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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

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18

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_{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.¹ 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.²⁻⁵

56 Some of the PFAS bioaccumulate in organisms and biomagnify along trophic chains,
57 particularly in aquatic ecosystems.^{6, 7} As a consequence, high trophic level aquatic organisms
58 including seabirds often have elevated tissue concentrations.^{1, 8} Oceanic and atmospheric
59 transport can contribute to the long-range dispersal of PFAS, their precursors and breakdown
60 products⁹⁻¹⁵ which are found in wildlife tissues worldwide, including remote areas like the
61 Arctic.^{8, 16}

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).¹⁷ 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.¹⁸ 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.¹⁷ 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.¹⁹⁻²³ Although fluorinated compounds are transferred at a lower degree
78 than lipophilic contaminants, they can still be found at high concentrations in eggs.^{19, 24-26} 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.²⁷⁻²⁹

83 In humans and in laboratory models, PFAS can cause cancer, affect immuno-
84 competence and disrupt the endocrine system.³⁰ 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.³¹⁻³⁷ 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.³⁸⁻⁴³ PFAS cover thousands of substances, the vast
91 majority are not regulated yet and some are increasingly detected in biota.⁴⁴ 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.^{45, 46} 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.⁴⁷ 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.⁴⁸ To the best of our knowledge, to date only
99 three studies detected emerging PFAS in avian eggs.⁴⁹⁻⁵¹

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 exposure⁴³; 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.⁵² 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).⁵³ 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.^{54, 55} 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.^{21, 29} 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 μL^{-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_oA),
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 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}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 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).⁵⁶ 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 PFAs. 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 PFAs 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.^{57, 58} 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).⁵⁹

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.⁶⁰ 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.⁶¹⁻⁶³ 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⁻¹ 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⁻¹ 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
Σ 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
Σ 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
Σ 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⁻¹ ww), followed by PFTriA (7.80 ± 3.35 ng g⁻¹ ww) and PFUnA (6.88 ± 3.22 ng g⁻¹
242 ww, Table 1). Together, these three PFAS represented 78.6% of the Σ PFAS. In both eggs, PFTriA
243 was dominant (28.9 ± 7.99 ng g⁻¹ ww), followed by linPFOS (25.4 ± 9.89 ng g⁻¹ ww) and
244 PFUnA (16.2 ± 5.54 ng g⁻¹ ww), representing 78.4% of the Σ 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.^{38, 64} Odd-chain PFCAs (e.g. C₉, C₁₁, C₁₃)
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.^{21, 27, 44, 65-67}

250 PFCAs in plasma reflect recent dietary uptake due to their efficient elimination from
251 birds' bodies.⁶⁸ 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).⁶⁹ 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.²³ 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 4th of June ± 8 days (mean ± SD).⁶⁰ 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*).^{23,70} 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 C₈ and
269 related PFAS were phased out or regulated in North America and in Europe.^{71,72} 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.^{45,48,73-79} 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⁻¹ ww; range: 0.10 – 0.40 ng g⁻¹
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.⁴⁶ 7:3 FTCA is an intermediate environmental degradation
275 product from fluorotelomer alcohols (FTOH), whose final degradation products are PFCAs.⁸⁰
276 It has been recently detected in human tissues and wildlife.^{46,81-87} 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⁻¹ much higher concentrations than those found in
279 kittiwake eggs reported here.⁷⁸ 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.⁸⁸⁻⁹⁰ Among emerging PFAS, ADONA was detected in a single egg at a
282 concentration of 0.11 ng g⁻¹ 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.^{91, 92} 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.^{50, 51} 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.²¹ 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.⁹³ 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.⁹⁴ 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.

