Supporting Information

Spatiotemporal Analysis of Perfluoroalkyl Substances in White-Tailed Eagle (*Haliaeetus albicilla)* Nestlings from Northern Norway – A Ten-Year Study

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Number of Pages: 18

Number of Figures: 4

Number of Tables: 8

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MATERIALS AND METHODS

Limit of detection (LOD)

The samples measured under LOD are important and sometimes kept in statistical analyses as they represent low measurements. In the present study, due to a change in the spectrometer used for PFAS measurements, the amount of points measured under LOD in the first five years of the decade is much higher than on the last five years. In addition, due to PFAS analyses being conducted in different years, the LOD varied in an important manner among years (LODs for each year and each homologue can be found in SI Table S3). Removing points under LOD might weaken the model selection by discarding the lowest points of the dataset in the first years only, which could lead to untrue decreasing time trends. In the present study, all years when more than 50% of the points were lower than the LOD of the specific year were removed. Then, all points under LOD were set to half of the maximum LOD for each compound independently. This allowed a straightforward comparison between years for each congener as the LOD has the same weight on each year of the decade for the temporal trend analysis. To ensure the robustness and the reliability of the selected models, we ran the model selection on a dataset where LODs were kept as they were measured originally, after removing years when more than 50% of the points were lower than LOD of the specific year. The same results were obtained for all selected PFAS (PFOA, PFNA, PFUnDA, PFTrDA and PFOS) apart from PFDA spatial trends that was decreasing in the study's southern part only when the highest LOD was used. When the original LODs were used, it showed no trends in both regions. This was clearly due to the important weight of points under LOD in the first years of the decade. Consequently, to avoid any misinterpretation of PFDA time trends, we removed this compound from the analyses. PFHxS temporal trends model output was also different between both methods. When the highest LOD was kept, PFHxS contamination was decreasing with

distance to the airports in both regions but the southern region had lower concentrations than the northern one. This is consistent with the results of the spatial analyses. However, when LOD were kept as their original measurements, the same decrease was observed but the difference in concentrations between both regions was not significant. Here the difference is negligible and this contaminant was included in the analyses. We conclude that keeping the highest LOD for each compound was the best method to lower the bias in the model selection without removing too much information from the dataset.

