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## A Bad Start in Life? Maternal Transfer of Legacy and Emerging Poly- and Substances to Eggs in an Arctic Seabird

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1 A bad start in life? Maternal transfer of legacy and  
2 emerging poly- and perfluoroalkyl substances to  
3 eggs in an Arctic seabird

4  
5 *William Jouanneau<sup>1,2,\*</sup>, Don-Jean Léandri-Breton<sup>1,3</sup>, Alexandre Corbeau<sup>4</sup>, Dorte Herzke<sup>5</sup>,*  
6 *Børge Moe<sup>6</sup>, Vladimir A. Nikiforov<sup>5</sup>, Geir W. Gabrielsen<sup>2</sup>, Olivier Chastel<sup>1</sup>*

7  
8 <sup>1</sup> Centre d'Etudes Biologiques de Chizé (CEBC), UMR 7372 CNRS - La Rochelle Université,  
9 17031 La Rochelle, France

10 <sup>2</sup> Norwegian Polar Institute, Fram Centre, NO-9296 Tromsø, Norway

11 <sup>3</sup> Department of Natural Resource Sciences, McGill University, Ste Anne-de-Bellevue, QC  
12 H9X 3V9, Canada

13 <sup>4</sup> ECOBIO (Ecosystèmes, biodiversité, évolution), UMR 6553 CNRS - Université de Rennes,  
14 35000 Rennes, France

15 <sup>5</sup> NILU – Norwegian Institute for Air Research, Fram Centre, NO-9296 Tromsø, Norway

16 <sup>6</sup> NINA – Norwegian Institute for Nature Research, NO-7485 Trondheim, Norway

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19 **Key-word:** black-legged kittiwake, *Rissa tridactyla*, top predator, Svalbard, PFAS, emerging  
20 contaminants

21 ABSTRACT

22 In birds, maternal transfer is a major exposure route for several contaminants, including poly-  
23 and perfluoroalkyl substances (PFAS). Little is known, however, about the extent of the transfer  
24 of the different PFAS compounds to the eggs, especially for alternative fluorinated compounds.  
25 In the present study we measured legacy and emerging PFAS, including Gen-X, ADONA and  
26 F-53B, in the plasma of pre-laying black-legged kittiwake females breeding in Svalbard and  
27 the yolk of their eggs. We aimed to 1/ describe the contaminant levels and patterns in both  
28 females and eggs, and 2/ investigate the maternal transfer, i.e. biological variables and the  
29 relationship between the females and their eggs for each compound. Contamination of both  
30 females and eggs were dominated by linPFOS then PFUnA or PFTriA. We notably found 7:3  
31 fluorotelomer carboxylic acid - a precursor of long-chain carboxylates - in 84% of the egg yolks,  
32 and provide the first documented finding of ADONA in wildlife. Emerging compounds were  
33 all below the detection limit in female plasma. There was a linear association between females  
34 and eggs for most of the PFAS. Analyses of maternal transfer ratios in females and eggs suggest  
35 that the transfer is increasing with PFAS carbon chain length, therefore the longest chain  
36 perfluoroalkyl carboxylic acids (PFCAs) were preferentially transferred to the eggs. The mean  
37  $\sum_{\text{PFAS}}$  in the second-laid eggs was 73% of that in the first-laid eggs. Additional effort on  
38 assessing the outcome of maternal transfers on avian development physiology is essential,  
39 especially for PFCAs and emerging fluorinated compounds which are under-represented in  
40 experimental studies.

41

42 SYNOPSIS

43 Investigating the maternal transfer of PFAS in avian top predators is an essential steps in  
44 understanding the adverse effects of these compounds on wildlife.

## 45 INTRODUCTION

46 Poly- and perfluoroalkyl substances (PFAS) are synthetic chemicals widespread  
47 globally.<sup>1</sup> Since the 1950s, thousands of compounds were developed in this family and used in  
48 a multitude of manufactured products (firefighting foams, waterproof clothing, non-stick  
49 cookware, coatings, food packaging, personal care products, dental floss, electronics, metal  
50 plating and even pesticides) owing to their extremely stable chemical structure and their  
51 surfactant properties. After they have been revealed as ubiquitous and toxic for organisms in  
52 the early 2000s, some of the historical most noxious compounds such as  
53 perfluorooctanesulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) were phased out by  
54 the main manufacturers and recommended for elimination by the Stockholm Convention on  
55 Persistent Organic Pollutants.<sup>2-5</sup>

56 Some of the PFAS bioaccumulate in organisms and biomagnify along trophic chains,  
57 particularly in aquatic ecosystems.<sup>6, 7</sup> As a consequence, high trophic level aquatic organisms  
58 including seabirds often have elevated tissue concentrations.<sup>1, 8</sup> Oceanic and atmospheric  
59 transport can contribute to the long-range dispersal of PFAS, their precursors and breakdown  
60 products<sup>9-15</sup> which are found in wildlife tissues worldwide, including remote areas like the  
61 Arctic.<sup>8, 16</sup>

62 In birds, during egg formation, the female transfers various substances required to  
63 enable and sustain the development of the embryo. However, in addition to essential compounds  
64 such as water, minerals, proteins, lipids, vitamins, antibodies or hormones, some contaminants  
65 may also be transferred. Persistent organic pollutants (POPs) have been shown to be transferred  
66 from mothers to eggs due to their physicochemical properties, via either lipids (e.g.  
67 polychlorinated biphenyls (PCBs), pesticides) or proteins (PFAS).<sup>17</sup> Therefore, the avian egg is  
68 considered as a relevant monitoring tool for a selection of organic contaminants exposure in  
69 many species, as its composition directly reflects that of maternal tissues.<sup>18</sup> However, the few

70 field studies concurrently comparing variability in, for instance, PCBs and organochlorine  
71 pesticides concentrations and composition in both eggs and mother tissues, showed that organic  
72 contaminants transfer can be influenced by a combination of biological factors (ex: egg size,  
73 egg mass, egg laying order, number of eggs in clutch, age of the female) and by the  
74 physicochemical properties of contaminants.<sup>17</sup> Despite PFAS being studied for almost two  
75 decades, their concentrations in egg have been poorly documented in wildlife, and very few  
76 studies evaluated maternal transfers in birds by concurrently measuring PFAS in the eggs and  
77 the mother in the field.<sup>19-23</sup> Although fluorinated compounds are transferred at a lower degree  
78 than lipophilic contaminants, they can still be found at high concentrations in eggs.<sup>19, 24-26</sup> Once  
79 accumulated into females, PFAS tend to bind to proteins (especially very low-density  
80 lipoproteins) synthesized in the liver, these being then transferred via blood to the ovary and  
81 finally to the eggs and the carbon-chain length has been suggested to be an important driver of  
82 PFAS transfer efficiency.<sup>27-29</sup>