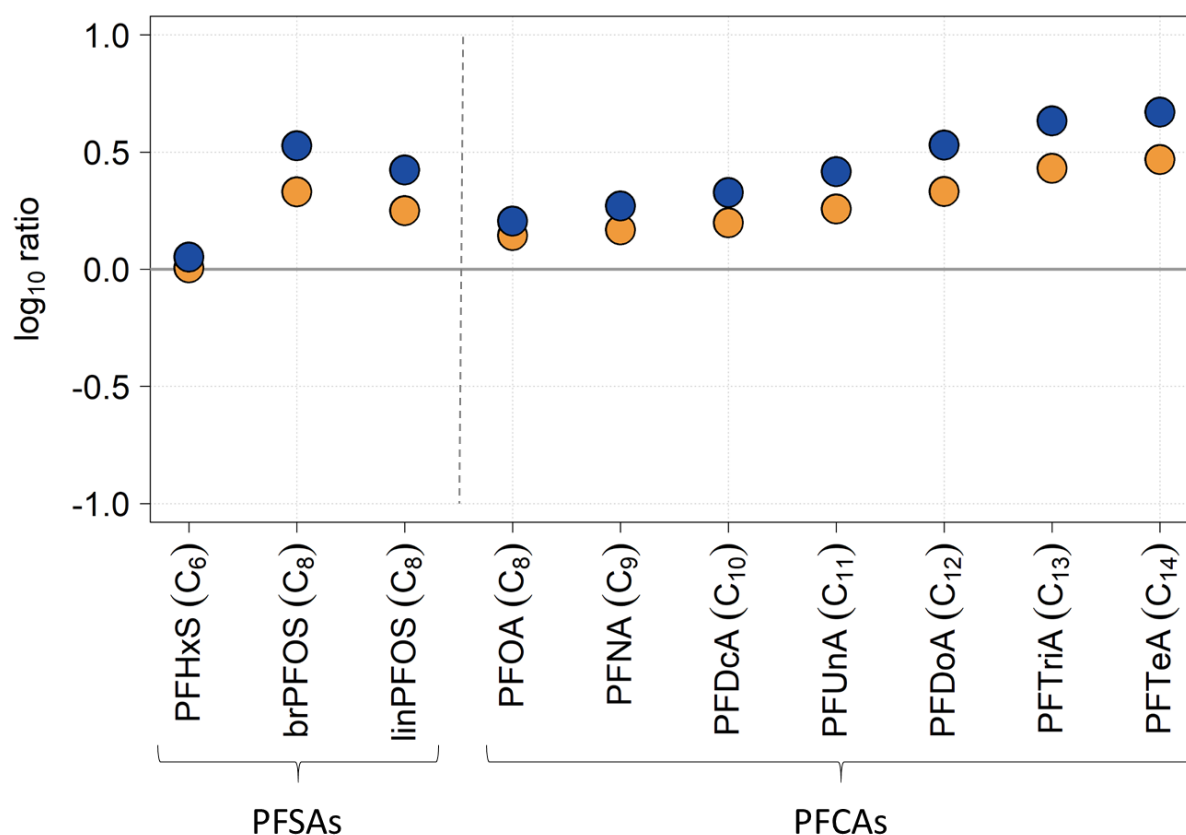
309

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,⁹⁵
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.⁹⁶ 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.^{29, 52, 96} 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.^{21, 27}
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.⁹⁷ 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.⁶ However, MTRs of PFCAs had no clear trends with the compounds
362 chain length in herring gulls of southern Norway.¹⁹ 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 (C₈) and a transfer kinetic barrier for PFTeA (C₁₄).

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

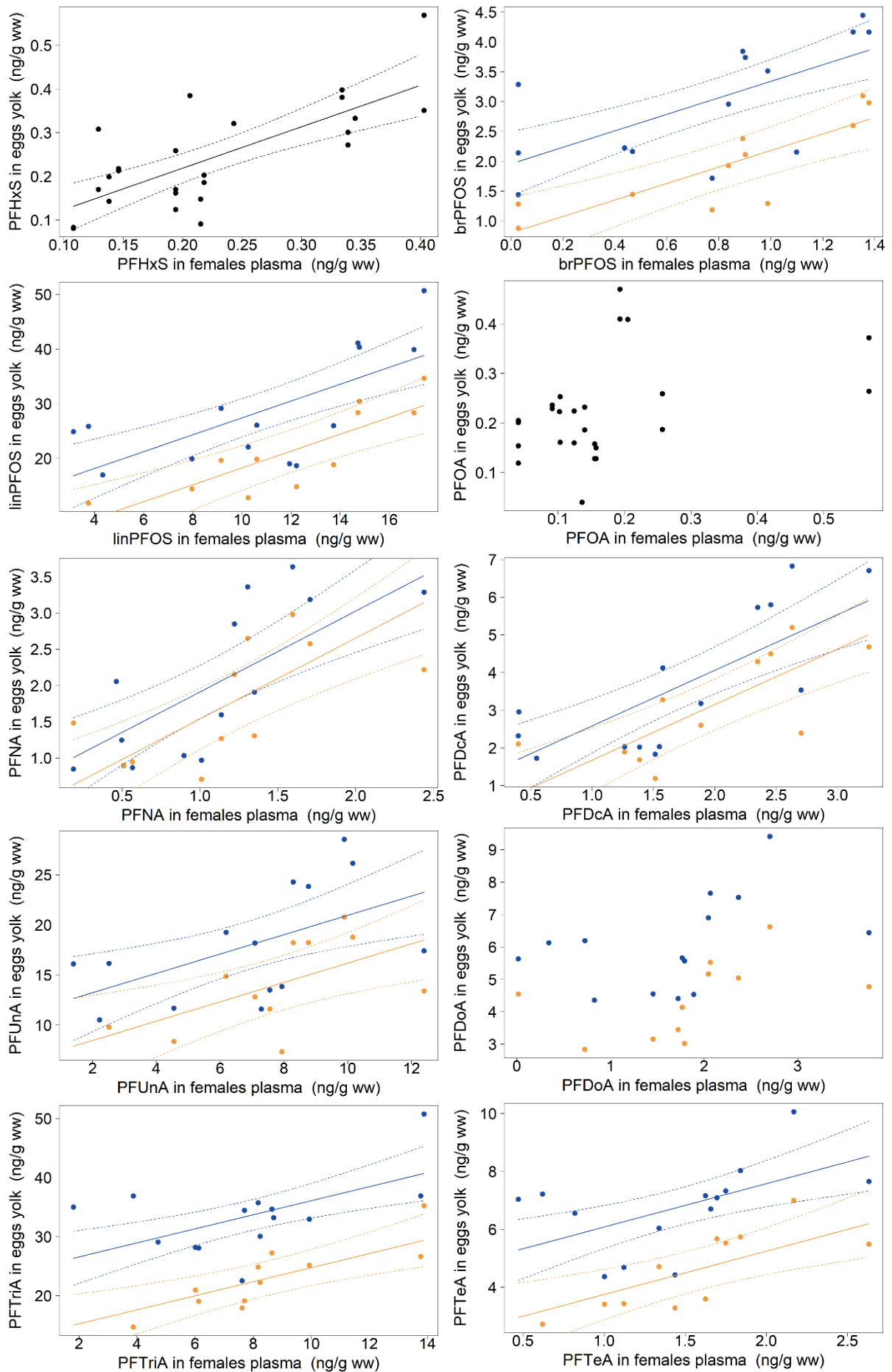
375 potentially induce adverse effects for the development of the embryo, as toxicity is known to
376 be dependent on the carbon chain length.^{41, 98, 99} However, the majority of the studies on the
377 effects of PFAS exposure on the embryo focused on PFOS and PFOA. These studies revealed
378 that PFOS and PFOA can impair hatchability and disrupt embryos' physiology, but at higher
379 concentrations than those measured in the present study.^{31, 32, 34-36, 100-102} 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.^{65, 103-107}