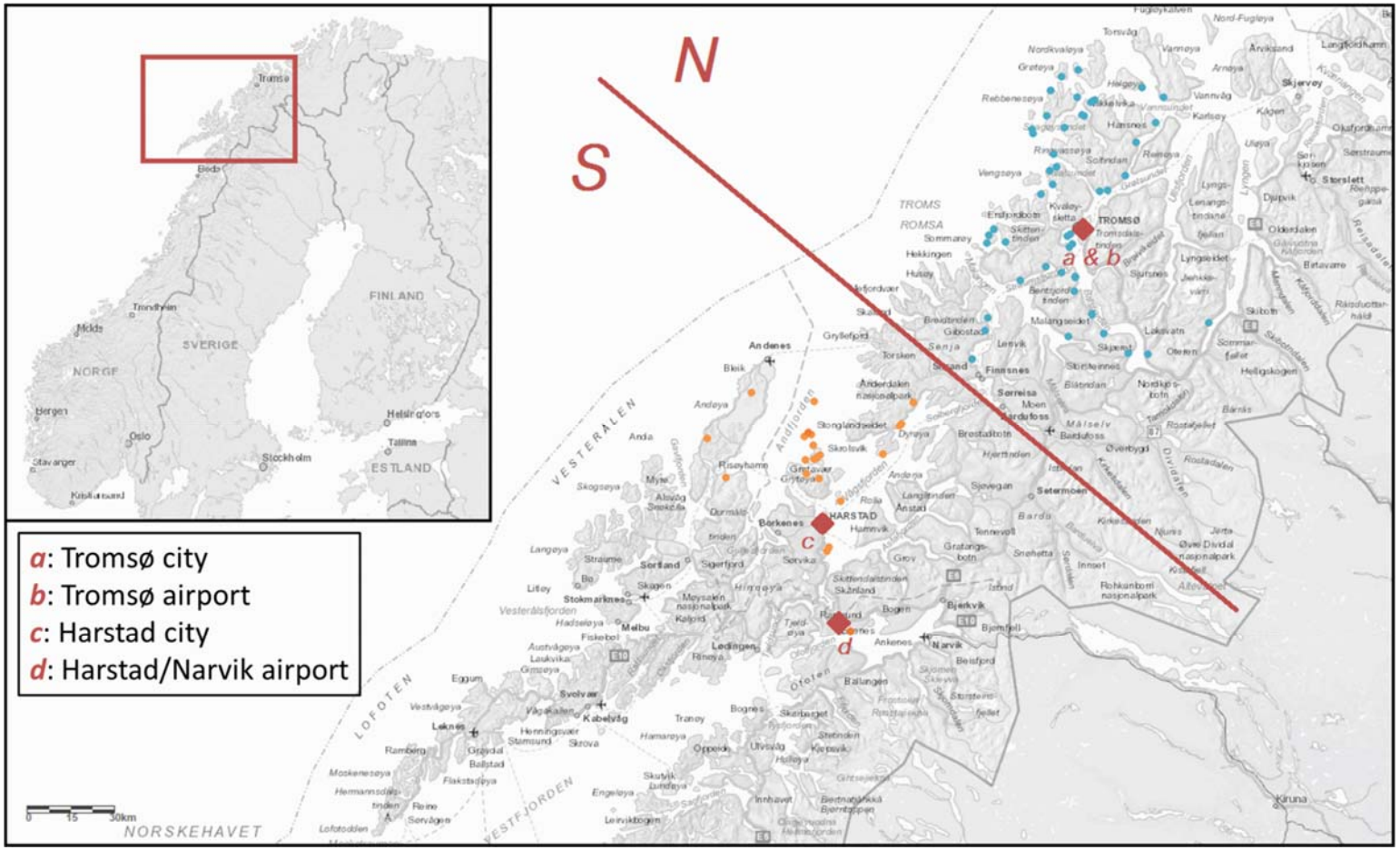

Figure S1. Map identifying the Troms County (68-70°N 15-22°E) in northern Norway. The region is divided in two regions (North: *N* and South: *S*) by the red line; blue points marked nests of the northern region, and orange points nests of the southern region (*a*: Tromsø city, *b*: Tromsø airport, *c*: Harstad city, *d*: Harstad/Narvik airport).

Table S1. List of targeted PFAS: their groups (PFCAs: perfluorinated carboxylic acids; PFSAs: perfluoroalkane sulfonates), abbreviations, chemical names, structures, Molecular weights and CAS-Number.

Group	Abbreviation	Chemical Name	Structure	Molecular Weight	CAS-number
PFCAs	PFOA	Perfluorooctanoic acid	$F(CF_2)_7COOH$	414.1	335-67-1
	PFNA	Perfluorononanoic acid	$F(CF_2)_8COOH$	464.1	375-95-1
	PFDA	Perfluorodecanoic acid	$F(CF_2)_9COOH$	514.1	335-76-2
	PFUnDA	Perfluoroundecoic acid	$F(CF_2)_{10}COOH$	564.1	2058-94-8
	PFDoDA	Perfluorododecanoic acid	$F(CF_2)_{11}COOH$	614.1	307-55-1
	PFTrDA	Perfluorotridecanoic acid	$F(CF_2)_{12}COOH$	664.1	72629-94-8
PFSAs	PFHxS	Perfluorohexane sulfonic acid	$F(CF_2)_6SO_3H$	400.1	355-46-4
	PFOS	Perfluorooctane sulfonic acid	$F(CF_2)_8SO_3H$	500.1	1763-23-1

Tables S2. List of standard reference material inter-day and intra-day variations for each PFAS for the analyses of samples collected during years 2013-2016, average and standard deviations (*SD*) are expressed as pg g^{-1} and relative *SD* (*RSD*) is expressed as %.

	inter-day variation (*n* = 4)			**intra-day variation (*n* = 3)**		
	average	***SD***	***RSD***	**average**	***SD***	***RSD***
PFOA	17783	788	4.4	17230	1528	8.9
PFNA	1973	505	25.6	1800	275	15.3
PFUnA	2690	374	13.9	2727	424	15.6
PFDcA	247	52	21.0	242	75	30.9
PFHxS	3230	659	20.4	2933	792	27.0
PFOS	27959	2271	8.1	26835	3811	14.2

Table S3. Detection feature of targeted PFAS: their groups, abbreviations, detection percentages and limits of detection (ng g^{-1}) for the six different periods. Compounds included in further analyses are given in bold.

