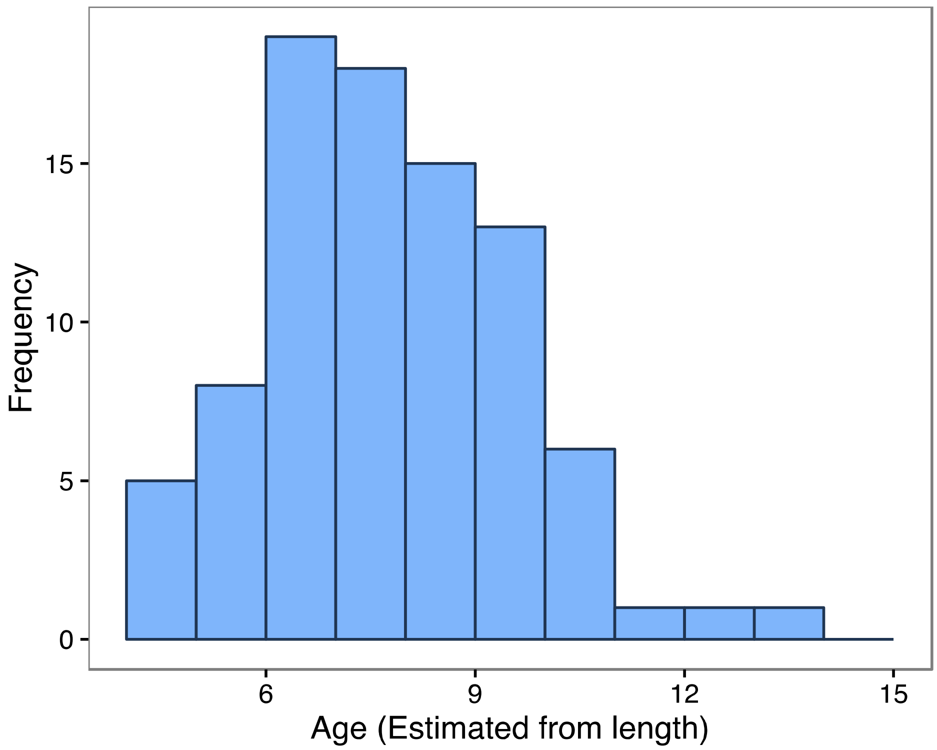

Figure S1. Age distribution of juvenile male pilot whales collected as part of this study and from the Faroese Environment Agency[1] between 1987 and 2013.

Table S1a. Ancillary data on pilot whales harvested in the Faroe Islands with tissues archived by the Faroese Natural History Museum and analyzed for PFASs in this study.

Sample ID	Sampling Date	Gender	Length (cm)	Age Category[a]
19940630-01	6/30/94	M	445	Juvenile
19940630-02	6/30/94	M	435	Juvenile
19960628-03	6/28/96	M	385	Juvenile
19960628-04	6/28/96	M	407	Juvenile
19960628-05	6/28/96	M	380	Juvenile
19981125-06	11/25/98	M	395	Juvenile
19981125-07	11/25/98	M	370	Juvenile
19990314-08	3/14/99	M	400	Juvenile
19990314-09	3/14/99	M	400	Juvenile
19990908-010	9/8/99	M	430	Juvenile
20000831-011	8/31/00	M	390	Juvenile
20000831-012	8/31/00	M	435	Juvenile
20000909-013	9/9/00	M	415	Juvenile
20060101-014	1/1/06	M	470	Juvenile
20060101-015	1/1/06	M	430	Juvenile
20060101-016	1/1/06	M	366	Juvenile
20060101-017	1/1/06	M	450	Juvenile
20060101-018	1/1/06	M	455	Juvenile
20060101-019	1/1/06	M	458	Juvenile
20060101-020	1/1/06	M	411	Juvenile
20110209-021	2/9/11	M	369	Juvenile
20110209-022	2/9/11	M	437	Juvenile
20110528-023	5/28/11	M	423	Juvenile
20110528-024	5/28/11	M	459	Juvenile
20110902-025	9/2/11	M	414	Juvenile
20110902-026	9/2/11	M	450	Juvenile
20111113-027	11/13/11	M	433	Juvenile
20111113-028	11/13/11	M	343	Juvenile
20111113-029	11/13/11	M	436	Juvenile
20111113-030	11/13/11	M	453	Juvenile
20111113-031	11/13/11	M	338	Juvenile
20111118-032	11/18/11	M	415	Juvenile
20111122-033	11/22/11	M	446	Juvenile
20111122-034	11/22/11	M	508	Adult
20130721-035	7/21/13	M	400	Juvenile
20130721-036	7/21/13	M	578	Adult
20130721-037	7/21/13	M	515	Adult
20130721-038	7/21/13	F	424	Adult

Table S1b. Ancillary data on pilot whales harvested in the Faroe Islands with tissues archived by the Faroese Natural History Museum and analyzed for PFASs in this study.

