

1
2
3
4 TEMPORAL VARIATION IN CIRCULATING CONCENTRATIONS OF
5 ORGANOCHLORINE POLLUTANTS IN A PELAGIC SEABIRD
6 BREEDING IN THE HIGH ARCTIC

7 JAN O. BUSTNES,*† BÅRD-JØRGEN BÅRDSSEN,† BØRGE MOE,‡ DORTE HERZKE,§ SVEINN
8 A. HANSSEN,† KJETIL SAGERUP,|| CLAUD BECH,# TORE NORDSTAD,# OLIVIER CHASTEL,††
9 SABRINA TARTU,†† ‡‡ and GEIR WING GABRIELSEN‡‡

10 † Norwegian Institute for Nature Research, Arctic Ecology Department, FRAM – High North
11 Research Centre on Climate and the Environment, NO-9296 Tromsø, Norway

12 ‡ Norwegian Institute for Nature Research, NO-7385 Trondheim, Norway

13 § Norwegian Institute for Air Research, FRAM – High North Research Centre on Climate and the
14 Environment, NO-9296 Tromsø, Norway

15 || Akvaplan-niva, FRAM – High North Research Centre on Climate and the Environment, NO-9296
16 Tromsø, Norway

17 # Department of biology, Faculty of Science and Technology, Norwegian University of Science and
18 Technology, NO-7491 Trondheim, Norway

19 †† Centre d'Etudes Biologiques de Chizé, Centre National de la Recherche Scientifique, FR-79360
20 Villiers en Bois, Deux-Sevres, France

21 ‡‡ Norwegian Polar Institute, FRAM – High North Research Centre on Climate and the Environment,
22 NO-9296 Tromsø, Norway

23 * Address of correspondence to jan.o.bustnes@nina.no

24
25 This is the peer reviewed version of the following article:

Bustnes, Jan Ove; Bårdsen, Bård-Jørgen; Moe, Børge; Herzke, Dorte; Hanssen, Sveinn Are; Sagerup, Kjetil; Bech, Claus; Nordstad, Tore; Chastel, Olivier; Tartu, Sabrina; Gabrielsen, Geir W.. Temporal variation in circulating concentrations of organochlorine pollutants in a pelagic seabird breeding in the high arctic. *Environmental Toxicology and Chemistry* 2017 ; Volum 36.(2) s. 442-448, which has been published in final form at DOI 10.1002/etc.3560. This article may be used for non-commercial purposes in accordance with Wiley Terms and Conditions for Self-Archiving

26

27

28 * Author and address of correspondence: Norwegian Institute for Nature Research, FRAM – High
29 North Research Centre on Climate and the Environment, N-9296 Tromsø, Norway (jan.o.
30 bustnes@nina.no, Phone: +47 77 75 04 07, Fax +47 77 75 04 01).

31

32 **Abstract:** The present study explored short-term temporal variations in circulating
33 concentrations of three legacy organochlorines (OCs) with different physicochemical
34 properties (polychlorinated biphenyl 153 [PCB-153], *p,p'*-dichlorodiphenyldichloroethylene
35 [DDE], and hexachlorobenzene [HCB]) in breeding kittiwakes (*Rissa tridactyla*) in a colony
36 in Svalbard (78°N), Norwegian Arctic. Concentrations were measured in blood of a large
37 number (n = 412-521 blood samples, depending on the data-analyses), of pre-breeding,
38 incubating and chick-rearing birds over a period of five years (2007-2011). PCB-153
39 concentrations were equal in male and female blood in the pre-breeding period, whereas
40 females had significantly lower concentrations during incubation and chick rearing, probably
41 due to their ability to eliminate OCs through egg laying. A similar temporal pattern was
42 observed with DDE although the lower concentrations in incubating females were not
43 significant. Males and females had similar concentrations of HCB over all reproductive
44 stages. The concentrations of all three compounds varied greatly between years. PCB-153
45 tended to decline over the study period while HCB showed an increasing trend, especially
46 among chick-rearing males late in the season. Concentrations of PCB-153 increased about 2.5
47 times from the pre-breeding to the chick-rearing period, concurrent with mobilization of body
48 lipids (reduced body mass). A similar, but less pronounced trend was found for HCB. For
49 DDE, however, kittiwakes had the highest concentrations in the pre-breeding period,
50 suggesting relatively high exposure in their winter areas. The present study documented large
51 variations in circulating concentrations of legacy OCs among and within breeding seasons in
52 kittiwakes, but the alterations within seasons were relatively consistent from year to year.

53

54 **Keywords:** Kittiwake Organochlorines Marine pollution Seasonal variation

55

56

INTRODUCTION

57 Organochlorines (OCs) are lipid-soluble pollutants that may undergo long-range transport to
58 the Arctic, where they bio-magnify in local food webs [1, 2]. Due to their high trophic
59 positions, arctic seabirds accumulate relatively high loads of such compounds [3, 4]. Many
60 OCs were, however, banned in most countries decades ago (e.g. DDT and PCB), which has
61 resulted in declining concentrations in arctic biota, also in seabirds [5, 6].

62 In seabirds, as in other wildlife species, OCs are distributed among different body
63 compartments: e.g., they may be stored in lipids within various organs, in adipose tissue, or
64 they may be circulating in the blood stream. There are three main processes determining the
65 concentrations of circulating OCs: 1) intake via food; 2) degree of remobilization of OCs
66 from adipose tissue; 3) removal through different elimination routes, including egg laying [7].
67 In addition, the lipid content of blood and tissue is also an important determinant for these
68 concentrations [8].

69 OC concentrations in blood of arctic seabirds may show high inter-annual variation
70 due to temporal variability in: 1) long-range transport through alterations in the atmosphere
71 and oceans [9-11] and/or; 2) diet; i.e. variation in the availability of prey with different lipid
72 and OC loads [12, 13]; 3) Variation in temperatures; i.e. at low temperatures the daily energy
73 expenditure and thus lipid metabolism increase, which remobilize more stored OCs, and
74 subsequently the circulating concentrations will increase [8, 14]; 4) Arctic seabirds may also
75 carry OCs during migration (bio-transport), and annual variation during breeding may be a
76 reflection of the variation in OCs exposure at the wintering grounds [15, 16].

77 Variation in circulating OC concentrations within breeding seasons may also arise
78 from altered transport, diet and remobilization due to temperature variability, but also because
79 the body condition (lipid stores) of seabirds often vary consistently within breeding seasons
80 [17, 18]. Moreover, there may be variation across the sexes, and females may eliminate some

81 of their OC loads through egg production. Although some seabird studies have found lower
82 concentrations of OCs in the blood of incubating females compared to males [19-21], the
83 importance of egg laying as a determinant for OCs in blood has been little studied in wild
84 birds [7, 22].