Group	Abbreviation	Detection (%)	Limit of Detection					
			2008	2009	2010	2011-2012	2013-2016	2017
PFCAs	**PFOA**	**77.58**	**0.30**	**2.30**	**1.40**	**0.30**	**0.02**	**0.05**
	PFNA	**95.15**	**0.24**	**0.40**	**1.10**	**0.30**	**0.09**	**0.09**
	PFDA	80.61	0.70	0.30	1.98	0.30	0.05	0.05
	PFUnDA	**86.67**	**0.80**	**0.80**	**4.60**	**0.12**	**0.03**	**0.03**
	PFDoDA	67.27	0.10	0.30	1.50	0.10	0.04	0.06
	PFTrDA	**74.55**	**0.20**	**0.40**	**0.90**	**0.60**	**0.06**	**0.06**
PFSAs	**PFHxS**	**83.03**	**0.10**	**0.30**	**1.20**	**0.50**	**0.10**	**0.10**
	PFOS	**98.79**	**2.00**	**0.60**	**0.80**	**0.18**	**0.18**	**0.18**

Table S4. Descriptive statistics (mean ± standard error (*SE*), median and range (min-max)) for each PFAS concentrations (ng g^{-1} ww) in plasma of white-tailed eagle nestlings from northern Norway for each year of the study period. "na" means that no samples have been measured above the limit of detection, "Measurements <LOD" is the number of points lower than the limit of detection, if ">50%" then this year was not included in the analyses.

PFOA

	Mean ± *SE*	**Median**	**Min-max**	***n***	**Measurements <LOD**
2008	2.06 ± 0.28	2.30	1.30-2.80	5	0
2009	na	na	na	0	>50%
2010	na	na	na	0	>50%
2011	0.81 ± 0.16	0.79	0.15-2.48	19	7
2012	0.93 ± 0.21	0.74	0.15-2.65	16	6
2013	1.39 ± 0.20	1.43	0.15-2.91	15	0
2014	1.17 ± 0.13	1.02	1.03-3.70	18	0
2015	1.52 ± 0.15	0.66	0.15-1.04	16	0
2016	1.34 ± 0.52	0.52	0.15-7.79	20	1
2017	0.75 ± 0.09	0.78	0.15-1.72	21	0

PFNA

	Mean ± *SE*	**Median**	**Min-max**	***n***	**Measurements <LOD**
2008	4.32 ± 0.12	4.30	3.90-4.60	5	0
2009	3.32 ± 0.37	2.80	0.55-7.50	19	1
2010	2.69 ± 0.42	2.50	0.55-5.50	15	2
2011	3.15 ± 0.66	2.25	0.55-12.9	19	0
2012	2.87 ± 0.64	2.14	0.55-9.44	16	5
2013	5.77 ± 1.44	3.46	0.55-22.8	15	0
2014	4.52 ± 0.37	1.90	1.025-3.70	18	0
2015	1.52 ± 0.15	3.16	1.66-10.6	16	0
2016	4.36 ± 0.73	3.49	0.55-11.8	20	0
2017	3.47 ± 0.62	2.66	0.55-13.7	21	0

PFUnDA

	Mean ± *SE*	Median	Min-max	*n*	Measurements <LOD
2008	2.54 ± 0.66	3.00	0.40-4.20	5	1
2009	3.98 ± 1.19	2.50	0.40-20.40	19	6
2010	na	na	na	0	>50%
2011	6.26 ± 0.72	6.00	1.09-13.4	19	0
2012	4.25 ± 0.60	4.19	1.01-8.83	16	0
2013	4.46 ± 0.57	4.09	1.38-8.81	15	0
2014	4.70 ± 0.40	1.90	1.03-3.70	18	0
2015	1.52 ± 0.15	4.56	2.05-5.99	16	0
2016	3.38 ± 0.24	3.05	1.71-5.24	20	0
2017	3.14 ± 0.36	2.94	1.00-7.35	21	0