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 C₁₁-C₁₄ PFCAs have also been found to be more
386 lipophilic than other short-chain PFAS, representing a large proportion of the \sum_{PFAS} in the fat
387 of marine mammals.¹⁰⁸ Seabirds egg yolk being constituted of 20 to 30% lipids,¹⁰⁹ 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.¹¹⁰
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 (LogK_{PW}).¹¹¹ 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 C₈ to C₁₀ and an increasing from C₁₀ to C₁₃ and that of PFASs was
402 decreasing with compounds chain length.¹¹²⁻¹¹⁴ 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.¹¹³ 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^{-1} 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	t -value	p -value
<u>PFHxS (R²m: 0.52; R²C: 0.72)</u>				
Female plasma	0.94	0.22	4.35	<0.001
<u>brPFOS (R²m: 0.59; R²C: 0.84)</u>				
Female plasma	1.25	0.36	3.51	<0.01
Egg number (2)	-1.18	0.18	-6.68	<0.001
<u>linPFOS (R²m: 0.55; R²C: 0.93)</u>				
Female plasma	1.45	0.39	3.74	<0.01
Egg number (2)	-9.52	1.14	-8.34	<0.001
<u>PFOA (Null model)</u>				
<u>PFNA (R²m: 0.48; R²C: 0.88)</u>				
Female plasma	1.10	0.30	3.64	<0.01
Egg number (2)	-0.38	0.14	-2.68	0.023
<u>PFDCa (R²m: 0.54; R²C: 0.93)</u>				
Female plasma	1.43	0.35	4.13	<0.01
Egg number (2)	-0.93	0.19	-5.02	<0.001
<u>PFUnA (R²m: 0.38; R²C: 0.95)</u>				
Female plasma	0.96	0.39	2.42	0.032
Egg number (2)	-5.29	0.54	-9.86	<0.001
<u>PFDoA (R²m: 0.34; R²C: 0.90)</u>				
Egg number (2)	-1.92	0.23	-8.52	<0.001

PFTriA (R²m: 0.61; R²C: 0.79)

Female plasma	1.09	0.37	2.98	0.012
Egg number (2)	-1.10	1.50	-7.36	<0.001

PFTeA (R²m: 0.53; R²C: 0.79)

Female plasma	1.34	0.52	2.59	0.024
Egg number (2)	-2.21	0.34	-6.60	<0.001

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.^{20, 23, 32} PFAS have been found to have relatively strong
435 binding abilities to yolk proteins (low-density lipoprotein, high-density lipoprotein and vitellin
436 proteins).¹¹⁵ All yolk proteins (except immunoglobulins) being synthesized in the liver, it makes
437 it the entry point for PFAS in egg yolks.¹¹⁶ 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.^{20, 23} 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

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