85 The circumpolar black-legged kittiwake (*Rissa tridactyla*) (hereafter 'kittiwake') is a
86 pelagic and surface-feeding seabird with a diet of different fish species and invertebrates
87 [23]. Arctic breeding kittiwakes usually lay two eggs (1-3) and incubate for approximately 27
88 days. Hatching occurs in early July, and parents rear chicks at the nest for 4-5 weeks [24]. The
89 kittiwake leaves the Arctic in October/November and returns in April [25, 26]. In order to
90 explore the short-term dynamics of circulating OCs, we measured three legacy OCs with
91 different physicochemical properties; polychlorinated biphenyl 153 (PCB-153); *p,p'*-
92 dichlorodipenyldichloroethylene (DDE) and; hexachlorobenzene (HCB) in more than 500
93 kittiwake blood samples from a colony at Svalbard, over 5 years. Samples were collected
94 during three reproductive stages (April: pre-breeding; mid-June: incubation; and late-July:
95 chick rearing). HCB is a semi-volatile compound with high long-range transport potential that
96 show high concentrations at remote locations such as the Arctic and the Antarctic. In
97 comparison, PCB-153 is a much heavier and very persistent molecule with a relatively lower
98 atmospheric transport potential. DDE is a metabolite of DDT and in seabirds, its occurrence
99 correlates to a varying degree with other legacy OCs such as PCB [14, 27-32], even if
100 historical industrial applications have been quite different. These three OCs were chosen as
101 key compounds because they have been linked to changes in physiology, reproduction and
102 survival of Svalbard kittiwakes and they represent a broad range of physiochemical properties
103 characteristic of the OC class of compounds [3, 33, 34].

104 In the present study, we hypothesized that egg laying induces differences in OC
105 circulating concentrations between the as females may reduce their OC loads through

106 contaminant deposition into their eggs. Consequently, we predicted that concentrations would
107 be equal for male and female kittiwakes prior to egg laying, but lower for females after egg
108 laying. Secondly, since body condition of kittiwakes tend to decline from the pre-laying to the
109 chick-rearing stages [18, 35, 36] we hypothesized increasing remobilization of OCs from
110 adipose tissue, thus increasing blood concentrations as breeding progressed. One central
111 question was whether changes in body mass (body lipids) over the breeding season could
112 explain alterations in OC concentrations, or if some other factors related to the different
113 reproductive stages would be of importance. Based on the different physiochemical properties
114 of the three OCs, we predicted different dynamics in kittiwakes, with a stronger fluctuation in
115 HCB concentrations over the breeding season, relative to PCB-153 and DDE (higher
116 volatility and lower K_{ow} and less lipophilic of HCB compared to PCB and DDE). We tested
117 the hypotheses both for wet weight and lipid normalized concentrations.

118

119 MATERIALS AND METHODS

120 *Study species and field procedures*

121 The kittiwake is a long-lived gull with a circumpolar distribution, breeding in
122 colonies on cliffs. In Svalbard, it feeds mainly on capelin (*Mallotus villosus*), polar cod
123 (*Boreogadus saida*) and amphipods [37, 38]. Krykkjefjellet, our study colony, is a seabird
124 cliff located 6 km southeast of Ny-Ålesund in Kongsfjorden, Svalbard (78°54'N, 12°13'E).
125 The kittiwakes were caught on their nest with a snare on a long fishing rod. All birds were in
126 adult breeding plumage: i.e. no dark patches on their heads or black fields on their outer
127 primaries [23]

128 Blood samples for contaminant analyses were taken from both sexes during the pre-
129 breeding, incubation and chick rearing periods between 2007 and 2011. Samples (~1.5 ml

130 blood) were taken from the brachial vein using a heparin treated 2 mL syringe and a 23G
131 needle. In the field, the samples were immediately stored in darkness at ambient temperatures
132 in June and July. In April, the samples were kept from freezing. When returning from the
133 field all the samples were frozen at -20°C.

134 Body mass and skull length (head and bill) were measured with an accuracy of 5 g and
135 0.1 mm, respectively, and the birds were banded with a metal and a three-letter coded plastic
136 ring [34]. Confirmation of breeding status (pre-laying, incubation and chick rearing) was done
137 by inspecting nest content using a mirror at the end of a long rod.

138 Permissions for fieldwork and blood sampling of the birds were granted by the
139 Governor of Svalbard and complied with the Norwegian Regulations on Animal
140 Experimentation.

141 *POPs analyses*

142 The concentrations of POPs in the blood samples were analyzed at the Norwegian
143 Institute for Air Research (NILU) in Tromsø, Norway. Details regarding the analyses are
144 provided in Herzke et al. [39] and in Nordstad et al. [36]. Lipid content in the blood samples
145 were analyzed gravimetrically. We analyzed blood lipids for all years, except in 2009 due to a
146 laboratory problem. We analyzed the correlations of different factors both on wet weight (all
147 years except 2009) and lipid normalized concentrations.

148 *Molecular sexing*

149 The sex of the birds was determined at the Centre d'Etudes Biologiques de Chizé
150 (CEBC-CNRS, University of La Rochelle). To sex the birds, DNA was extracted from red
151 blood cells and the CHD gene was amplified in a PCR procedure, as described by
152 Weimerskirch et al. [40].

153

154 *Statistical Analysis*

155 Statistical analyses and plotting of results were carried out in R [41]. All tests were
156 two-tailed, the null-hypothesis was rejected at an α -level of 0.05, and we used the treatment
157 contrast comparing each level of a factor to its baseline level. Standard plotting diagnostics
158 tools were used in all analyses, and as none of the residuals was normally distributed, the
159 responses were \log_e -transformed.

160 Prior to the statistical analyses, we assessed collinearity in several steps. Firstly, we
161 assessed the extent to which kittiwake body mass varied as a function of the other predictors
162 by forming a set of different *a priori* models. The selected model explained >60 % of the
163 variance in body mass. As body mass was related to year, period and sex in addition to two
164 interactions involving period we chose to separate the analyses between the sexes. Secondly,
165 collinearity was then assessed by checking if the effect sizes or their level of statistical
166 significance differed depending on whether each of the other predictors was excluded or not.
167 Thirdly, Variance Inflation Factor (VIF), with a cut-off value of 5, was used to assess which
168 predictors that are collinear and consequently should be dropped prior to the analyses [42]. As
169 we were unable to reveal any collinearity, we concluded that our separation of the analyses of
170 contaminants between the sexes were sufficient to avoid potential problems with serious
171 confounding.

172 We performed our statistical analyses in several steps. Firstly, we performed a set of
173 overall analyses in order to assess the extent in which the different responses varied according
174 to period and sex (keeping body mass out due to the collinearity issues outlined above), by
175 fitting linear mixed-effect models (LME) [43] with reproductive period, sex and their
176 interaction as fixed effects, whereas the constant term for year was used as a random effect
177 (i.e. random intercepts only). Secondly, we performed more detailed analyses (on each sex

178 separately), where we fitted linear models (LMs), using the *lm* function in *R* in order to assess
179 the effects of body mass, period and year. In these analyses, we calculated the second-order
180 *Akaike's Information Criterion* (AICc) values for several candidate models [42, 44]. The
181 models used for inference were selected by: 1) forming a set of candidate models where we
182 rescaled and ranked models relative to the model with the lowest AICc value (Δ_i denotes this
183 difference for model *i*) and then by 2) selecting the simplest model with a $\Delta_i \leq 1.5$ even
184 though we also provide Akaike's weights. Model selection was performed using the
185 *AICcmodavg* library in *R* [45]. We kept body mass in all models based on our *a priori*
186 expectations, whereas the other covariates were excluded or included in the different
187 candidate models. It can be argued that LMEs using individual as a random effect represent a
188 more correct statistical approach than the LM approach applied in the above analyses [42].
189 Nonetheless, there are several reasons why we applied LMs and not LMEs. Firstly, out of our
190 total sample of 529 observations, 27 were data from unknown individuals. Second and more
191 importantly, approximately 50% of our observations (from known individuals) were single
192 samples taken from one individual and 70% consisted of individuals being samples only once
193 or twice (both sexes), whereas only 15% (males) and 10% (females) consisted of individuals
194 that had been sampled ≥ 4 times. Thirdly, as a precautionary action we fitted LME versions of
195 all selected models, i.e. a model with the same fixed effects and random intercepts only, using
196 the *nlme* library [46]. As neither the estimates nor the statistical significance for our estimates
197 changed notably when comparing the reported output for the analyses using period and body
198 mass as predictors to LMEs (results not shown), we conclude that our inference were not
199 sensitive to our choice of statistical approach.