Sample ID	Sampling Date	Gender	Length (cm)	Age Category[a]
20130721-039	7/21/13	M	406	Juvenile
20130721-040	7/21/13	F	442	Adult
20130721-041	7/21/13	F	320	Juvenile
20130721-042	7/21/13	F	324	Juvenile
20130721-043	7/21/13	M	376	Juvenile
20130721-044	7/21/13	F	424	Adult
20130721-045	7/21/13	M	526	Adult
20130721-046	7/21/13	F	454	Adult
20130721-047	7/21/13	F	431	Adult
20130721-048	7/21/13	F	443	Adult
20130721-049	7/21/13	M	459	Juvenile
20130721-050	7/21/13	F	339	Juvenile
20130721-051	7/21/13	M	431	Juvenile
20130730-052	7/30/13	F	429	Adult
20130730-053	7/30/13	F	431	Adult
20130808-054	8/8/13	M	532	Adult
20130808-055	8/8/13	F	452	Adult
20130808-056	8/8/13	F	408	Adult
20130808-057	8/8/13	M	421	Juvenile
20130808-058	8/8/13	M	425	Juvenile
20130808-059	8/8/13	F	324	Juvenile
20130808-060	8/8/13	M	545	Adult
20130808-061	8/8/13	F	425	Adult
20130811-062	8/11/13	M	440	Juvenile
20130811-063	8/11/13	F	338	Juvenile
20130811-064	8/11/13	F	309	Juvenile
20130811-065	8/11/13	F	438	Adult
20130811-066	8/11/13	M	482	Juvenile
20130811-067	8/11/13	F	402	Adult
20130811-068	8/11/13	F	430	Adult
20130811-069	8/11/13	M	564	Adult
20130811-070	8/11/13	M	415	Juvenile
20130811-071	8/11/13	M	340	Juvenile
20130811-072	8/11/13	M	414	Juvenile
20130811-073	8/11/13	F	346	Juvenile
20130811-074	8/11/13	M	542	Adult
20130814-075	8/14/13	F	431	Adult
20130814-076	8/14/13	F	298	Juvenile
20130814-077	8/14/13	F	443	Adult

Table S1c. Ancillary data on pilot whales harvested in the Faroe Islands with tissues archived by the Faroese Natural History Museum and analyzed for PFASs in this study.

Sample ID	Sampling Date	Gender	Length (cm)	Age Category[a]
20130814-078	8/14/13	F	426	Adult
20130814-079	8/14/13	F	434	Adult
20130814-080	8/14/13	M	502	Adult
20130814-081	8/14/13	F	427	Adult
20130814-082	8/14/13	M	430	Juvenile
20130827-083	8/27/13	F	454	Adult
20130827-084	8/27/13	M	493	Juvenile
20130827-085	8/27/13	M	426	Juvenile
20130827-086	8/27/13	M	394	Juvenile
20130827-087	8/27/13	F	303	Juvenile

[a]Age was determined by size following Bloch et al.[4] with adults defined by lengths >500 cm for males and >378 cm for females.

Table S2. Supporting data on pilot whale muscle tissue collected and analyzed by the Faroese Environment Agency.[1]