PFTrDA

	Mean ± *SE*	Median	Min-max	*n*	Measurements <LOD
2008	0.64 ± 0.15	0.70	0.30-1.10	5	2
2009	na	na	na	0	>50%
2010	na	na	na	0	>50%
2011	4.09 ± 0.68	3.97	0.30-11.15	19	3
2012	2.00 ± 0.59	1.25	0.30-7.77	16	6
2013	1.82 ± 0.60	1.04	0.30-8.89	15	0
2014	1.82 ± 0.18	1.90	1.03-3.70	18	0
2015	1.52 ± 0.15	2.29	0.30-3.28	16	0
2016	1.79 ± 0.14	1.81	0.84-2.98	20	0
2017	0.93 ± 0.20	0.30	0.30-3.22	21	1

PFHxS

	Mean ± *SE*	Median	Min-max	*n*	Measurements <LOD
2008	2.66 ± 0.44	2.40	2.00-4.40	5	0
2009	1.75 ± 0.34	1.60	0.25-6.80	19	1
2010	na	na	na	0	>50%
2011	0.74 ± 0.12	0.52	0.25-1.75	19	7
2012	na	na	na	0	>50%
2013	1.54 ± 0.28	0.90	0.25-3.18	15	0
2014	1.45 ± 0.14	1.90	1.03-3.70	18	0
2015	1.52 ± 0.15	0.83	0.25-2.34	16	0
2016	1.36 ± 0.33	0.78	0.25-4.65	20	0
2017	0.61 ± 0.08	0.61	0.25-1.84	21	1

	PFOS				
	Mean ± *SE*	**Median**	**Min-max**	***n***	**Measurements <LOD**
2008	40.18 ± 5.95	40.80	22.3-59.1	5	0
2009	48.51 ± 13.82	28.10	1.00-247	19	1
2010	29.75 ± 4.32	25.00	9.60-56.4	15	0
2011	46.34 ± 7.13	46.74	6.07-133	19	0
2012	52.53 ± 15.32	34.43	1.00-248	9	1
2013	29.83 ± 7.93	17.62	6.72-118	15	0
2014	27.75 ± 2.75	1.90	1.03-3.70	18	0
2015	1.52 ± 0.15	17.21	11.8-41.0	16	0
2016	21.22 ± 2.58	19.88	3.85-44.8	20	0
2017	18.45 ± 1.73	16.93	8.02-44.7	21	0

Table S5. Model selection for temporal trends based on the lowest second-order Akaike's information criterion corrected for small sample sizes (AICc). The predicting variables were year, geographical region (Region), the square root of the sampling year (Year²) and the interaction between sampling year and region (Year × Region). The most parsimonious models are given in bold. w_i: Akaike's weight; Δ_i: difference between the model with the smallest AICc-value and the model of interest (*i*).

	Year	Region	Year x Region	Year²	K	AICc	w_i	ΔAICc
PFOA								
Mod4	**X**				**4**	**338.47**	**0.43**	**0.00**
Mod6	X			X	5	339.58	0.24	1.11
Mod3	X	X			5	340.54	0.15	2.97
Mod5	X	X		X	6	341.66	0.09	3.19
Mod2	X	X	X		6	342.28	0.06	3.81
Mod1	X	X	X	X	7	343.82	0.03	5.36
PFNA								
Mod4	**X**				**4**	**335.18**	**0.42**	**0.00**
Mod6	X			X	5	336.90	0.18	1.71
Mod3	X	X			5	337.11	0.16	1.92
Mod1	X	X	X	X	7	337.76	0.11	2.58
Mod2	X	X	X		6	338.81	0.07	3.62
Mod5	X	X		X	6	338.86	0.07	3.67
PFUnDA								
Mod1	X	X	X	X	7	259.54	0.46	0.00
Mod6	**X**			**X**	**5**	**259.86**	**0.39**	**0.32**
Mod5	X	X		X	6	261.87	0.14	2.33
Mod4	X				4	270.87	0.00	11.34
Mod3	X	X			5	272.65	0.00	13.11
Mod2	X	X	X		6	274.02	0.00	14.48