RESULTS AND DISCUSSION

201 *Differences between sexes*

202 Studies of different seabird species have shown that males may have higher circulating
203 concentrations of OCs than females during breeding [19-21], but this does not appear to be
204 consistent [47]. Differences in OC loads between sexes have been attributed to both egg
205 laying and diet specialization [7, 22]. Since we had data both prior to – and after egg laying (n
206 = 521), we were able to test the hypothesis that such differences was an effect of female
207 kittiwakes eliminating OCs through the eggs. In the LMEs, i.e. when year was used as a
208 random factor, the sexes had equal levels of PCB-153 in the pre-laying period, whereas males
209 had higher concentrations during incubation and chick rearing (~10%): interactions being
210 significant for lipid-normalized concentrations ($p = 0.033$; Supplemental Data, Table S4A),
211 and marginally significant ($p = 0.075$; Supplemental Data, Table S3A) for wet weight
212 concentrations (Figure 1A; Supplemental Data, Figure S1A; bar plots are used for visualizing
213 model predictions, whereas points are used to show descriptive statistics). For DDE the
214 differences were in the same direction as PCB-153, but the blood concentrations tended to
215 decline over the breeding season in both sexes and the interactions between period and sex
216 were not significant ($p = 0.107$, Figure 1B; Supplemental Data, Figure S1B, Table S3B, S4B).
217 This suggests that egg laying has some impact on the circulating concentrations of DDE in the
218 breeding season, but the effect appear to be weak, and it is important to note the variation
219 among and within breeding seasons (Figure 2A-C; Supplemental Data, Figure S2A-C). That
220 is, in some years females had equally high blood concentrations as males during both
221 incubation and chick-rearing periods. This might be a result of poor feeding conditions in
222 which females are forced to emancipate their body lipid reserves and thereby remobilize more
223 OCs during egg laying and incubation periods [21]. For HCB, however, there appeared to be
224 no difference (interaction: $p = 0.38$) between sexes in any of the periods (Figure 1C;

225 Supplemental Data, Figure S1C, Table S3C, S4C). This was unexpected since maternal
226 transfer of contaminants to eggs often favors low K_{ow} and/or less persistent OCs, whereas
227 more lipophilic compounds such as PCB-153 are more likely to be retained in the mother's
228 adipose tissue [48-50]. The lack of difference between the sexes after egg laying could result
229 from the relatively high continuous exposure of HCB in our study area. Hence, the observed
230 air concentrations on Svalbard are 80 pg/m^3 for HCB and only 10 pg/m^3 for PCB, showing
231 different background exposure [51]. Moreover, in the same location and time period as the
232 present study, blood concentrations of HCB increased more during incubation fast in common
233 eiders (*Somateria mollissima*), a benthic top predator, than did PCB-153, suggesting high
234 intake of HCB during pre-breeding accumulation of body reserves [14]. Hence, female
235 kittiwakes in Kongsfjorden may rapidly regain the loads lost through the eggs, and thus
236 potential differences between sexes may not be detected. Alternatively, HCB has relatively
237 low affinity for adipose tissue compared to other compounds [50, 52], and this compound
238 may therefore be more readily redistributed from body fat to blood after egg laying.

239

240 *Temporal variation in OCs and the effect of body condition*

241 Descriptive data on concentrations for both sexes ($n = 270$ males and 224 females) can
242 be found in SI Table 5. The sexes were analyzed separately, and for each contaminant, the
243 same models were selected and used for inferences (Supplemental Data, Table S1, S2). For
244 wet weight, the best models explained 37% and 34% of the variance in blood concentrations
245 for PCB-153 for male and female kittiwakes, respectively (Table S6A), whereas the
246 corresponding values were 16% and 21% for DDE (Table S6B), and 19% and 18% for HCB
247 (Table S6C). The corresponding values for lipid-normalized concentrations were within the
248 same order of magnitude, although lipid data for 2009 was lacking (Supplemental Data, Table
249 S7A-C).

250 The variation in wet weight blood concentrations among years was generally the most
251 important variance component explaining 23% and 30% (partial R^2) of the variation in PCB-
252 153 in males and females, respectively. The corresponding values were 8% and 12% for
253 DDE, and 10% and 8% for HCB, respectively. For PCB-153, the mean wet weight
254 concentrations over the whole seasons varied from ~4000-8000 pg/g (wet weight) between
255 years in males, and between ~2200-7500 pg/g in females (Supplemental Data, Table S5). The
256 individuals sampled from 2009 to 2011 had lower levels (negative estimates for these years
257 after controlling for body mass) when compared to the first year of the study (2007), whereas
258 the levels in 2008 were slightly higher, although not significantly different from 2007 (SI
259 Table 6a). Interestingly, 2008 was a very cold summer [14] and kittiwakes may have been
260 forced to increase daily energy expenditure by metabolizing more lipids and thus remobilizing
261 more PCB to the blood [14]. The lipid-normalized concentrations for PCB-153
262 (Supplemental Data, Figure S2A) showed a similar pattern as for wet weight, especially in
263 females (Supplemental Data, Table S7A, Figure S2A), suggesting that PCBs are declining in
264 the study area consistent with declining background exposure [53].

265 For DDE, mean concentrations in males varied from ~2000-3500 pg/g (wet weight)
266 between years, over all seasons, and between ~1700-3300 pg/g in females (Supplemental
267 Data, Table S5). Males sampled in 2009 had lower wet weight concentrations compared to
268 2007, whereas the concentrations in other years (2008 and 2010-2011) were not significantly
269 different from 2007. For the females, however, all years except 2008 showed lowered levels
270 compared to 2007, similar to PCB-153 (Supplemental Data, Table S6B, Figure S2B). For
271 lipid-normalized concentrations, however, year did not improve the statistical models and was
272 not included in the best model after controlling for reproductive period and body mass
273 (Supplemental Data, Table S7B, Figure S2B). Consistent temporal trends of legacy OCs may
274 be impossible to document with certainty over just five years, and for DDE it seems more

275 likely that the trends observed are results of differences in lipid content in blood between
276 years.