Sample ID	Sampling Date	Gender	Length (cm)	Age Category[a]
19860712-088	7/12/86	M	385	Juvenile
19860712-089	7/12/86	M	422	Juvenile
19861026-090	10/26/86	M	452	Juvenile
19861026-091	10/26/86	M	360	Juvenile
19861101-092	11/1/86	M	391	Juvenile
19861101-093	11/1/86	M	400	Juvenile
19870722-094	7/22/87	M	380	Juvenile
19870802-095	8/2/87	M	398	Juvenile
19870802-096	8/2/87	M	408	Juvenile
19870819-097	8/19/87	M	410	Juvenile
19870819-098	8/19/87	M	380	Juvenile
19880610-099	6/10/88	M	379	Juvenile
19880610-0100	6/10/88	M	415	Juvenile
19970826-0101	8/26/97	M	390	Juvenile
19970826-0102	8/26/97	M	420	Juvenile
19970924-0103	9/24/97	M	360	Juvenile
19970924-0104	9/24/97	M	430	Juvenile
19971202-0105	12/2/97	M	381	Juvenile
19971202-0106	12/2/97	M	419	Juvenile
20010627-0107	6/27/01	M	440	Juvenile
2001076-0108	7/6/01	M	450	Juvenile
2001076-0109	7/6/01	M	354	Juvenile
2001076-0110	7/6/01	M	418	Juvenile
20020903-0111	9/3/02	M	350	Juvenile
20020903-0112	9/3/02	M	390	Juvenile
20020903-0113	9/3/02	M	440	Juvenile
20060828-0114	8/28/06	M	385	Juvenile
20060828-0115	8/28/06	M	390	Juvenile
20060828-0116	8/28/06	M	430	Juvenile
20070703-0117	7/3/07	M	405	Juvenile
20070703-0118	7/3/07	M	380	Juvenile
20070713-0119	7/13/07	M	370	Juvenile
20090105-0120	1/5/09	M	380	Juvenile
20090105-0121	1/5/09	M	405	Juvenile
20100624-0122	6/24/10	M	370	Juvenile
20100624-0123	6/24/10	M	416	Juvenile
20100702-0124	7/2/10	M	387	Juvenile
20100702-0125	7/2/10	M	440	Juvenile

Table S3. Analytical parameters for target analytes and isotopically labeled compounds.

Analytes	Precursor ion (*m/z*)	Product ions (*m/z*)	Collision energy (V)
PFBS	299	80	-56
		99	-38
PFHxS	399	99	-50
		80	-80
$^{13}C_4$-PFHxS	403	103	-80
PFHxA	313	269	-12
		119	-30
$^{13}C_4$-PFHxA	315	270	-12
PFHpA	363	169	-26
		319	-16
PFOS	499	80	-104
		99	-100
$^{13}C_4$-PFOS	503	99	-100
PFOSA	498	169	-50
		78	-62
$^{13}C_8$-PFOSA	506	78	-62
PFNA	463	419	-22
$^{13}C_5$-PFNA	468	423	-22
PFOA	413	369	-20
		169	-22
$^{13}C_4$-PFOA	417	372	-20
PFHpS	449	80	-92
		99	-84
PFDS	599	99	-104
		80	-104
PFDA	513	219	-30
		469	-22
$^{13}C_2$-PFDA	515	470	-20
PFUnA	563	519	-22
		169	-44
$^{13}C_2$-PFUnA	565	520	-22
PFDoA	613	569	-18
		169	-34
$^{13}C_2$-PFDoA	615	570	-34
PFTrA	663	619	-28
		169	-38
PFTeA	713	669	-24
		169	-38

Table S4. Method recoveries based on duplicate analysis of six calf-liver samples spiked with 1 to 20 ng g^{-1} of each PFAS.

Compound	Average Recovery %	Standard Deviation	RSD%
PFBS	74.7	5.29	7.08
PFHxS	97.3	6.20	6.37
PFHpS	126	15.1	12.0
PFOS	102	7.10	6.97
PFDS	81.2	19.4	23.9
FOSA	104	8.44	8.11
PFHxA	104	5.74	5.52
PFHpA	129	8.16	6.35
PFOA	102	6.01	5.88
PFNA	104	6.58	6.34
PFDA	101	5.40	5.35
PFUnA	99.7	5.27	5.29
PFDoA	103	6.10	5.92
PFTrA	84.4	37.5	44.4*
PFTeA	79.4	36.9	46.5*

*RSDs for PFTeA and PFTrA were higher (44%-47%) because of the tendency of longer-chained PFASs to sorb to surfaces during sample preparation and analysis. Such variability will bias trend analysis toward non-significant findings.[5] So, we include these compounds in all statistical analysis but results may underestimate potential changes.

Table S5. Flux of FOSA derived from the Devon ice cap, Devon Island, Nunavut, Canada[2] and corresponding air concentrations (C_A).