83 In humans and in laboratory models, PFAS can cause cancer, affect immuno-  
84 competence and disrupt the endocrine system.<sup>30</sup> Regarding developing avian embryos, some  
85 PFAS compounds were found to be responsible for a lower heart rate, an enlarged liver, thyroid  
86 hormones and immune system disruption, as well as a lower hatching success and a lower  
87 survival.<sup>31-37</sup> However, the consequences of PFAS exposure remain poorly investigated in  
88 wildlife. Recent studies on kittiwakes and other seabirds of the Kongsfjord area suggest that  
89 exposure to long-chain perfluorinated carboxylic acids (PFCAs) can be associated with  
90 physiological and fitness impairments.<sup>38-43</sup> PFAS cover thousands of substances, the vast  
91 majority are not regulated yet and some are increasingly detected in biota.<sup>44</sup> Furthermore,  
92 development and manufacturing of alternatives to the legacy PFAS remains largely  
93 uncharacterized in terms of risks, despite recent evidences of their environmental occurrence in  
94 wildlife tissues.<sup>45, 46</sup> Alternative fluorinated compounds are now continuously introduced to the

95 market and despite many of them being still unidentified, some are now known as  
96 bioaccumulative.<sup>47</sup> As an example, emerging fluorinated chemicals such as F-53B, ADONA or  
97 HFPO-DA (Gen-X) and short-chained precursors are part of these alternative PFAS which have  
98 been very scarcely screened for in wildlife before.<sup>48</sup> To the best of our knowledge, to date only  
99 three studies detected emerging PFAS in avian eggs.<sup>49-51</sup>

100 In the present study we aimed to: 1/ describe concentration and composition of emerging  
101 and legacy PFAS in the egg yolk of an Arctic-breeding population of black-legged kittiwakes  
102 (*Rissa tridactyla*) facing significant PFAS exposure<sup>43</sup>; 2/ investigate the maternal transfer, i.e.  
103 the relationship between emerging and legacy PFAS found in the eggs yolk with those found in  
104 the plasma of the corresponding females sampled during the pre-laying stage (i.e. before egg-  
105 laying) as well as the effects of biological variable (laying date, clutch size, laying order, female  
106 condition). We expect strong correlations of contaminant concentrations between females and  
107 their eggs with highly contaminated adults laying highly contaminated eggs, but a decrease in  
108 eggs contamination with laying order.<sup>52</sup> We also hypothesize that variations in the transfer of  
109 PFAS to the egg should be influenced by the physicochemical properties of the different PFAS  
110 compounds, especially the carbon chain length.

111

## 112 MATERIALS AND METHODS

### 113 **Sample collection**

114 The study was conducted in a Black-legged kittiwake colony (“Krykkjefjellet” in  
115 Kongsfjorden, Svalbard, 78°53'48"N 12°11'43"E) from May to June 2019. On fourteen nests,  
116 adult females of black-legged kittiwake (hereafter “kittiwakes”) were captured a few days  
117 before expected laying using a noose at the end of a fishing rod. Females of each nest ( $n = 14$ )  
118 were individually identified using a numbered metal ring (Stavanger Museum) and a three digits  
119 plastic ring to allow individual identification from a distance. Molecular sexing of the adults

120 was performed at the Centre d'Etude Biologiques de Chizé (CEBC), following Fridolfsson and  
121 Ellegren (1999).<sup>53</sup> Immediately after capture, a 2 mL blood sample was taken from the alar vein  
122 using a heparinized 2.5 mL syringe and a 25G needle. Blood was then transferred in a 2 mL  
123 Eppendorf tube and stored in a cooler. At the end of each day, the samples were centrifuged for  
124 10 minutes. Both tubes of red blood cells (RBC, for molecular sexing) and plasma (for PFAS)  
125 were stored in a -20 °C freezer until analyses. The nests corresponding to the sampled birds  
126 were then monitored daily using a mirror attached to an 8 m rod. As soon as the first egg was  
127 laid, it was collected ( $n = 14$ ) and replaced by a plastic dummy-egg in order to avoid nest  
128 desertion. Similarly, the second-laid egg was collected ( $n = 11$ ) the day it was laid and the  
129 dummy-egg was removed. If they were no second-laid egg after 10 days, the dummy-egg was  
130 retrieved. All egg samples were processed in the lab immediately. The eggs were measured  
131 (height and diameter) with an electronic caliper and weighted to the nearest 0.01g. Eggs were  
132 opened and the yolk and the albumen were separated and stored in 2 mL Eppendorf tubes at -  
133 20°C in a freezer until assayed. All eggs were collected less than 24h after they were laid, and  
134 thus were at a similar development stage, avoiding contaminants to be absorbed by the growing  
135 embryo and allowing a comparison between eggs. Sample size was limited by authorization to  
136 collect eggs and by the number of accessible nests in the colony. The sampling of birds and  
137 eggs was approved by the Governor of Svalbard and by the Norwegian Animal Research  
138 Authority (NARA, permit number 19970).

139

#### 140 **Poly and perfluoroalkyl substances extraction and analysis**

141 We used a method described by Sletten *et al.* (2016) adapted from Powley *et al.* (2005)  
142 to measure PFAS in females' plasma.<sup>54, 55</sup> For yolk samples (~0.20 g), after mixing methanol  
143 and the samples spiked with internal standards (listed in the SI), an additional initial step  
144 consisting in thoroughly mixing the solution using zirconium beads was added. This step

145 ensured that yolk was completely homogenized with the solvent. PFAS concentrations were  
146 only measured in yolk as this is where the most part of these contaminants are located in bird  
147 eggs.<sup>21, 29</sup> Quantification was conducted by ultrahigh-performance liquid chromatography  
148 triple-quadrupole mass spectrometry (UHPLC–MS/MS). The chromatograms were quantified  
149 with LCQuan software (version 2.6, Thermo Fisher Scientific Inc., Waltham, MA, USA),  
150 applying the isotopic dilution method. An eight-point calibration curve with a concentration  
151 range from 0.02 to 10.0 pg  $\mu\text{L}^{-1}$  was used. The following “legacy” PFAS were screened in both  
152 plasma and yolk: perfluorooctane sulfonamide (FOSA), perfluorobutanesulfonic acid (PFBS),  
153 perfluoropentanesulfonic acid (PFPS), perfluorohexanesulfonic acid (PFHxS),  
154 perfluoroheptanesulfonic acid (PFHpS), branched perfluorooctanesulfonic acid (brPFOS),  
155 linear perfluorooctanesulfonic acid (linPFOS), perfluorononanesulfonic acid (PFNS),  
156 perfluorodecanesulfonic acid (PFDcS), perfluorobutanoic acid (PFBA), perfluoropentanoic  
157 acid (PFPA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA),  
158 perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid  
159 (PFDcA), perfluoroundecanoic acid (PFUnA), perfluorododecanoic acid (PFD<sub>o</sub>A),  
160 perfluorotridecanoic acid (PFTriA), perfluorotetradecanoic acid (PFTeA),  
161 perfluorohexadecanoic acid (PFHxDA) and methyl perfluorohexane sulphonamide  
162 (MeFHxSA). Additionally, PFAS of emerging concern were measured: methylperfluorooctane  
163 sulfonamide (MeFOSA), ethylperfluorooctane sulphonamide (EtFOSA),  
164 perfluorooctanesulfonamidoacetic acid (FOSAA), methyl perfluorooctane sulfonamidoacetic  
165 acid (MeFOSAA), ethyl perfluorooctane sulfonamidoacetic acid (EtFOSAA), 4:2  
166 fluorotelomer sulfonic acid (4:2 FTS), 6:2 fluorotelomer sulfonic acid (6:2 FTS), 8:2  
167 fluorotelomer sulfonic acid (8:2 FTS), 10:2 fluorotelomer sulfonic acid (10:2 FTS), 3:3  
168 fluorotelomer carboxylic acid (3:3 FTCA), 5:3 fluorotelomer carboxylic acid (5:3 FTCA), 7:3  
169 fluorotelomer carboxylic acid (7:3 FTCA), a mixture of 6:2 and 8:2 chlorinated polyfluorinated