277 The mean concentrations of HCB in male kittiwakes varied from ~1500-2900 pg/g
278 (wet weight) between years over the whole seasons, and between ~1600-3200 pg/g in females
279 (Supplemental Data ,Table S5). For HCB, however, the wet weight concentrations tended to
280 increase over the years when controlling for body mass (Supplemental Data, Table S6C,
281 Figure S2C). The temporal patterns were, however, different between the sexes: males
282 showed increasing levels of HCB from 2009 to 2011 (relative to 2007), whereas females
283 showed lower levels in 2008 and higher levels in 2009 compared to 2007 (Supplemental Data,
284 Table S5C, Figure S2C). For lipid-normalized concentrations, there was still an increase of
285 HCB in 2008 and 2010 compared to 2007 after controlling for body mass and reproductive
286 period (Supplemental Data, Table S7C, Figure S2C). Hence, despite the short period, wet
287 weight concentrations showed a directional increase for HCB, especially in males (Figure 2C;
288 Supplemental Data, Figure S2C). The changes in HCB concentrations may be expected since
289 the background exposure of HCB in Kongsfjorden is increasing [53]. This explanation is
290 strengthened by the fact that the most pronounced increase seemed to occur late in the
291 breeding season, when the birds had spent about four months in the Kongsfjorden area.

292 Although the inter-year variation in blood concentrations was large, there was also
293 considerable variation between the different reproductive stages. For PCB-153 the lowest wet
294 weight concentrations were generally found in the pre-breeding periods, increasing on
295 average approximately 2.5 times until the chick-rearing periods (Figure 2A; Supplemental
296 Data, Table S5). HCB increased on average about 1.2 times over the same period (Figure 2C;
297 Supplemental Data, Table S5). For both compounds, the increase occurred concurrently with
298 reductions in body mass (Figure 3), and the changes in body mass (lipid stores) eliminated
299 reproductive stage as a significant predictor in the statistical models (Supplemental Data,

300 Table S6A, S6C). For PCB-153, body mass explained 26.5% of the variation in males, but
301 only 9% in females, possibly an effect of egg laying. The values for HCB were lower: 6% and
302 3% for males and females, respectively. For PCB-153, the lipid-normalized concentrations
303 showed similar patterns as wet weight (Supplemental Data, Table S6A, Figure S2A), whereas
304 for lipid-normalized HCB concentrations, the best model also included reproductive stage.
305 The effect of body mass was, however, not included in the best model for females
306 (Supplemental Data, Table S6C, Figure S2C). The relatively low explanatory power of the
307 statistical models for HCB compared to PCB-153 may again originate from the higher local
308 exposure of HCB and more rapid remobilization of this compound.

309 DDE showed a different pattern compared to the other compounds, as the highest
310 concentrations were found in the pre-breeding periods, mean concentrations being
311 approximately 1.25 times higher than during incubation, with a slight increase during chick
312 rearing (Figure 2B; Supplemental Data, Table S5). Changes in body mass explained 5% and
313 3% of the wet weight concentrations of DDE in male and female kittiwakes respectively,
314 whereas the corresponding values for the reproductive stage predictor were 9% and 8%
315 (Supplemental Data, Table S6B). The lipid-normalized DDE concentrations (Supplemental
316 Data, Figure S2B) showed the same pattern as for wet weight concentrations with regard to
317 reproductive stage and body mass (Supplemental Data, Table S7B). Hence, despite decreasing
318 body mass from April to June (Figure 3), DDE in blood decreased, suggesting that the
319 breeding area in Kongsfjord has lower background exposure of DDE, or the mother
320 compound DDT, than the winter areas. Kittiwakes thus seem to eliminate DDE from their
321 bodies relatively fast until concentrations reach an equilibrium with their breeding
322 environment [7]. This indicates that kittiwakes are net-transporters of DDE/ DDT to the High
323 Arctic. The kittiwakes breeding in Kongsfjorden winter at 40-60°N in the North Atlantic,
324 roaming across the western- (Grand Banks and Labrador Sea), central (Mid Atlantic) and

325 eastern parts (Mid-Atlantic Ridge to Portugal/Ireland) [25, 26]. Exposure of organic
326 pollutants in this offshore region is not well studied: i.e. whether the high levels of DDE
327 compared to other legacy OCs in pre-breeding kittiwakes can be attributed to this region is
328 not known. However, Espin et al. [54] reported that razorbills (*Alca torda*) were exposed to
329 high amounts of DDT along the coast of Spain, which was almost completely metabolized
330 when they reached their breeding grounds in Northern Europe. In Norwegian lesser black-
331 backed gulls (*Larus fuscus*), high levels of DDE were found in eggs and blood of birds
332 wintering in African lakes where DDE is a dominating contaminant, compared to gulls
333 wintering in areas dominated by PCB [15]. This may indicate that DDE and/or DDT are
334 compounds prone to bio-transport.

335 Diet variability may cause variation in circulating concentrations of OCs [13]. In
336 2007, when the study started, 75% of the kittiwake diet during chick rearing consisted of
337 capelin, a relatively lipid-rich fish, whereas in the other years capelin made up <15% of the
338 diet. In 2010, low trophic krill (Euphausiids) made up nearly 50% of the diet. In 2009 and
339 2011 nearly 50% of the diet was made up by polar cod (G.W. Gabrielsen, Norwegian Polar
340 Institute, unpublished data). Although high concentrations of OCs in 2007 could potentially
341 result from high intake of lipid-rich capelin, this cannot explain the high levels in 2008.
342 Hence, there seems to be no consistent pattern in the diet data, e.g. trophic position of prey,
343 coherent with the variation in OC concentrations in the present study. There may be several
344 reasons for this, notably that the diet data has been collected in a limited period of the
345 breeding stage (during chick rearing), whereas blood has been sampled over the whole
346 breeding season. It might also be that diet samples were intended for the chicks, whereas the
347 adults are feeding on different prey as found in some other seabirds [55].

348 The present study demonstrated large variations in circulating concentrations of
349 different legacy OCs during breeding in high arctic kittiwakes. Firstly, egg laying seemed to

350 reduce circulating levels of PCB-153 in females relative to males. This effect, however, was
351 surprisingly not significant for DDE, and not found for HCB. The OCs behaved differently in
352 the birds and relatively simple statistical models may explain much of the variation in
353 circulating concentrations of PCB-153, the most persistent compound. For HCB, however, the
354 present study suggests that local exposure during the breeding season may be more important
355 relative to the other compounds. The fact that HCB tended to increase over the years, supports
356 this explanation since background HCB is increasing in the Kongsfjorden area [53]. This is
357 worrying since HCB has been associated with lower adult survival probabilities in this
358 kittiwake population [34]. For DDE, the situation seems more complex since the birds appear
359 carry of this compound from the wintering grounds. For all three compounds, however, the
360 present study demonstrates great variance between different reproductive stages, with mean
361 PCB-153 concentrations increasing approximately 2.5 times over the breeding season.
362 Moreover, these differences seem to be consistent among years, and much of the differences
363 can be explained by reductions of the birds' lipid stores. This could be a result of energetic
364 stress during the breeding period [18]. The importance of different environmental factors (e.g.
365 climate variables) in causing variability is poorly understood, and more data over several
366 years is necessary to elucidate such links. Moreover, variation in diet probably has a great
367 impact on OC intake of kittiwakes, and should be addressed in future studies, e.g. by
368 measuring isotopes. The present study, however, emphasizes that sampling time is an
369 important factor if seabird tissue, such as blood, should be used for monitoring purposes. In
370 the future this might become an even more important issue since many seabirds are
371 threatened, notably pelagic species such as kittiwakes [56], and the need for non-invasive
372 sampling methods increases. Blood sampling is a viable alternative to various forms of
373 invasive sampling.