Year	Flux (ng m^{-2} yr^{-1})	Back-calculated C_A *(pg m^{-3})*
2007	2.07	2.4
2006	2.19	2.5
2005	3.09	3.6
2004	1.42	1.6
2003	2.45	2.8
2002	5.99	6.9
2001	15.74	18.2
2000	13.27	15.4
1999	14.37	16.7
1998	13.94	16.2
1997	18.86	21.9
1996	8.06	9.3
1995	1.51	1.8
1994	2.45	2.8
1993	3.19	3.7

[a] We used mean atmospheric FOSA concentrations from three cruise transects (North Sea: 2.66 pg m^{-3}; Canary Islands to Nfld., Canada: 2.03 pg m^{-3}; Norwegian Sea: 2.48 pg m^{-3}) to estimate atmospheric concentrations between August 2007 and January 2008 (2.39 pg m^{-3}). We use this average atmospheric value and the measurements from ice core data in 2007 of 2.07 ng m^{-2} to calculate: $C_{Air} = \alpha \cdot F$, where: $\alpha = 1.15$. For modelled values between 2007 and 2020 (Figure 5A), we assumed concentrations linearly declined to the minimum value of 1.6 pg m^{-3} in 2004, which is similar to average observations from remote high-mountain top in Zurich[6] in 2010 of 1.7 pg m^{-3}.

Figure S2. Map of satellite telemetry data from three pilot whales tagged off the coast of the Faroe Islands (show in blue) in the summer 2000. Adapted from Bloch et al.[3]

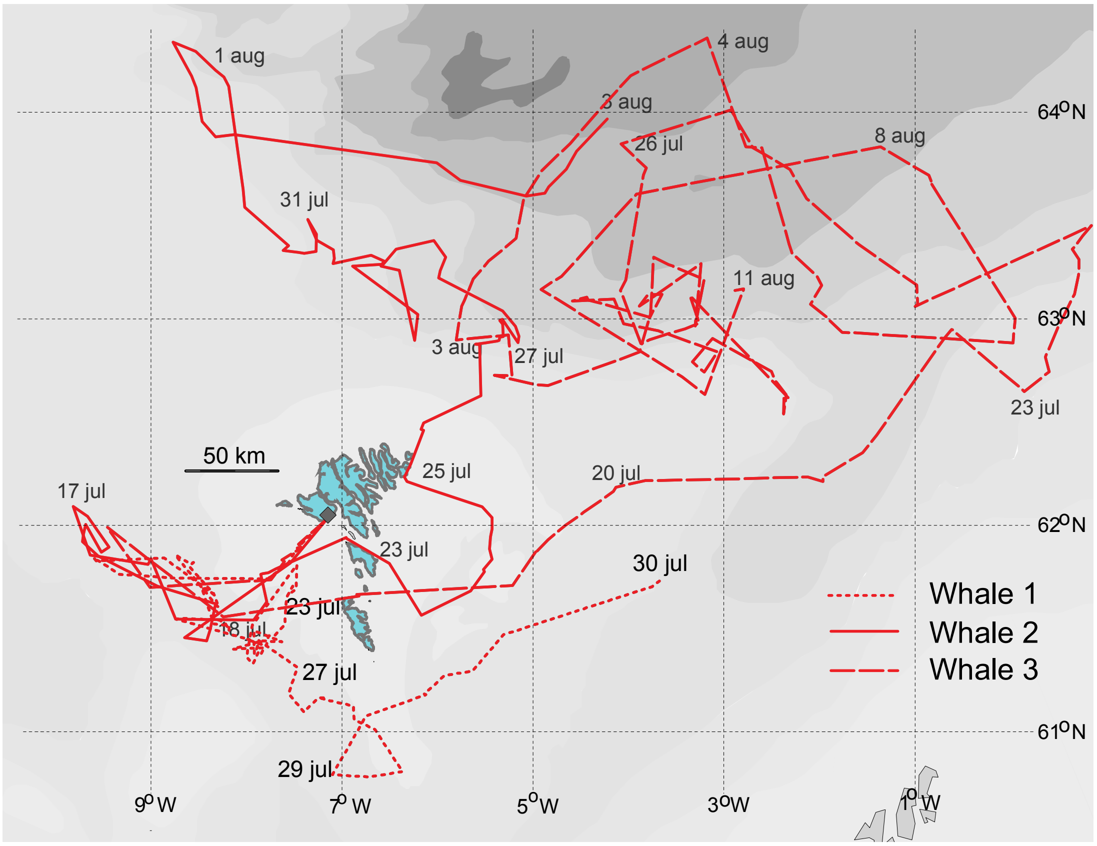

Table S6. Partitioning model for FOSA in seawater and squid.