170 ether sulfonate (F-53B), dodecafluoro-3H-4,8-dioxanonanoate (ADONA) and  
171 hexafluoropropylene oxide dimer acid (Gen-X), see Supporting Information (SI), Table S1. All  
172 concentrations are expressed in  $\text{ng g}^{-1}$  wet weight (ww).

173

#### 174 **Quality control**

175 To guarantee the quality and control for reproducibility and precision of the PFAS  
176 analyses method, one blank and a standard reference material (human serum AM-S-Y-1908  
177 INSPQ within the Arctic Monitoring and Assessment Program ring test) were concurrently  
178 analyzed every 15 samples. The determined concentrations varied between 78 and 125 % of the  
179 target value. The recovery of the  $^{13}\text{C}$  labelled internal standard was calculated by adding a  
180 recovery standard at the end of the sample processing to every sample. The recoveries of all  
181 internal standard compounds varied between 66 and 120 %. Limit of detection (LOD) was  
182 defined as three times the signal to noise ratio for the specific matrix, or in the case of detection  
183 in the blanks as the sum of the average of the blank level and 3 times standard deviation. LOD  
184 varied depending on the compounds and ranged from 0.004 to 0.500  $\text{ng g}^{-1}$  for the PFAS.

185

#### 186 **Statistical analyses**

187 Among PFAS, only compounds detected in >70 % of both pre-laying females plasma  
188 and eggs yolk were included in analyses to increase statistics robustness. For each included  
189 PFAS, values <LOD were set to half of the LOD of the specific compound.

190 First, we tested for each PFAS the relationship between concentrations in eggs and the  
191 effects of the following biological variables: the laying date, the egg mass and the rank of the  
192 egg in the clutch by laying order (egg number: 1 or 2) as well as both interactions between the  
193 laying date or the egg mass and the egg number, using linear mixed effect models (LMMs: lme  
194 function from “nlme” R-package, v.3.1.147).<sup>56</sup> The females body condition (residuals of the

195 linear correlation between the skull length and the mass) was not included as an explanatory  
196 variable in the model as it was correlated to the laying date (LM:  $t=-4.054$ ,  $p<0.001$ ).

197 Second, LMMs were built to test the maternal transfer efficiency of each individual  
198 PFCA. The maternal transfer ratio, calculated as  
199 *concentration in egg/concentration in female* for each compound, was used as the  
200 response variable and log-transformed to meet the assumptions of linear models. The number  
201 of carbons in the PFCA chain, the egg number as well as the interaction between these two  
202 variables were used as predictors. For these models, all PFAS concentrations lower than LOD  
203 were removed from the dataset to avoid outliers in ratios ( $n=15$ ).

204 Third, we investigated for each PFAS the effect of the concentration in females plasma  
205 and the egg number on the concentration in eggs using LMMs.

206 Each model of the three steps were then ranked and rescaled according to the Akaike's  
207 Information Criterion for small sample size (AICc): in a set of models from the full to the null  
208 model, the model with the lowest AICc was selected for all steps independently. If the  $\Delta AICc$   
209 with the next ranked model was  $<2$ , the most parsimonious was chosen.<sup>57, 58</sup> The nest number  
210 was used as a random intercept in all models. The normality of residuals was verified through  
211 inspection of diagnosis plots (residuals vs fitted values and Q-Q plots). For all models a  
212 significance level of  $\alpha<0.05$  was used. All statistical analyses were performed using R (v.4.0.0;  
213 R Core Team, 2020).<sup>59</sup>

214

## 215 RESULTS AND DISCUSSION

### 216 PFAS concentrations in females and their eggs

217 In the studied population, kittiwakes arrived in the colony in early April after migrating  
218 from pelagic areas of the western North-Atlantic Ocean.<sup>60</sup> During the two-months pre-laying  
219 period that follows migration, females feed mostly on small fish and crustaceans in the marine

220 system near the colony before laying a clutch of usually two eggs.<sup>61-63</sup> Only 3 nests were  
 221 observed to have single egg clutches in the present study. Females having a single egg clutch  
 222 and females having a two eggs clutch had PFAS concentrations of similar magnitude in plasma  
 223 (respectively 8.46-35.4 and 12.4-89.4 ng g<sup>-1</sup> ww). Therefore they were both considered the same  
 224 way in analyses.

225 The following legacy PFAS were detected in >70% of both females plasma and eggs  
 226 yolk and therefore included in analyses: PFHxS, brPFOS, linPFOS, PFOA, PFNA, PFDcA,  
 227 PFUnA, PFDoA, PFTriA, PFTeA (SI Table S1). Among legacy compounds, both PFDcS and  
 228 PFHxDA were above LOD in 84% of the eggs. However, in maternal plasma PFDcS and  
 229 PFHxDA were above LOD in only 24% and 0% of the samples, respectively. All other legacy  
 230 PFAS were not detected in maternal plasma nor in eggs. None of the emerging PFAS were  
 231 detected in the maternal plasma. In contrast, 7:3 FTCA was detected in 84% of the eggs whereas  
 232 ADONA was only detected in a single egg (SI Table S1). None of the other emerging PFAS  
 233 investigated were detected in the yolk. PFDcS found in <70% of the females and PFHxDA, 7:3  
 234 FTCA and ADONA found in none of the females were therefore excluded from statistical  
 235 analyses, but descriptive statistics of the compounds in eggs are provided in SI Table S2.

236

237 **Table 1.** Descriptive statistics (mean  $\pm$  standard deviation *SD*, median and range *min-max*) for  
 238 PFAS concentrations (ng g<sup>-1</sup> ww) in plasma and yolk of black-legged kittiwakes from Svalbard.