374

375

SUPPLEMENTAL DATA

376 Supplemental Data (Table S1-S7, Figure S1, S2) is available on the Wiley Online library at

377 DIO:

378 *Acknowledgment*- We are grateful to the staff at Ny Ålesund research station (Kings Bay and

379 Norwegian Polar Institute) and for valuable support during fieldwork, and two anonymous

380 reviewers for comments that greatly improved the manuscript. The Norwegian Research

381 Council through the COPOL and AVITOX projects funded the study (Project numbers:

382 176073 and 234423) and by the Institut Paul-Émile Victor (IPEV Programme 330 to O

383 Chastel).

384 *Data availability*- Data, associated metadata, and calculation tools are available from the

385 corresponding author (jan.o.bustnes@nina.no).

386

REFERENCES

387

- 388 1. Borgå K, Gabrielsen GW, Skaare JU. 2001. Biomagnification of organochlorines
389 along a Barents Sea food chain. *Environ Pollut* 113: 187-198.
390
- 391 2. Kozak K, Polkowska Z, Ruman M, Koziol K, Namiesnik J. 2013. Analytical studies
392 on the environmental state of the Svalbard Archipelago provide a critical source of
393 information about anthropogenic global impact. *Trac-Trends Anal Chem* 50: 107-126.
394
- 395 3. Letcher RJ, Bustnes, JO, Dietz D, Jenssen BM, Jørgensen EJ, Sonne C, Verreault J,
396 Vijayan MM, Gabrielse, GW. 2010. Effect assessment of persistent organic pollutants
397 in arctic wildlife and fish. *Sci Tot Environ* 408: 2995-3043.
398
- 399 4. Bourgeon S, Leat EHK, Strøm H, Furness R.W, Magnusdottir E, Fisk AT, Ellis S,
400 Petersen Æ, Olafsdottir K, Borgå K, Hanssen SA, Gabrielsen GW, Bustnes JO. 2012.
401 Individual variation in biomarkers of health: influence of persistent organic pollutants
402 in great skuas (*Stercorarius skua*) breeding along a geographic gradient. *Environ Res* ,
403 118: 31-39.
404
- 405 5. Riget F, Bignert A, Braune B, Stow J, Wilson S. 2010. Temporal trends of legacy
406 POPs in Arctic biota, an update. *Sci Tot Environ* 408: 2874-2884.

- 407
408
409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427
428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444
445
446
447
448
449
6. Helgason LB, Sagerup K, Gabrielsen GW. 2011. *Organohalogen pollutants in seabird eggs from Northern Norway and Svalbard*. In Loganathan, B.G., Lam, P.K.S. (eds.). *Global Contamination Trends of Persistent Organic Chemicals*. CRC Press. Pp 547-570.
 7. Drouillard KG. 2000. Modeling the toxicokinetics and biomagnification of polychlorinated biphenyls (PCBs) in birds. Ph.D. Thesis, Trent University, Peterborough, Ontario, Canada, p. 201.
 8. Haddad S, Poulin P, Krishnan K. 2000. Relative lipid content as the sole mechanistic determinant of the adipose tissue: blood partition coefficients of highly lipophilic organic chemicals. *Chemosphere* 40: 839-843.
 9. Macdonald RW, Harner T, Fyfe J. 2005. Recent climate change in the Arctic and its impact on contaminant pathways and interpretation of temporal trend data. *Sci Total Environ* 342: 5-86.
 10. Noyes PD, McElwee MK, Miller HD, Clark BW, Van Tiem LA, Walcott KC, Erwin KN, Levin ED. 2009. The toxicology of climate change: Environmental contaminants in a warming world. *Environ Int* 35: 971-986.
 11. Borgå K, Saloranta TM, Ruus A. 2010. Simulating climate change-induced alterations in bioaccumulation of organic contaminants in an arctic marine food web. *Environ Toxicol Chem* 29: 1349-1357.
 12. Hebert CE, Weseloh DVC, Idrissi A, Arts MT, O'Gorman R, Gorman OT, Locke B, Madenjian CP, Roseman EF. 2008. Restoring piscivorous fish populations in the Laurentian Great Lakes causes seabird dietary change. *Ecology* 89: 891-897.
 13. Bustnes JO, Erikstad KE, Bakken V, Mehlum F, Skaare JU. 2000. Feeding ecology and the concentration of organochlorines (OCs) in glaucous gulls. *Ecotoxicology* 9: 175-182.
 14. Bustnes JO, Moe B, Herzke D, Hanssen SA, Nordstad T, Fenstad A, Borgå K, Gabrielsen GW. 2012. Temporal dynamics of circulating persistent organic pollutant in a fasting seabird under different environmental conditions. *Environ Sci Technol* 46: 10287-10294.
 15. Bustnes JO, Helberg M, Strann KB, Skaare JU. 2006. Environmental pollutants in endangered vs. increasing subspecies of lesser black-backed gulls along the Norwegian Coast. *Environ Pollut* 144: 893-901.

- 450 16. Leat EHK, Bourgeon, S, Magnusdottir E, Gabrielsen GW, Grecian J, Hanssen SA,
451 Olafsdottir K, Petersen A, Phillips RA, Strøm H, Ellis S, Fisk AT, Bustnes JO,
452 Furness RW, Borgå K. 2013. Influence of wintering area on persistent organic
453 pollutants in a breeding migratory seabird. *Mar Ecol Prog Ser* 491: 277-293.
454
- 455 17. Moreno, J. 1989. Strategies of mass change in breeding birds. *Biol J Linnean Soc* 37:
456 297–310.
457
- 458 18. Moe B, Langseth I, Fyhn M, Gabrielsen GW, Bech C. 2002. Changes in body
459 condition in breeding kittiwakes *Rissa tridactyla*. *J Avian Biol* 33: 225-234.
460
- 461 19. Bustnes JO, Bakken V, Skaare J U, Erikstad KE. 2003. Age and accumulation of
462 persistent organochlorines: a study of arctic breeding glaucous gulls. *Environ Toxicol*
463 *Chem* 22: 2173-2179.
464
- 465 20. Bustnes JO, Tveraa T, Henden JA, Varpe Ø, Skaare JU. 2007. Reproductive
466 performance and organochlorine pollutants in an Antarctic marine top predator: the
467 south polar skua. *Environ Int* 33: 911-918.
468
- 469 21. Bustnes JO, Tveraa T, Fauchald P, Helberg M, Skaare JU. 2008. The potential impact
470 of environmental variation on the concentrations and ecological effects of pollutants in
471 a marine avian top predator. *Environ Int* 34: 193-201.
472
- 473 22. Norstrom RJ, Clark TP, Jeffrey DA, Won HT, Gilman AP. 1986. Dynamics of
474 organochlorine compounds in herring gulls (*Larus argentatus*): I. distribution and
475 clearance of [¹⁴C]DDE in free-living herring gulls (*Larus argentatus*). *Environ Toxicol*
476 *Chem* 5: 41-48.
477
- 478 23. Cram, S, Simmons KEL. 1983. Handbook of the birds of Europe the Middle East and
479 North Africa. The Birds of the Western Palearctic. Vol. III. Oxford University Press,
480 Oxford.
481
- 482 24. Moe B, Stempniewicz L, Jakubas D, Angelier F, Chastel O, Dienessen F, Gabrielsen
483 GW, Hanssen F, Karnovsky N, Rønning, B, Welcker J, Wojczulanis-Jakubas K, Bech
484 C. 2009. Climate change and phenological responses of two seabird species breeding
485 in the high-Arctic. *Mar Ecol Prog Ser* 393: 235–246.
486
- 487 25. Frederiksen M, Moe B, Daunt F, Phillips RA, Barrett RT, Bogdanova MI, Boulinier T,
488 Chardine JW, Chaste, O, Chivers LS, Christensen-Dalsgaard S, Clement-Chastel C,
489 Colhoun K, Freeman R, Gaston AJ, Gonzalez-Solis J, Goutte A, Gremillet D,
490 Guilford T, Jensen G, Krasnov Y, Lorentsen S-H, Mallory ML, Newell M, Olsen B,
491 Shaw D, Steen H, Strom S, Systad GH, Thorarinsson TL, Anker-Nilssen T. 2012.
492 Multicolony tracking reveals the winter distribution of a pelagic seabird on an ocean
493 basin scale. *Diversity and Distribution* 18: 530-542.