Parameter	Description	Equation/Value	Reference
Seawater			
C_W	Seawater FOSA (pg L^{-1}),	$C_A K_{AW}$	
C_A	FOSA in air (pg m^{-3})	Table S5	
K_{AW}[a]	Air-water partitioning coefficient (unitless)	-3.6	Vierke et al.[7]
Squid			
C_S	FOSA in Squid (pg g-1 wet weight)	$\nu_{LD} K_{OW} + \nu_{ND} \beta K_{OW} + \nu_{WD}$	
ν_{LD}	Lipid fraction (unitless)	0.014	Bloch et al.[4]
ν_{ND}	Non-lipid organic matter (NLOM) content (unitless)	0.16	Bloch et al.[4]
vWD	Moisture content (unitless)	1- ν_{ND} - ν_{LD}	
$logKOW$	Octanol-water partition coefficient (unitless)	5.8	U.S. EPA[8]
$\boldsymbol{\beta}$	Sorptive capacity of NLOM relative to octanol	0.035	Arnot and Gobas[9]

[a] K_{AW} for FOSA from an experimental study[7] assuming a pKa value of 6.24.

Table S7a. Bioaccumulation model for pilot whales.

Parameter	Description	Equation	Reference
$\boldsymbol{\frac{dM_B}{dt}}$	Change in mass of FOSA over time	$W_B \cdot \left(k_A C_A + k_D \sum \left(P_i C_{D,i}\right)\right) - (k_O + k_E + k_U + +k_M) \cdot M_B$	
Growth			
W_B	Weight of pilot whale (kg)	$189.68e^{\frac{0.2802}{0.120}(1-e^{-0.12t})}$	Bloch et al.[4]
L	Length of whale (cm)	$228.92e^{0.1209/0.13(1-e^{-0.13t})}$	Bloch et al.[4]
Respiration			
k_A	Rate constant for inhalation (L/kg/day)	$\frac{E_A G_A}{W_B}$	
G_A	Inhalation rate (L/day)	$f V_T$	
f	Breaths/day	$33\, {W_B}^{-0.42}$	Limoges et al.[10]
V_T	L	$0.074{W_B}^{0.9}$	Lafortuna and Jahoda[11]
k_O	Rate constant for exhalation (1/day)	$\frac{k_A}{K_{BA}}$	
K_{BA}	Whale-Air partition coefficient (L/kg)	$(\nu_{LB}/\rho_L + \nu_{NB}\beta/\rho_{NLOM})K_{OA} + \frac{\nu_{WB}}{K_{AW}\rho_w}$	
K_{OA}	Octanol-air partition coefficient (L/kg)	$\frac{K_{OW}}{K_{AW}}$	

Table S7b. Bioaccumulation model for pilot whales cont'd.

Ingestion			
k_D	Rate constant for ingestion (1/day)	$\frac{E_D G_D}{W_B}$	
G_D	Ingestion Rate (kg/day)	$G_D = 0.123\ W_B{}^{0.8}$	Innes et al.[12]
E_D	Assimilation efficiency (%)	$(10^{-9} K_{OW} + 1.025)^{-1}$	Gobas and Arnot[13]
kE	Rate constant for fecal egestion (1/day)	$\frac{E_D G_F K_{GB}}{W_B}$	
GF	Fecal egestion rate (kg/day)	$\{(1-\varepsilon_L)\nu_{LD} + (1-\varepsilon_N)\nu_{ND} + (1-\varepsilon_W)\nu_{WD}\}G_D$	
KGB	Gut-whale partition coefficient	$(\nu_{LG}K_{OW} + \nu_{NG}K_{OW}\beta + \nu_{WG})/K_{BW}$	
$\boldsymbol{\nu_{LG}}$	Lipid fraction in gut	$(1-\varepsilon_L)\nu_{LD}/\{(1-\varepsilon_L)\nu_{LD} + (1-\varepsilon_N)\nu_{ND} + (1-\varepsilon_W)\nu_{WD}\}$	
$\boldsymbol{\nu_{NG}}$	NLOM fraction in gut	$(1-\varepsilon_N)\nu_{ND}/\{(1-\varepsilon_L)\nu_{LD} + (1-\varepsilon_N)\nu_{ND} + (1-\varepsilon_W)\nu_{WD}\}$	
$\boldsymbol{\nu_{WG}}$	Water fraction in gut	$(1-\varepsilon_W)\nu_{WD}/\{(1-\varepsilon_L)\nu_{LD} + (1-\varepsilon_N)\nu_{ND} + (1-\varepsilon_W)\nu_{WD}\}$	
$\boldsymbol{K_{BW}}$	Whale-water partition coefficient	$(\nu_{LB} + \nu_{NB}\beta)K_{OW} + \nu_{WB}$	
Urination			
kU	Rate constant for urination (1/day)	$\frac{G_U}{W_B K_{BU}}$	
KBU	Whale-urine partition coefficient	$(\nu_{LB}/\rho_L + \nu_{NB}\beta/\rho_{NLOM})K_{OW} + \nu_{WB}/\rho_W$	