	Prelaying females (n=14)			First laid eggs (n=14)			Second laid eggs (n=11)		
	Mean $\pm$ <i>SD</i>	Median	Min-max	Mean $\pm$ <i>SD</i>	Median	Min-max	Mean $\pm$ <i>SD</i>	Median	Min-max
PFHxS	0.23 $\pm$ 0.09	0.21	0.11 - 0.40	0.26 $\pm$ 0.14	0.26	0.08 - 0.57	0.22 $\pm$ 0.09	0.20	0.08 - 0.40
brPFOS	0.75 $\pm$ 0.49	0.86	0.028 - 1.38	3.00 $\pm$ 1.01	3.12	1.44 - 4.45	1.93 $\pm$ 0.77	1.93	0.88 - 3.10
linPFOS	10.8 $\pm$ 4.70	11.3	3.14 - 17.42	28.6 $\pm$ 10.3	25.9	17.0 - 50.7	21.3 $\pm$ 7.90	19.6	11.8 - 34.7
PFOA	0.17 $\pm$ 0.13	0.14	0.04 - 0.57	0.23 $\pm$ 0.12	0.22	0.04 - 0.47	0.21 $\pm$ 0.08	0.19	0.13 - 0.41
PFNA	1.06 $\pm$ 0.61	1.07	0.19 - 2.44	1.98 $\pm$ 1.07	1.75	0.85 - 3.64	1.74 $\pm$ 0.80	1.48	0.71 - 2.98
PFDcA	1.71 $\pm$ 1.56	1.56	0.40 - 3.26	3.63 $\pm$ 1.88	3.07	1.73 - 6.83	3.07 $\pm$ 1.38	2.60	1.19 - 5.20
PFUnA	6.88 $\pm$ 3.22	7.42	1.40 - 12.4	17.9 $\pm$ 5.78	16.8	10.5 - 28.6	14.0 $\pm$ 4.55	13.4	7.33 - 20.8
PFDoA	1.78 $\pm$ 0.98	1.78	0.02 - 3.76	6.07 $\pm$ 1.46	5.90	4.36 - 9.41	4.40 $\pm$ 1.19	4.55	2.84 - 6.62

PFTriA	7.80 ± 3.35	7.93	1.81 - 13.9	33.5 ± 6.45	33.8	22.5 - 50.8	23.0 ± 5.65	22.3	14.7 - 35.2
PFTeA	1.44 ± 0.60	1.53	0.471 - 2.63	6.74 ± 1.55	7.07	4.36 - 10.1	4.60 ± 1.38	4.71	2.72 - 7.00
$\Sigma$ PFSA	11.77 ± 5.03	12.5	3.37 - 18.9	31.9 ± 11.1	28.4	19.5 - 55.2	23.4 ± 8.48	22.1	12.9 - 37.8
$\Sigma$ PFCAs	20.73 ± 8.85	21.8	5.09 - 36.5	70.1 ± 15.4	67.5	48.0 - 109	51.2 ± 13.3	48.2	33.8 - 78.4
$\Sigma$ PFASs	32.5 ± 13.5	33.5	8.46 - 51.9	102 ± 25.9	98.2	68.5 - 164	74.5 ± 21.4	71.1	46.6 - 116

239

240 The most abundant PFAS found in maternal plasma were linPFOS (mean ± SD: 10.8 ±  
241 4.70 ng g<sup>-1</sup> ww), followed by PFTriA (7.80 ± 3.35 ng g<sup>-1</sup> ww) and PFUnA (6.88 ± 3.22 ng g<sup>-1</sup>  
242 ww, Table 1). Together, these three PFAS represented 78.6% of the  $\Sigma$ PFAS. In both eggs, PFTriA  
243 was dominant (28.9 ± 7.99 ng g<sup>-1</sup> ww), followed by linPFOS (25.4 ± 9.89 ng g<sup>-1</sup> ww) and  
244 PFUnA (16.2 ± 5.54 ng g<sup>-1</sup> ww), representing 78.4% of the  $\Sigma$ PFAS. For example, incubating  
245 glaucous gulls (*Larus hyperboreus*) sampled in the same area showed dominating linPFOS,  
246 then PFUnA and PFTriA in both plasma and eggs.<sup>38, 64</sup> Odd-chain PFCAs (e.g. C<sub>9</sub>, C<sub>11</sub>, C<sub>13</sub>)  
247 being more abundant than even-numbered chain is a frequent pattern in seabirds and their eggs,  
248 this is believed to be linked to the degradation process of precursor compounds such as  
249 fluorotelomer alcohols (FTOH), as well as a selective bioaccumulation.<sup>21, 27, 44, 65-67</sup>

250 PFCAs in plasma reflect recent dietary uptake due to their efficient elimination from  
251 birds' bodies.<sup>68</sup> In contrast, PFOS and other PFSA show much longer half-lives in plasma,  
252 suggesting that blood acts as a significant reservoir for them. Thus, occurrence of PFCAs may  
253 represent more recent and local inputs (marine foraging area close to Svalbard), whereas PFSA  
254 would rather represent contamination before birds enter the Svalbard areas to breed (winter and  
255 early spring in the West Atlantic).<sup>69</sup> Moreover, plasma samples may represent a snapshot of  
256 recent exposure to PFAS while egg concentrations integrate a longer period of time of egg  
257 formation, rather reflecting recent days and weeks.<sup>23</sup> In some cases even revealing the exposure  
258 at the wintering grounds, hampering direct comparisons of PFAS patterns in plasma and eggs.  
259 Kittiwakes in Svalbard nonetheless arrive on the breeding grounds by mid-April and females  
260 of the present study were caught on the 4<sup>th</sup> of June ± 8 days (mean ± SD).<sup>60</sup> Before they laid,

261 they were thus feeding locally for a longer time than the length of most PFAS clearance time  
262 measured in eggs of PFAS fed hens (*Gallus gallus*).<sup>23,70</sup> Despite different metabolisms and egg-  
263 laying pattern (and thus excretion) between these two species, PFAS concentrations of  
264 kittiwakes plasma and egg yolks could therefore mainly represent local contamination.