PFTrDA								
Mod6	**X**			**X**	**5**	**323.26**	**0.66**	**0.00**
Mod5	X	X		X	6	325.36	0.23	2.10
Mod1	X	X	X		7	327.59	0.08	4.33
Mod4	X				4	330.39	0.02	7.13
Mod3	X	X			5	332.51	0.01	9.25
Mod2	X	X	X		6	333.18	0.00	9.92
PFHxS								
Mod2	X	X	X		6	298.49	0.41	0.00
Mod3	**X**	**X**			**5**	**298.94**	**0.33**	**0.46**
Mod1	X	X	X		7	300.70	0.14	2.22
Mod5	X	X		X	6	300.80	0.13	2.32
Mod4	X				4	312.68	0.00	14.19
Mod6	X			X	5	314.80	0.00	16.31
PFOS								
Mod4	**X**				**4**	**360.06**	**0.50**	**0.00**
Mod6	X			X	5	363.12	0.18	2.06
Mod3	X	X			5	362.18	0.17	2.12
Mod5	X	X		X	6	364.26	0.06	4.2
Mod2	X	X	X		6	364.33	0.06	4.27
Mod1	X	X	X		7	366.44	0.02	6.38

Table S6. Model selection for spatial trends. These analyses are similar to the temporal analyses above except that sampling year was replaced with the distance to the nearest airport (Dist; see Table S5 for further details).

	Dist	Region	Dist x Region	K	AICc	w_i	ΔAICc
PFOA							
Mod3	**X**			**4**	**335.55**	**0.61**	**0.00**
Mod2	X	X		5	337.11	0.28	1.57
Mod1	X	X	X	6	338.81	0.12	3.26
PFNA							
Mod3	**X**			**4**	**335.91**	**0.55**	**0.00**
Mod1	X	X	X	6	337.43	0.26	1.52
Mod2	X	X		5	338.03	0.19	2.21
PFUnDA							
Mod3	**X**			**4**	**271.12**	**0.56**	**0.00**
Mod2	X	X		5	272.24	0.32	1.12
Mod1	X	X	X	6	274.25	0.12	3.13
PFTrDA							
Mod3	**X**			**4**	**331.86**	**0.68**	**0.00**
Mod2	X	X		5	333.99	0.24	2.13
Mod1	X	X	X	6	336.12	0.08	4.26
PFHxS							
Mod2	**X**	**X**		**5**	**304.03**	**0.66**	**0.00**
Mod1	X	X	X	6	305.43	0.33	1.41
Mod3	X			4	311.55	0.02	7.52
PFOS							
Mod3	**X**			**4**	**368.44**	**0.66**	**0.00**
Mod2	X	X		5	370.36	0.25	1.92
Mod1	X	X	X	6	372.50	0.09	4.05