- 494
495 26. Schultner J, Moe B, Chastel O, Tartu S, Bech C, Kitaysky AS. 2014. Experimental
496 evidence for corticosterone as a mediator of carry-over effects between breeding and
497 migration. *Mar Ecol Prog Ser* 496: 125-133.
498
- 499 27. Wania, F.; Mackay. D. Tracking the distribution of persistent organic pollutants.
500 *Environ. Sci. Technol.* **1996**, *30*, 390-396.
501
- 502 28. AMAP. AMAP Assessment 2002: Persistent Organic Pollutants in the Arctic. Arctic
503 Monitoring and Assessment Program (AMAP), Oslo, 2004, Norway. xvi + 310 pp.
504 www.amap.no
505
- 506 29. Weber K, Goerke H. 2003. Persistent organic pollutants (POPs) in antarctic fish:
507 levels patterns, changes. *Chemosphere* 53: 667-678.
508
- 509 30. Simonich SL, Hites RA. Global distribution of persistent organochlorine compounds.
510 *Science* 269: 1851-1854.
511
- 512 31. Steffen LC, Borgå K, Skaare JU, Bustnes JO. 2006. The occurrence of organochlorine
513 pollutants in marine avian top predators along a latitudinal gradient. *Environ Sci*
514 *Technol* 40: 5139-5146.
515
- 516 32. Bustnes, J.O. 2006. Pinpointing potential causative agents in mixtures of persistent
517 organic pollutants in observational field studies: A review of glaucous gull studies. *J*
518 *Toxicol Environ Health, Part A.* 69: 97-108.
519
- 520 33. Tartu S, Angelier F, Herzke D, Moe B, Bech C, Gabrielsen GW, Bustnes JO, Chastel
521 O. 2014. The stress of being contaminated? Adrenocortical function and reproduction
522 in relation to persistent organic pollutants in female black legged kittiwakes. *Sci Tot*
523 *Environ* 476-477C: 553-560.
524
- 525 34. Goutte A, Barbraud C, Herzke D, Bustamante P, Angelier F, Tartu S, Clément-Chastel
526 C, Moe B, Bech C, Gabrielsen GW, Bustnes JO, Chastel O. 2015. Survival rate and
527 breeding outputs in a high Arctic seabird exposed to legacy persistent organic
528 pollutants and mercury. *Environ Pollut* 200: 1-9.
529
- 530 35. Henriksen EO, Gabrielsen GW, Skaare JU. 1996. Levels and congener pattern of
531 polychlorinated biphenyls in kittiwakes (*Rissa tridactyla*), in relation to mobilization
532 of body-lipids associated with reproduction. *Environ Pollut* 92: 27-37.
533
- 534 36. Nordstad T, Moe B, Bustnes JO, Gabrielsen GW, Bech C, Chastel O, Herzke D. 2012.
535 Relationships between POPs and baseline corticosterone levels in black-legged
536 kittiwakes (*Rissa tridactyla*) across their breeding cycle. *Environ Pollut* 164: 219-
537 226.

- 538
539 37. Mehlum, F.; Gabrielsen, G.W. The diet of high arctic seabirds in coastal and ice-
540 covered, pelagic areas near the Svalbard archipelago. *Polar Res* 12: 1-20.
541
- 542 38. Strøm H. 2005. Black-legged kittiwake, *Rissa tridactyla*. in: Kovacs KM, Lydersen C.,
543 eds. Birds and mammals of Svalbard. Grafisk Nord AS, Tromsø.
544
- 545 39. Herzke D, Nygård T, Berger U, Huber S, Røv N. 2009. Perfluorinated and other
546 persistent halogenated organic compounds in European shag (*Phalacrocorax*
547 *aristotelis*) and common eider (*Somateria mollissima*) from Norway: A suburban to
548 remote pollutant gradient. *Sci Tot Environ* 408: 340-348.
549
- 550 40. Weimerskirch H, Lallemand J, Martin J. 2005. Population sex ratio variation in a
551 monogamous long-lived bird, the wandering albatross. *J Anim Ecol* 74: 285-291.
552
- 553 41. Team, R. C. 2013. R: a language and environment for statistical computing. R
554 Foundation for Statistical Computing, Vienna, Austria.
555
- 556 42. Zuur AF, Ieno EN, Walker NJ, Saveliev A, Smith GM. 2009. Mixed effects models
557 and extensions in ecology with R. Springer, New York, USA.
558
- 559 43. Pinheiro JC, Bates DM. 2000. Mixed effect models in S and S-PLUS. Springer, New
560 York, USA.
561
- 562 44. Burnham KP, Anderson DR. 2002. Model selection and multimodel inference. A
563 practical information-theoretic approach. Springer-Verlag, New York.
564
- 565 45. Mazerolle M.J. 2013. AICcmodavg: Model selection and multimodel inference based
566 on (Q)AIC(c).
567
- 568 46. Pinheiro JC, Bates DM, DebRoy S, Deepayan S. 2012. R. D. C. Team. nlme: linear
569 and nonlinear mixed effects model. R package.
570
- 571 47. Helberg M, Bustnes JO, Erikstad KE, Kristiansen KO, Skaare JU. 2005. Relationships
572 between reproductive performance and organochlorine pollutants in great-black
573 backed gulls (*Larus marinus*). *Environ Pollut* 134: 475-483.
574
- 575 48. Drouillard KG, Norstrom RJ. 2001. Quantifying maternal and dietary sources of 2,2
576 ',4,4 ',5,5 '-hexachlorobiphenyl deposited in eggs of the ring dove (*Streptopelia*
577 *risoria*). *Environ Toxicol Chem* 20: 561-567.
578
- 579 49. Verreault J, Agudo Villa R, Gabrielsen GW, Skaare JU, Letcher RJ. 2006. Maternal
580 transfer of organohalogen contaminants and metabolites to eggs of Arctic-breeding
581 glaucous gulls. *Environ Pollut* 144: 1053-1060.