265

### 266 **Emerging PFAS in females and their eggs**

267 A broad number of shorter-chain alternatives, supposedly less bioaccumulative and  
268 toxic than long-chain persistent PFAS, have been synthesized since the 2000's when C<sub>8</sub> and  
269 related PFAS were phased out or regulated in North America and in Europe.<sup>71,72</sup> Even so, some  
270 of them have been found in biota, sometimes with higher levels of bioaccumulation and toxicity  
271 than the long-chain PFAS they replaced.<sup>45,48,73-79</sup> In the present study, we measured 7:3 FTCA  
272 above LOD in 84% of the eggs (mean  $\pm$  SD: 0.21  $\pm$  0.09 ng g<sup>-1</sup> ww; range: 0.10 – 0.40 ng g<sup>-1</sup>  
273 ww). To the best of our knowledge, this is the first report of this compound in seabird eggs, and  
274 the second for Arctic top predators.<sup>46</sup> 7:3 FTCA is an intermediate environmental degradation  
275 product from fluorotelomer alcohols (FTOH), whose final degradation products are PFCAs.<sup>80</sup>  
276 It has been recently detected in human tissues and wildlife.<sup>46,81-87</sup> In eggs of osprey (*Pandion*  
277 *haliaetus*), tawny owl (*Strix aluco*) and common kestrel (*Falco tinnunculus*), this compound  
278 ranged from <0.24 (LOD) to 2.7 ng g<sup>-1</sup> much higher concentrations than those found in  
279 kittiwake eggs reported here.<sup>78</sup> Literature regarding fluorotelomer toxicity is scarce and even  
280 non-existent for avian species, but some were found to be potentially more toxic than long-  
281 chain PFAS.<sup>88-90</sup> Among emerging PFAS, ADONA was detected in a single egg at a  
282 concentration of 0.11 ng g<sup>-1</sup> ww, which is the first documented finding for this compound in  
283 wildlife (chromatograms for ADONA measurements are provided in SI Figure S1). To the best  
284 of our knowledge, this compound was detected only twice before in biota, in blood of humans  
285 living close to a production plant using ADONA in Germany and in human milk from women

286 in China.<sup>91, 92</sup> Therefore we recommend additional investigations for this compound in bird  
287 eggs. Other fluorinated alternatives such as F-53B and Gen-X were not detected in any of the  
288 samples, F-53B was very recently discovered in herring gull eggs from Germany (despite not  
289 being officially used in Europe) and, along with Gen-X, in 100% of black-tailed gull (*Larus*  
290 *crassirostris*) eggs from South Korea with an increasing trend over time.<sup>50, 51</sup> None of the  
291 analyzed emerging PFAS were found in females plasma in the present study, which means that  
292 kittiwakes foraging grounds are not important sources. However, PFAS accumulation in a  
293 specific tissue dependent on their physicochemical characteristics.<sup>21</sup> Thus, although we detected  
294 7:3 FTCA and ADONA in eggs yolk and not in maternal plasma, these compounds may be  
295 present in other organs of the birds. Another hypothesis for the presence of emerging PFAS in  
296 yolk but not in females' plasma could be a very high transfer efficiency of these compounds  
297 since some emerging PFAS (including ADONA) have been found to bind to human and rats  
298 liver proteins at least as strongly as PFOA or PFOS.<sup>93</sup> A significant transfer of precursors such  
299 as FTOH in eggs could also be a reason for the detection of 7:3 FTCA in eggs but not in females,  
300 however this is highly speculative as these compounds were not measured in the present study.  
301 Adults blood plasma is frequently used in birds to describe the extent of local contamination to  
302 a wide range of pollutants.<sup>94</sup> However, in our study this tissue shows its limitation as a tool to  
303 document the contamination of the environment, as all emerging compounds investigated and  
304 detected in egg yolks could not be detected in any of the plasma samples. For future studies,  
305 we suggest systematic screening for emerging PFAS in biota to increase the overview of the  
306 exposure to these compounds and we suggest that the use of eggs would be more pertinent  
307 rather than adults plasma. Toxicological studies are also strongly needed to evaluate the threats  
308 of the alternative fluorinated compounds in biota.