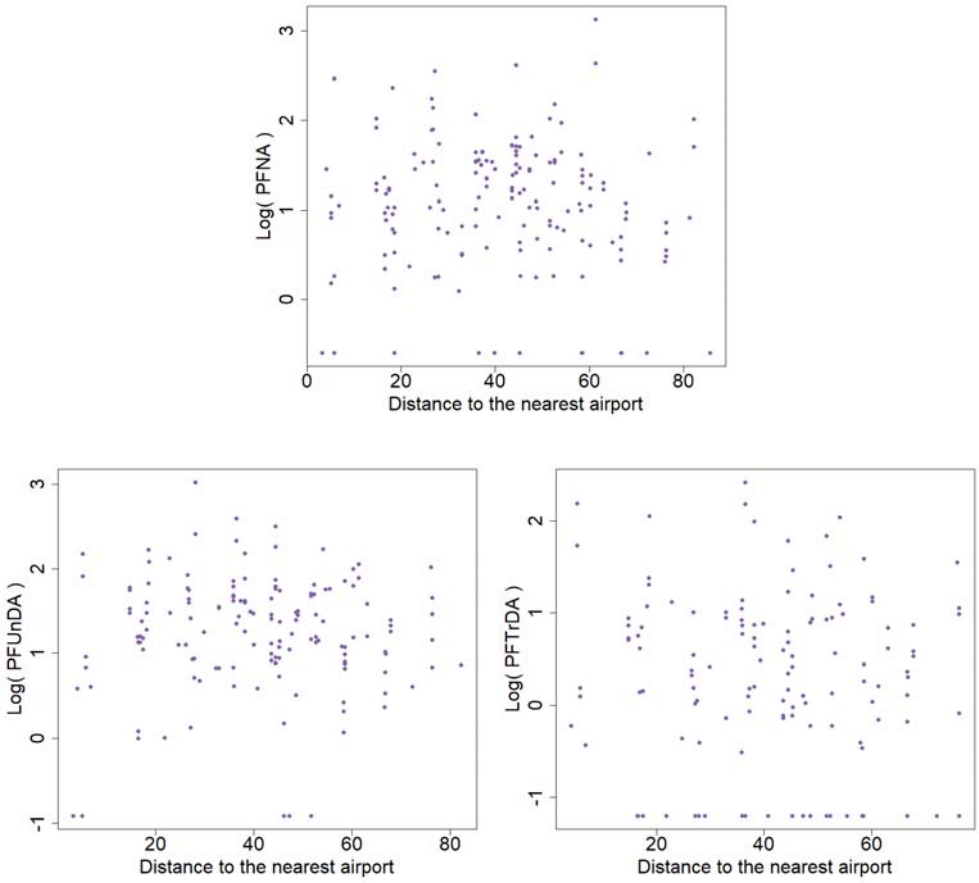

Figure S2. Spatial trends of the concentrations of PFNA, PFUnDA and PFTrDA in white-tailed eagle nestlings from northern Norway with distance to the nearest airport (km). No lines means no significant trend.

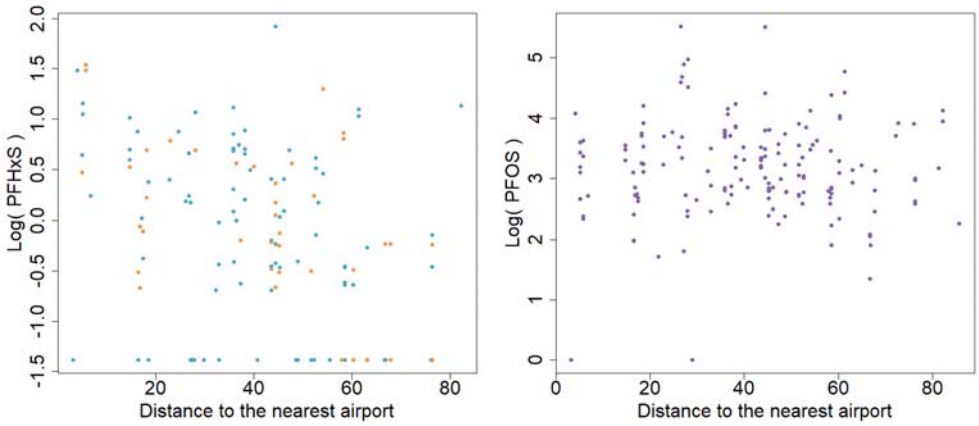

Figure S3. Spatial trends of the concentrations of PFHxS and PFOS in white-tailed eagle nestlings from northern Norway with distance to the nearest airport (km), orange points for the southern region and blue points for the northern one, purple is used if the trend is similar in both regions, no lines means no significant trend.

PFDoDA temporal variations:

Tables S7 and S8 and Figure S4 are related to PFDoDA temporal trends. Too few samples have been measured above the limit of detection (LOD; 0.10-1.50 ng g^{-1}) between years 2008 and 2012 (30% of the samples above LOD). A lower LOD (0.04-0.06 ng g^{-1}) used from 2013 to 2017 allowed a higher detection (97% of the samples above LOD). Consequently, years from 2013 to 2017 can be used for model selection and analyses.

Year was the only variable in the model explaining the temporal variation in C_{12} PFCA, the quadratic time component was also included (SI Table S7). The trend is positive until 2015, when the breaking point occurs, from 2015 to 2017 the trend is negative (SI Table S8 and Figure S4). This trend is similar to C_{11} and C_{13} PFCAs pattern on the last years of the study (see in the article for discussion of the results). This is not surprising as all those compounds were - since their direct production has stopped - mainly products of the same fluorotelomers precursors degradation.[1,2]

Table S7. Model selection for PFDoDA temporal trends (see Table S5 for technical details; model selection on the period 2013-2017 only).