- 582
583 50. Louis C, Dirtu AC, Stas M, Guiot Y, Malarvannan G, Das K, Costa DP, Crocker
584 DE, Covaci A, Debier C. 2014. Mobilisation of lipophilic pollutants from blubber in
585 northern elephant seal pups (*Mirounga angustirostris*) during the post-weaning fast.
586 *Environ Res* 132: 438–448.
587
- 588 51. Wilson S, Hung H, Katsoyiannis A, Kong D, Oostdam JV, Riget F, Bignert A. 2014.
589 Trends in Stockholm Convention Persistent Organic Pollutants (POPs) in Arctic Air,
590 Human media and Biota. *Arctic Monitoring and Assessment Programme* (AMAP).
591
- 592 52. Lehman-McKeeman LD, Parkinson A, Ogilvie BW, Shen DD. 2008. Disposition of
593 toxicants in: Casarett and Doull's Toxicology, the basic science of poisons, 7 ed.
594 McGraw Hill, Kansas, US.
595
- 596 53. Hung H, Kallenborn R, Breivik K, Su Y, Brorstrom-Lunden E, Olafsdottir, K,
597 Thorlacius JM, Leppanen S, Bossi R, Skov H, Mano S, Patton GW, Stern G, Sverk, E,
598 Fellin, P. 2010. Atmospheric monitoring of organic pollutants in the Arctic under the
599 Arctic Monitoring and Assessment Programme (AMAP): 1993-2006. *Sci Tot Environ*
600 408: 2854-2873.
601
- 602 54. Espín S, Martínez-López E, Gómez-Ramírez, P, María-Mojica P, García-Fernández AJ.
603 2010. Assessment of organochlorine pesticide exposure in a wintering population of
604 razorbills (*Alca torda*) from the southwestern Mediterranean. *Chemosphere* 80: 1190-
605 1198.
606
- 607 55. Erikstad KE, Reiertsen TK, Barrett RT, Vikebo F, Sandvik H. 2013. Seabird-fish
608 interactions: the fall and rise of a common guillemot *Uria aalge* population. *Mar Ecol*
609 *Prog Ser* 475: 276.
610
- 611 56. Paleczny M, Hammill E, Karpouzi V, Pauly D. 2015. Population trends of the world's
612 monitored seabirds, 1950-2010. *PLoS One* DOI:10.1371/journal.pone.0129342
613
614
615
616
617
618
619
620
621

622 **Figure legends:**

623

624 **Figure 1.** Estimated, i.e. model predictions with precision [± 1 standard error (SE)] as bars,
625 wet weight concentrations (Log pg/g, from LMEs where we controlled for year as a random
626 factor) of (A) polychlorinated biphenyl 153 [PCB-153], (B) *p,p'*-
627 dichlorodiphenyldichloroethylene [DDE], and (C) hexachlorobenzene [HCB]) in male and
628 female kittiwakes in different reproductive stages; pre-breeding, incubation and chick-rearing.
629 Data from Kongsfjorden, Svalbard, 2007-2011.

630 **Figure 2.** Plot showing descriptive statistics, i.e. the average (points), median (-) as well as
631 the 25th and 75th percentiles (bars), for wet weight concentrations (pg/g wet weight) of of (A)
632 polychlorinated biphenyl 153 [PCB-153], (B) *p,p'*-dichlorodiphenyldichloroethylene [DDE],
633 and (C) hexachlorobenzene [HCB]) in male (blue bars) and female (red bars) kittiwakes in
634 different reproductive stages; pre-breeding, incubation and chick-rearing. Data from
635 Kongsfjorden, Svalbard, 2007-2011.

636 **Figure 3.** Plot showing descriptive statistics, i.e. the average (points), median (-) as well as
637 the 25th and 75th percentiles (bars), for body mass in male (blue) and female (red) kittiwakes
638 in different reproductive stages; pre-breeding, incubation and chick-rearing. Data from
639 Kongsfjorden, Svalbard, 2007-2011.