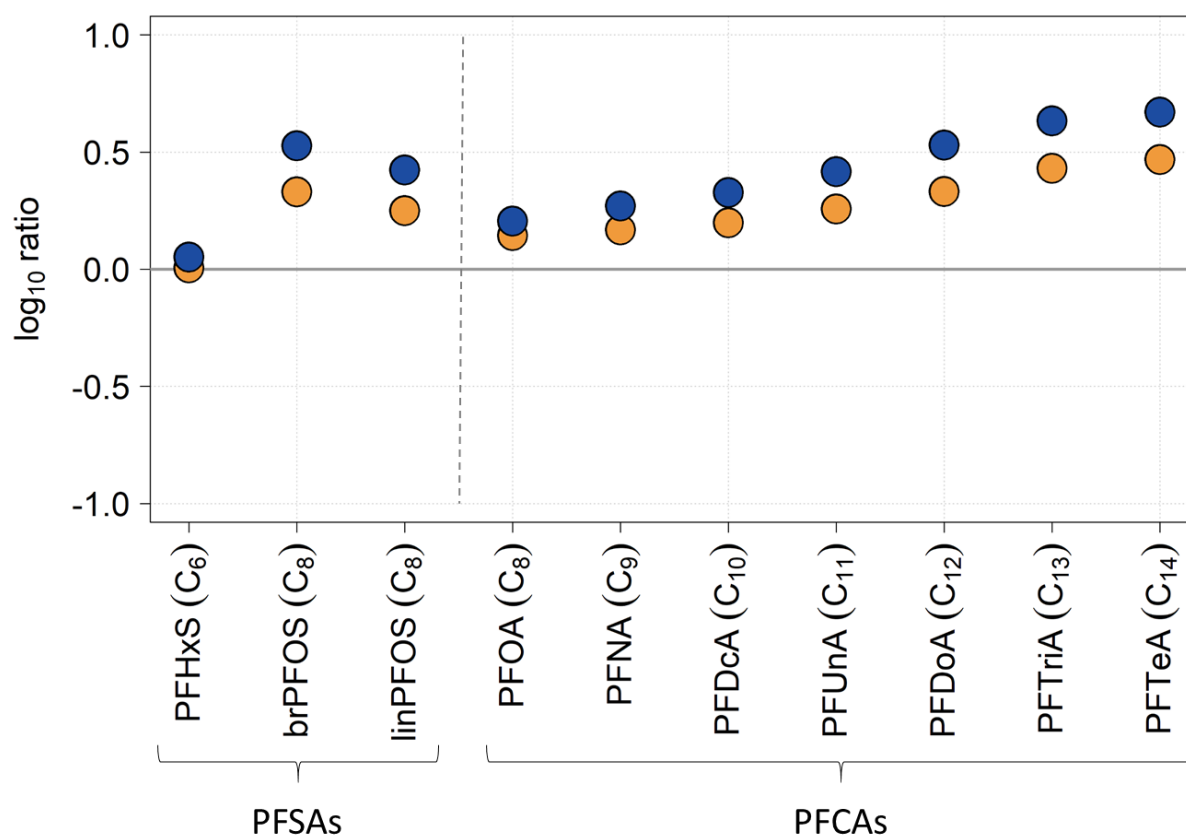
309

### 310 **Relationship between biological factors and PFAS concentrations in eggs**

311 The variables “laying date” and “egg mass” were excluded from all best models  
312 explaining PFAS variations in eggs (SI Table S3). Therefore, developing embryos from eggs  
313 laid early or late in the season should have a similar exposure to PFAS. Moreover, juvenile  
314 survival is known to be affected by egg size (and therefore egg mass) in various avian species,<sup>95</sup>  
315 however egg mass was unrelated to PFAS contamination in our study. Relationship between  
316 laying date and PFAS concentrations in eggs is not commonly studied, but in great tits (*Parus*  
317 *major*) a negative and linear correlation was found between PFOS concentration of eggs and  
318 the laying date but no relationship with any of the other PFAS was found.<sup>96</sup> In the present study,  
319 the absence of relationship between the laying date and any of the PFAS concentrations in eggs  
320 might be due to the highly synchronized and time-limited laying period during which significant  
321 variations of PFAS concentrations in Svalbard water would be unlikely. In the same study on  
322 great tits, a positive and linear relationship was also noted between PFOS concentration and the  
323 egg mass, which was presumably attributed to the lipoprotein content of the eggs, heavier eggs  
324 having a higher lipoprotein content, yet the result of the present study do not support this  
325 hypothesis. All selected PFAS, but PFHxS and PFOA (null model selected), were in higher  
326 concentration in first- than in second-laid eggs (all  $t < -2.57$ , all  $p < 0.03$ ; Figure 2; SI Table S3).  
327 This difference in PFAS concentrations between eggs of the same clutch is generally observed  
328 in gulls, but it is not always the case in other birds with larger clutches.<sup>29, 52, 96</sup> As a consequence,  
329 toxicological impairments on hatchability and survival would be more frequent for the first-laid  
330 egg, which could be critical for small clutch size species, including kittiwake. Decreasing body  
331 contaminant burden along the laying sequence is the main hypothesis to explain the decreasing  
332 concentrations in eggs. In species with larger clutch sizes, daily exogenous intakes or food  
333 shortage during laying could be of higher importance, leading to variations in blood circulating  
334 PFAS and thus in eggs along the laying sequence. In kittiwakes, the similar concentrations  
335 observed in both eggs for PFHxS and PFOA might be the consequence of low concentrations

336 of these two compounds in maternal plasma, resulting in a lower transfer efficiency to eggs.

337



338 **Figure 1.** Maternal transfer ratios ( $\log_{10}$  ratios) of PFAS in black-legged kittiwakes from

339 Svalbard. The PFASs and PFCAs are ordered by carbon chain length (given in parentheses).

340 Ratios are calculated as

341  $\log_{10}(\text{mean concentration in eggs}/\text{mean concentration in plasma})$  for first- and

342 second-laid eggs. Blue and orange colors are used for first- and second-laid eggs respectively.

343 The grey horizontal solid line ( $\log_{10}$  ratio = 0) stand for a 1:1 relationship between female and

344 eggs.

345

### 346 PFAS transfer ratios between females and their eggs

347 Maternal transfer ratios (MTRs) were all higher than 0, which means that eggs are an

348 important excretion route for all measured PFAS compounds in kittiwakes (Figure 1). The

349 model selection showed that both the length of the PFAS carbon chain, the egg number and



350 their interaction were significant predictors of the MTRs for PFCAs (SI Table S4). MTRs were  
351 increasing with the carbon chain length both for first- and second-laid eggs ( $t=12.5$ ,  $p<0.001$   
352 and  $t=6.87$ ,  $p<0.001$  respectively; Figure 1). This suggests a preferential transfer of PFCAs with  
353 the longest chains. Similar findings that those of the present study were documented for PFCAs  
354 maternal transfer measured between females liver and eggs yolk in herring (*Larus argentatus*)  
355 and common guillemots (*Uria aalge*) respectively from North America and the Baltic Sea.<sup>21, 27</sup>  
356 This pattern may originate from a selective binding of the longest chain length PFCAs to  
357 proteins and a subsequent facilitated transfer to the egg. Such a selective binding have been  
358 revealed in *in-vitro* studies with a variable affinity of PFAS to bovine serum albumin depending  
359 on the carbon-chain length.<sup>97</sup> Moreover, long-chain PFAS generally have greater  
360 bioaccumulation potential than short-chain homologues, they are thus more likely to be  
361 transferred to the eggs.<sup>6</sup> However, MTRs of PFCAs had no clear trends with the compounds  
362 chain length in herring gulls of southern Norway.<sup>19</sup> A linear relationship between MTRs and  
363 PFCAs chain length depict the absence of compounds-specific transfer mechanisms. However,  
364 a leveling off of the curve on the extremes (see Figure 1) could be the sign of additional transfer  
365 mechanisms for PFOA (C<sub>8</sub>) and a transfer kinetic barrier for PFTeA (C<sub>14</sub>).

366 The extent of the MTRs increase with chain length was different between first- and  
367 second-laid eggs ( $t=-3.25$ ,  $p<0.01$ ; Figure 1), leading to an increasing difference in the MTRs  
368 with PFCAs carbon chain length between both eggs. Consequently, second-laid eggs received  
369 a lower proportion of females plasma contaminants than first-laid eggs for the compounds with  
370 the longest chains. Such a difference could be a consequence of the preferential transfer of the  
371 longest chain PFCAs, an increasing transfer of PFCAs with the chain length would lead to a  
372 higher depletion of these compounds in females and therefore a lower transfer in their second-  
373 laid eggs.