Table S8. List of bioaccumulation model parameters

Parameter	Description	Value	Reference
Partition Coefficients			
$logK_{OW}$	Octanol-water partition coefficient (unitless)	5.8	U.S. EPA[8]
$logK_{AW}$	Air-water partition coefficient (unitless)	-3.7	Vierke et al.[7a]
Squid			
v_{LD}	Lipid fraction (unitless)	0.14	Bloch et al.[4]
v_{ND}	Non-lipid organic matter (NLOM) content (unitless)	0.16	Bloch et al.[4]
v_{WD}	Moisture content (unitless)	0.826	
Pilot Whale			
v_{LB}	Lipid fraction in whale (unitless)	0.21	Innes et al.[12]
v_{NB}	NLOM fraction in whale (unitless)	0.373	Innes et al.[12]
v_{WB}	Water fraction in whale (unitless)	0.417	
β	Relative sorption capacity of NLOM to that of octanol (unitless)	0.035	Arnot and Gobas[9]
E_A	Uptake efficiency in lungs (unitless)	1	Vestergren[14]; Gebbink[15]
f	Breathing rate frequency (Breaths/day)	$33\times W_B(t)^{-0.42}$	Mortola and Limoges[10]
V_T	Volume of breath (L)	$0.074\times W_B(t)^{0.9}$	Lafortuna and Jahoda[11]
E_D	Uptake efficiency in GIT (unitless)	$(A_{ED}K_{OW}+B_{ED})^{-1}$	Gobas and Arnot[13]
A_{ED}	Constant for E_D eq. (unitless)	1.0×10^{-9}	Gobas and Arnot[13]
B_{ED}	Constant for E_D eq. (unitless)	*1.025*	Gobas and Arnot[13]
ε_L	Lipid assimilation efficiency (unitless)	*0.9*	Gobas and Arnot[13]
ε_N	NLOM assimilation efficiency (unitless)	*0.5*	Gobas and Arnot[13]
ε_W	Water assimilation efficiency (unitless)	*0.55*	Gobas and Arnot[13]
G_U	Urinary excretion rate (L/day)	*300 at full size. Proportional to* W_B	Binnington et al.[16]

[a]Measured value at a waste-water treatment plant assuming pKa = 6.24.

Table S9. Results of temporal regressions for each compound adjusted and unadjusted for length of pilot whale using traditional regression for PFOS and FOSA and maximum likelihood estimation (MLE) for the remaining compounds containing non-detects. PFBS, PFHpS, PFDS, and PFHpA were infrequently detected and so were excluded from analysis.

	Unadjusted Model		Adjusted Model			
Compound	Slope (yr^{-1})	p-value	Slope	p-value	Length (cm^{-1})	p-value
PFBS	-	-	-	-	-	-
PFHxS	-0.54	0.89	-0.87	0.82	-0.2%	0.49
PFHpS	-	-	-	-	-	-
PFOS	2.8%	<0.0001	3.0%	<0.0001	-0.3%	0.03
PFDS	-	-	-	-	-	-
FOSA (Before 2003)	7.4%	0.04	7.4%	0.06	0.3%	0.97
FOSA (After 2003)	-13.0%	<0.0001	- 12.6%	<0.0001	0.2%	0.08
PFHxA	-	-	-	-	-	-
PFHpA	-	-	-	-	-	-
PFOA	-3.3%	0.052	-3.1%	0.07	-0.3%	0.30
PFNA	6.6%	<0.0001	7.0%	<0.0001	-0.4%	0.06
PFDA	8.2%	<0.0001	8.3%	<0.0001	-0.2%	0.28
PFUnA	6.1%	<0.0001	6.2%	<0.0001	-0.06%	0.66
PFDoA	4.4%	<0.0001	4.4%	<0.0001	-0.009%	0.95
PFTrA	7.4%	0.0003	7.5%	0.004	0.05%	0.93
PFTeA	9.8%	0.67	9.8%	0.67	-0.07%	0.96

Table S10. Measured concentrations (pg g^{-1} wet weight) of FOSA in five European flying squid (*Todarodes sagittatus*) caught off the coast of the Faroe Islands in 2010.

ID	FOSA
S1	232
S2	260
S3	177
S4	386
S5	363
Mean	**284**

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