	Year	Region	Year x Region	Year²	K	AICc	w_i	ΔAICc
PFDoDA								
Mod6	**X**			**X**	**5**	**207.72**	**0.61**	**0.00**
Mod5	X	X		X	6	209.23	0.29	1.51
Mod1	X	X	X	X	7	211.38	0.10	3.66
Mod4	X				4	216.45	0.01	8.72
Mod3	X	X			5	218.67	0.00	10.95
Mod2	X	X	X		6	220.92	0.00	13.20

Table S8. Factors affecting temporal trends of PFDoDA concentrations (ng g^{-1} ww) in white-tailed eagle chicks from northern Norway, estimated by mixed linear regression models on the period 2013-2017.

PFDoDA (R^2m: 0.17, R^2c: 0.58)				
Year	772	228	3.39	<0.01
Year²	-0.20	0.06	-3.39	<0.01

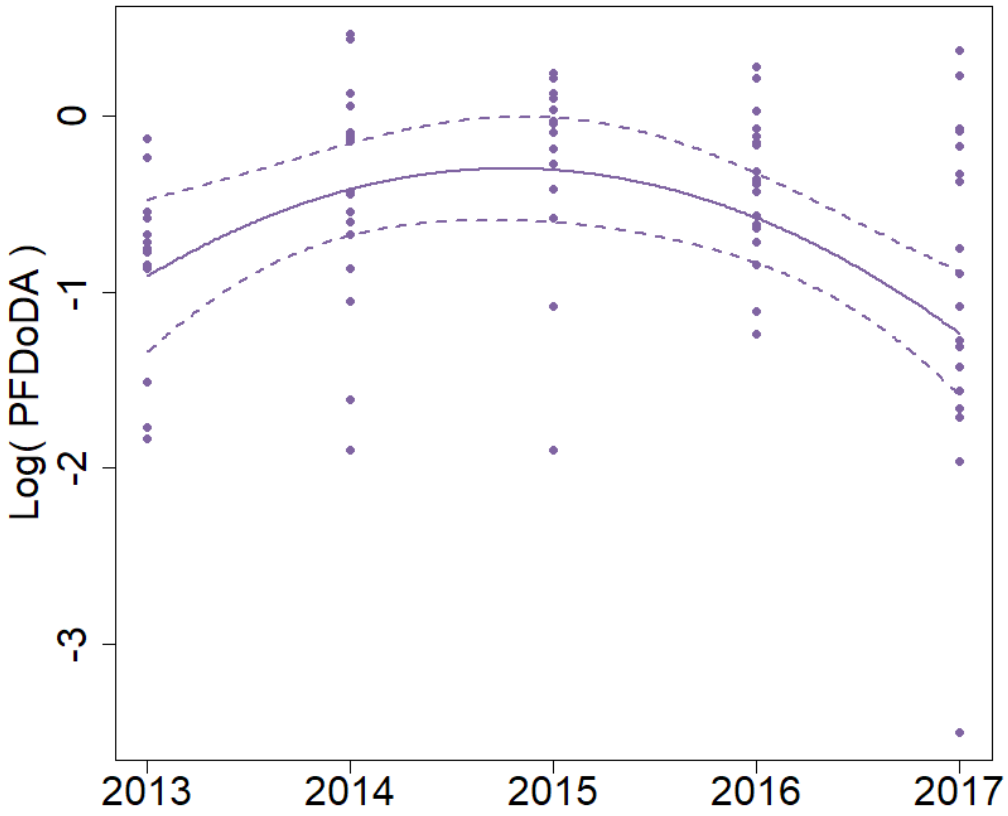

Figure S4. Temporal trends for the concentrations of PFDoDA in white-tailed eagle nestlings from northern Norway on the period 2013-2017. The solid line refers to a statistically significant time trend (and are based on the selected models presented in SI Table S8), lined by the dotted lines representing 95% Confident Intervals.

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