640

641

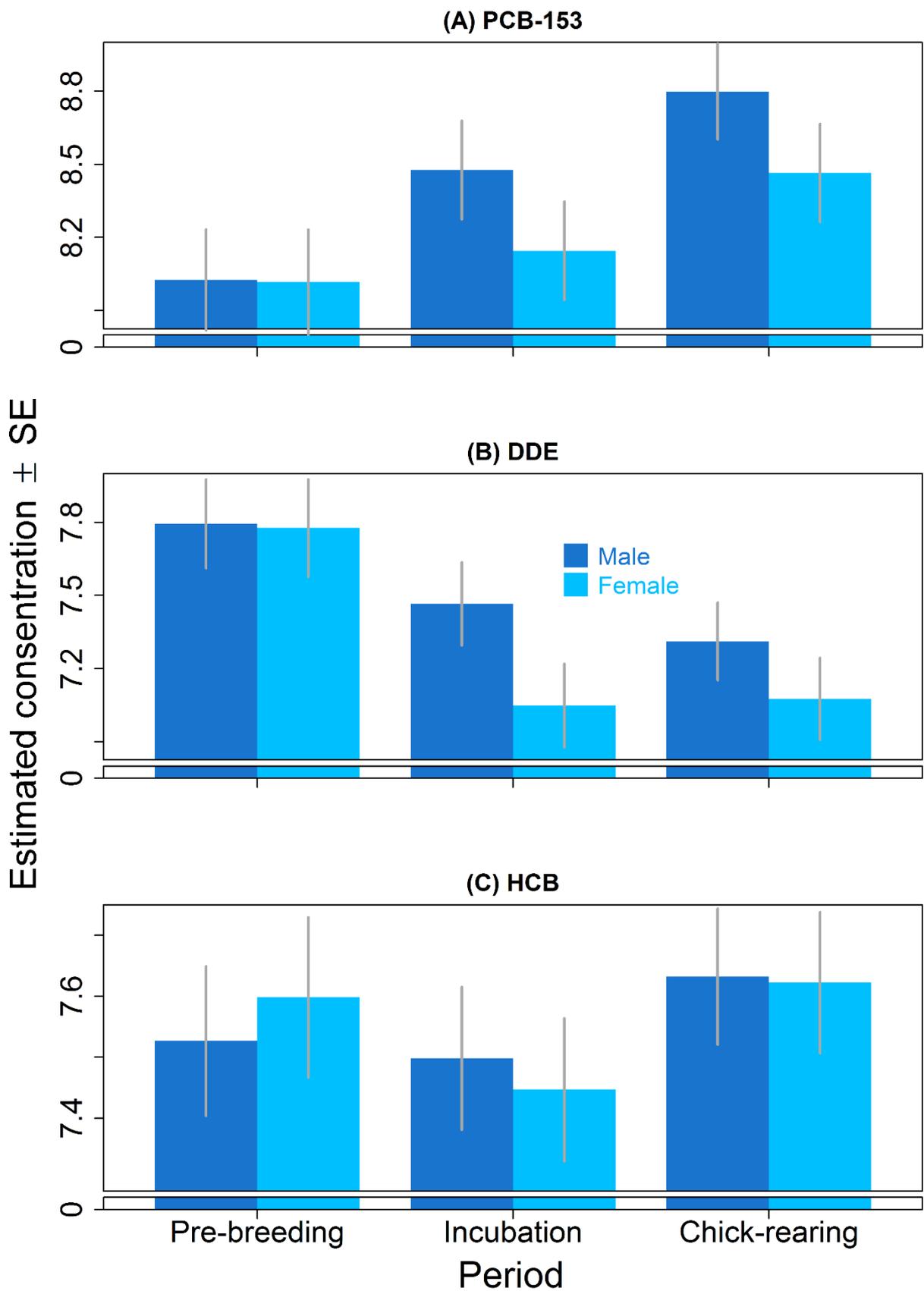
642

643

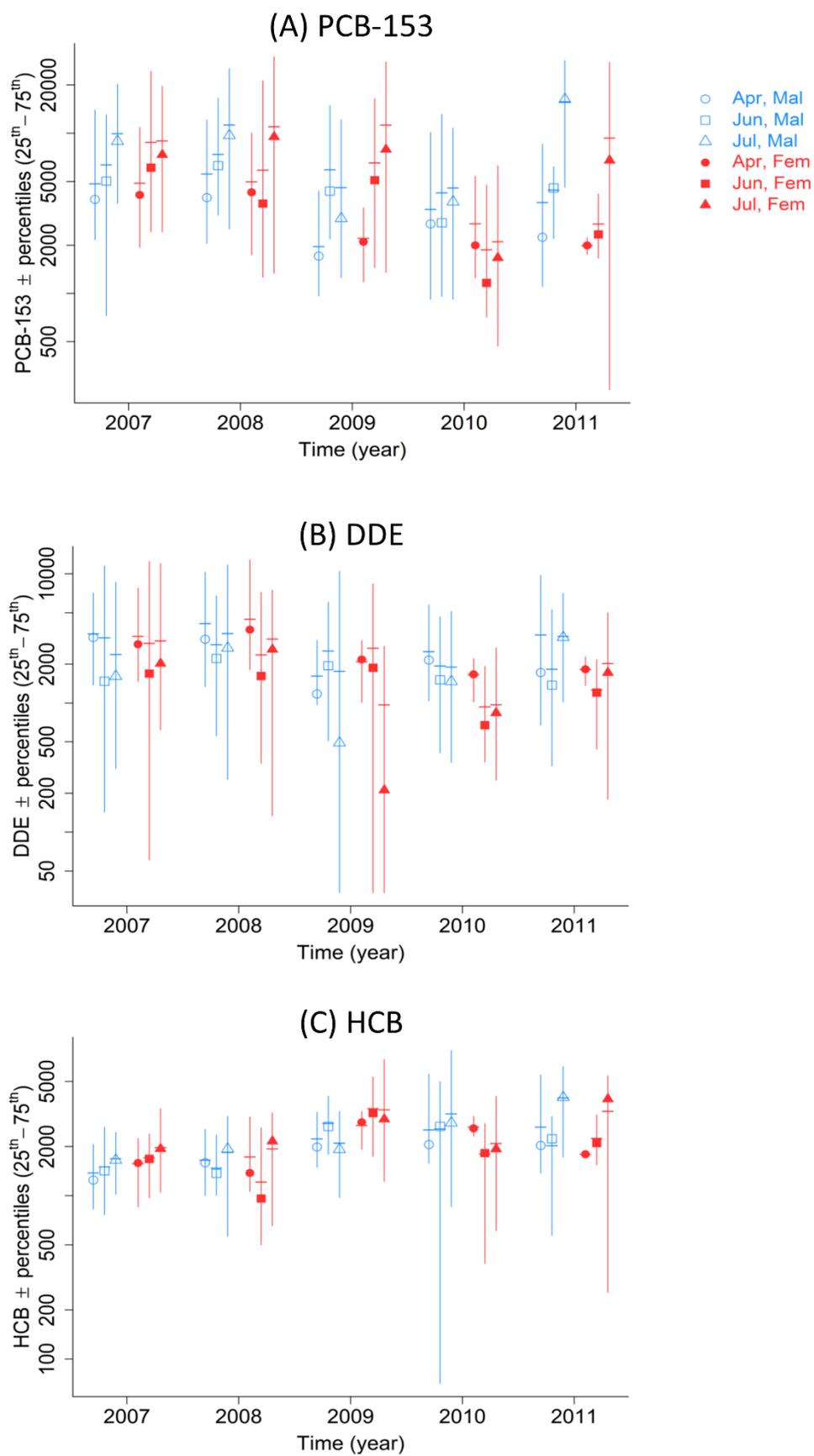
644

645

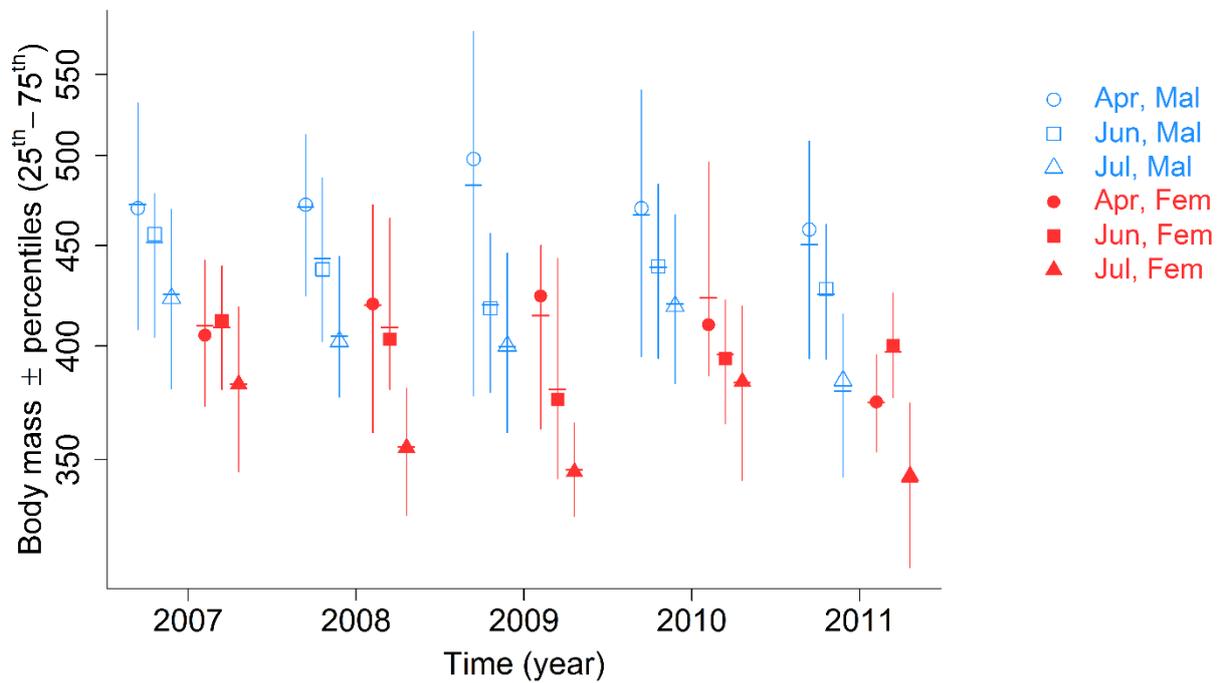
646 Fig.1.



648 Fig. 2



650 **Fig. 3.**



651

652