374 Facilitated transfer of PFCAs with the longest carbon chains is concerning as it could

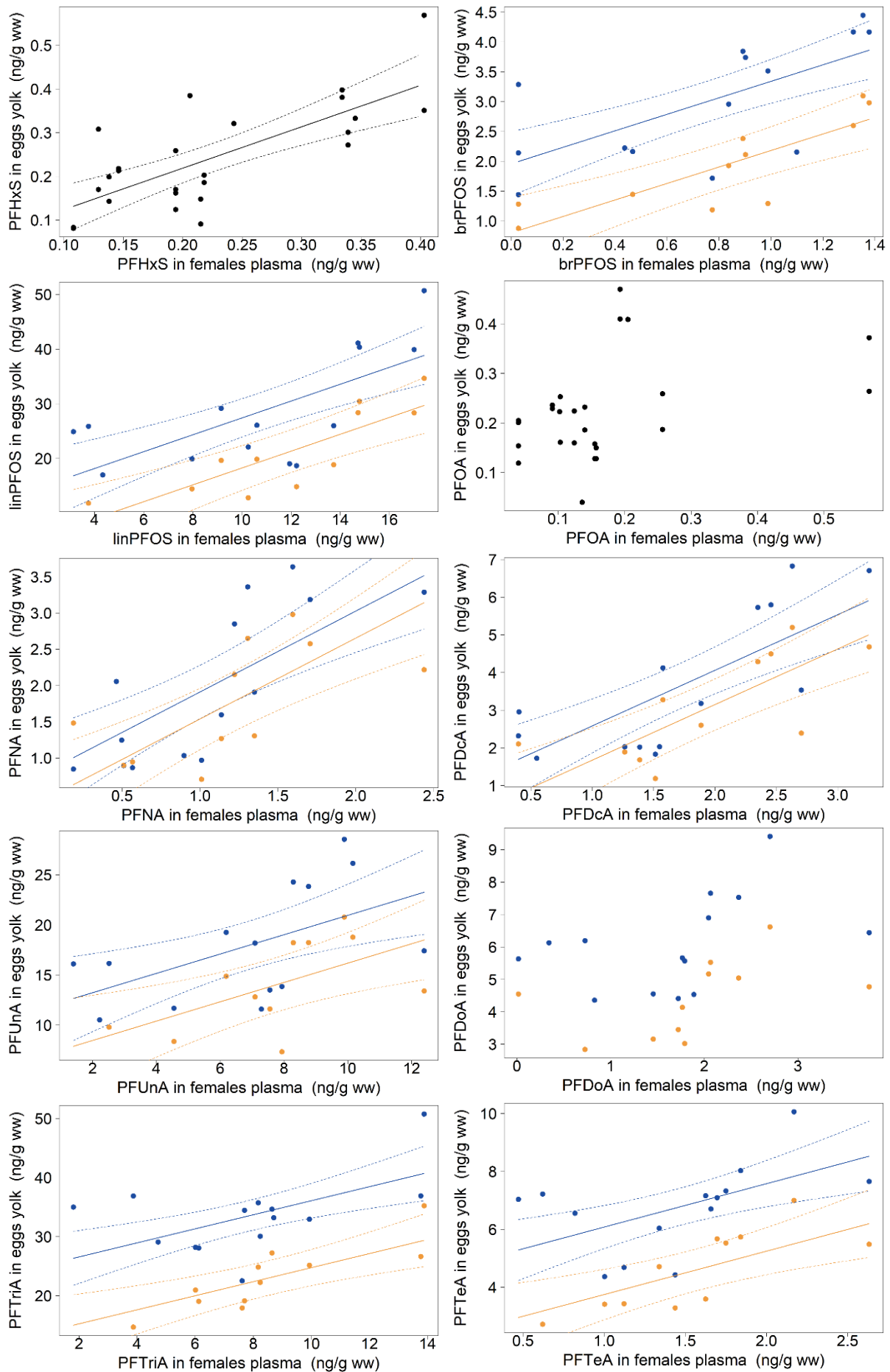
375 potentially induce adverse effects for the development of the embryo, as toxicity is known to  
376 be dependent on the carbon chain length.<sup>41, 98, 99</sup> However, the majority of the studies on the  
377 effects of PFAS exposure on the embryo focused on PFOS and PFOA. These studies revealed  
378 that PFOS and PFOA can impair hatchability and disrupt embryos' physiology, but at higher  
379 concentrations than those measured in the present study.<sup>31, 32, 34-36, 100-102</sup> Studies on the  
380 consequences of long-chain PFCAs *in ovo* exposure on birds' embryo are needed, especially as  
381 long chain PFCAs are widely detected in wildlife, despite decreasing concentrations in the last  
382 few years.<sup>65, 103-107</sup>

383         Among PFASs, PFHxS MTR was lower than those of brPFOS and linPFOS (LMM:  $t=-$   
384 10.4,  $p<0.001$  and  $t=-8.56$ ,  $p<0.001$  respectively), suggesting, as for PFCAs, an increasing  
385 transfer with carbon chain length. PFOS and C<sub>11</sub>-C<sub>14</sub> PFCAs have also been found to be more  
386 lipophilic than other short-chain PFAS, representing a large proportion of the  $\sum_{\text{PFAS}}$  in the fat  
387 of marine mammals.<sup>108</sup> Seabirds egg yolk being constituted of 20 to 30% lipids,<sup>109</sup> the maternal  
388 transfer of PFAS could be both lipid- and protein-driven. Concerning PFOS, brPFOS was  
389 significantly more transferred to the eggs than linPFOS (LMM:  $t=-2.50$ ,  $p=0.02$ ) despite a  
390 similar carbon chain length for these isomers. This may be a consequence of different transfer  
391 mechanisms for the branched and linear forms. However, contrarily to linPFOS, brPFOS is a  
392 mixture of different isomers and not all of them are covered by the labeled standards used for  
393 analyses, potentially leading to lower analytical precision in the measurements of brPFOS  
394 concentrations.

395         PFAS are bound to proteins (including serum albumin, fatty acid binding proteins and  
396 organic anion transporters), which affects their distribution, bioaccumulation and excretion.<sup>110</sup>  
397 In marine mammals, the trophic magnification factors of individual PFAS were found to have  
398 a significant relationship with their protein-water partition coefficients ( $\text{LogK}_{\text{PW}}$ ).<sup>111</sup> Therefore,  
399 PFASs transfer is affected by their binding affinity to proteins. In hooded seals (*Cystophora*

400 *cristata*) and humans, the transfer efficiency of PFCAs presented a U-shape with a decreasing  
401 transfer proficiency from C<sub>8</sub> to C<sub>10</sub> and an increasing from C<sub>10</sub> to C<sub>13</sub> and that of PFASs was  
402 decreasing with compounds chain length.<sup>112-114</sup> The relationship between MTRs and PFASs  
403 chain length described in the present study is different. However, this U-shape trend in  
404 mammals has been described as the integrated result of opposite trends in the transfer from  
405 maternal blood to placenta and from placenta to cord blood.<sup>113</sup> Similar trends than those of the  
406 present study were found in the transfer of PFASs and PFCAs from maternal blood to placenta,  
407 this has been described as the consequence of a high water content in blood and a higher protein  
408 and lipid content in the placenta. As a consequence, short chain PFASs with greater water  
409 solubility and less capacity to bind with proteins were less transferred to placenta. Similar  
410 transfer mechanisms between females blood and eggs yolk could lead to the MTRs pattern  
411 observed in kittiwakes.

412         Investigating lipid and protein content of female plasma and egg yolk in future studies  
413 would help in understanding the mechanisms behind maternal transfer of PFAS. Sampling  
414 females before and after egg-laying as well as males, would also be interesting in order to  
415 evaluate if females are depleted in the longest chain PFCAs after laying and compared to males.



417 **Figure 2.** PFAS concentrations (ng g<sup>-1</sup> ww) relationship between females plasma and yolks of  
 418 first- and second-laid eggs of black-legged kittiwakes from Svalbard. The solid line refers to a  
 419 statistically significant linear maternal transfer (see Table 2), with dotted lines representing 95%  
 420 confidence intervals and no line representing no significant relationship. Blue and orange colors  
 421 are used for first- and second-laid eggs respectively when concentrations are significantly  
 422 different between them, black is used if the trends are similar between egg numbers.

423

424 **Table 2.** Factors affecting each PFAS concentrations in black-legged kittiwakes' eggs from  
 425 Svalbard, estimated by mixed linear regression models. Significant *p*-values are bolded.

Parameter	Estimate	SE	<i>t</i> -value	<i>p</i> -value
<b><u>PFHxS (R<sup>2</sup>m: 0.52; R<sup>2</sup>C: 0.72)</u></b>				
Female plasma	0.94	0.22	4.35	<b>&lt;0.001</b>
<b><u>brPFOS (R<sup>2</sup>m: 0.59; R<sup>2</sup>C: 0.84)</u></b>				
Female plasma	1.25	0.36	3.51	<b>&lt;0.01</b>
Egg number (2)	-1.18	0.18	-6.68	<b>&lt;0.001</b>
<b><u>linPFOS (R<sup>2</sup>m: 0.55; R<sup>2</sup>C: 0.93)</u></b>				
Female plasma	1.45	0.39	3.74	<b>&lt;0.01</b>
Egg number (2)	-9.52	1.14	-8.34	<b>&lt;0.001</b>
<b><u>PFOA (Null model)</u></b>				
<b><u>PFNA (R<sup>2</sup>m: 0.48; R<sup>2</sup>C: 0.88)</u></b>				
Female plasma	1.10	0.30	3.64	<b>&lt;0.01</b>
Egg number (2)	-0.38	0.14	-2.68	<b>0.023</b>
<b><u>PFDCa (R<sup>2</sup>m: 0.54; R<sup>2</sup>C: 0.93)</u></b>				
Female plasma	1.43	0.35	4.13	<b>&lt;0.01</b>
Egg number (2)	-0.93	0.19	-5.02	<b>&lt;0.001</b>
<b><u>PFUnA (R<sup>2</sup>m: 0.38; R<sup>2</sup>C: 0.95)</u></b>				
Female plasma	0.96	0.39	2.42	<b>0.032</b>
Egg number (2)	-5.29	0.54	-9.86	<b>&lt;0.001</b>
<b><u>PFDoA (R<sup>2</sup>m: 0.34; R<sup>2</sup>C: 0.90)</u></b>				
Egg number (2)	-1.92	0.23	-8.52	<b>&lt;0.001</b>

**PFTriA (R<sup>2</sup>m: 0.61; R<sup>2</sup>C: 0.79)**

Female plasma	1.09	0.37	2.98	<b>0.012</b>
Egg number (2)	-1.10	1.50	-7.36	<b>&lt;0.001</b>

**PFTeA (R<sup>2</sup>m: 0.53; R<sup>2</sup>C: 0.79)**

Female plasma	1.34	0.52	2.59	<b>0.024</b>
Egg number (2)	-2.21	0.34	-6.60	<b>&lt;0.001</b>

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426

427 **Relationship between PFAS concentrations in females and their eggs**

428 In this study, eggs laid by the sampled females were collected, enabling the study of the  
429 actual profile of the maternal transfer of PFAS. Among PFAS, a positive and linear association  
430 between yolks and female plasma was observed for the following compounds: PFHxS, brPFOS,  
431 linPFOS, PFNA, PFDcA, PFUnA, PFTriA and PFTeA (all  $t > 2.42$ , all  $p < 0.032$ , SI Table S5,  
432 Table 2 and Figure 2). A similar linear relationship between females and eggs was documented  
433 in tree swallows (*Tachycineta bicolor*) for PFOS, in great tits for PFOS and PFOA, as well as  
434 for PFHxS, PFOS and PFOA in hens.<sup>20, 23, 32</sup> PFAS have been found to have relatively strong  
435 binding abilities to yolk proteins (low-density lipoprotein, high-density lipoprotein and vitellin  
436 proteins).<sup>115</sup> All yolk proteins (except immunoglobulins) being synthesized in the liver, it makes  
437 it the entry point for PFAS in egg yolks.<sup>116</sup> A linear relationship between females and eggs for  
438 most of the PFAS as found in the present study suggests that the efficiency of PFAS transfer  
439 mechanisms are not dependent on the concentration in females' plasma. These linear  
440 relationships observed between females' plasma and eggs yolk therefore cannot explain the  
441 differences found in the PFAS patterns presented above. The affinity of the different PFAS for  
442 proteins varies to a large extent, suggesting binding site-specific interactions and facilitated  
443 transport of some compounds.

444 However, PFAS are not always linearly transferred as found between females and eggs  
445 for PFHxA in hens and PFCAs (except PFOA) in great tits.<sup>20, 23</sup> A comparable absence of  
446 relationship was found for PFOA and PFDoA in the present study. At low levels, the transfer

447 dynamic of contaminants could be different. This could have been the case for PFOA, for which  
448 we found the lowest concentrations in both the females and the eggs. However, this was not the  
449 case for PFHxS which was also in very low concentrations in both females and eggs. Additional  
450 studies are needed to reveal whether the non-linear transfer of PFOA and PFDoA stems from  
451 real biological mechanisms or is due to low statistical resolution caused by the low sample size.  
452 The linear relationship between both eggs and females for most PFAS validates the use of eggs  
453 to assess the female's exposure in kittiwakes, however, we emphasized the relative importance  
454 of sampling eggs of similar laying numbers to avoid biased estimation due to the decrease in  
455 contamination with the laying-sequence.

456         To conclude, there was no measurable effects of the maternal plasma concentrations of  
457 PFAS on the efficiency of the transfer mechanisms into the yolk. However, the physicochemical  
458 characteristics of the different compounds (carbon chain length in particular) seem to affect the  
459 transfer, with a likely facilitated transportation of the longest chain PFCAs into the yolk. Effects  
460 of these long chain compounds on the development of seabird embryos are thus crucial to  
461 investigate as development represents a critical period to contaminants exposure. As highly  
462 contaminated females lay highly contaminated eggs, kittiwake embryos might be at strong risk  
463 as they are impacted twice by the female exposure to contaminants: 1/ via impaired incubation  
464 and chick-rearing behaviors and 2/ via maternal transfer to the egg with impaired embryonic  
465 and early days physiology and development. Therefore, for further studies, estimating the  
466 outcomes of the maternal transfer of PFAS on avian development physiology is essential to  
467 assess if this leads nestlings to a bad start in life, especially for emerging fluorinated compounds  
468 which are under-represented in experimental studies.

469

470 ASSOCIATED CONTENT

471 **Supporting Information**

472 The following files are available free of charge.  
473 Internal standards used in PFAS analyses; List of targeted PFAS; descriptive statistics in eggs  
474 for PFDcS, PFHxDA and 7:3 FTCA; chromatograms of the measurement of ADONA in one  
475 egg; model selection for the relationship between PFAS and biological variables in eggs; model  
476 selection for maternal transfer ratios; model selections for the maternal transfer of PFAS in eggs  
477 (PDF).

478

## 479 AUTHORS INFORMATION

### 480 **Corresponding Authors**

481 \*E-mail: [w.jouanneau@gmail.com](mailto:w.jouanneau@gmail.com)

482

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