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## Survival rate and breeding outputs in a high Arctic seabird exposed to legacy persistent organic pollutants and mercury

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### ► To cite this version:

Aurélie Goutte, Christophe Barbraud, Dorte Herzke, Paco Bustamante, Frédéric Angelier, et al.. Survival rate and breeding outputs in a high Arctic seabird exposed to legacy persistent organic pollutants and mercury. *Environmental Pollution*, 2015, 200, pp.1-9. 10.1016/j.envpol.2015.01.033 . hal-01118127

**HAL Id: hal-01118127**

**<https://hal.sorbonne-universite.fr/hal-01118127>**

Submitted on 18 Feb 2015

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1 Survival rate and breeding outputs in a high Arctic seabird exposed to legacy  
2 persistent organic pollutants and mercury

3

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26

27 **Abstract**

28 Chronic exposure to pollutants may represent a threat for wildlife. We tested whether adult  
29 survival rate, breeding probability and breeding success the year of sampling and the  
30 following year were affected by blood levels of mercury or persistent organic pollutants in  
31 Svalbard black-legged kittiwake *Rissa tridactyla*, by using capture–mark–recapture models  
32 over a five-year period. Survival rate was negatively linked to HCB levels in females, to  
33 chlordane mixture and oxychlordane, tended to decrease with increasing PCBs or DDE levels,  
34 but was unrelated to mercury. Breeding probability decreased with increasing mercury levels  
35 during the sampling year and with increasing CHL or HCB levels during the following year,  
36 especially in males observed as breeders. Surprisingly, the probability of raising two chicks  
37 increased with increasing HCB levels. Although levels of these legacy pollutants are expected  
38 to decline, they represent a potential threat for adult survival rate and breeding probability,  
39 possibly affecting kittiwake population dynamics.

40

41 **Capsule abstract:** Negative effects of pollutants were detected on future breeding  
42 probabilities and on adult survival rate in a High Arctic seabird species.

43

44 **Keywords:** heavy metals, kittiwake, population, pesticides, PCBs

45

## 46 **1. Introduction**

47 Contaminants, such as mercury (Hg) and persistent organic pollutants (hereafter  
48 POPs) may represent a threat for wildlife, because of their detrimental effects on  
49 developmental, neurological, physiological, endocrine and immune functions (Barron et al.,  
50 1995; Bustnes et al., 2003a; Tan et al., 2009; Letcher et al., 2010). Despite a growing  
51 environmental concern during the last decades, the demographic consequences of pollution  
52 remain poorly evaluated in free-living vertebrates. Only a few long-term monitoring studies  
53 have addressed the consequences of environmental pollutants on survival rate and long-term  
54 reproductive outputs. Hg or POP levels were negatively related to long-term breeding  
55 probability and success in the wandering albatross *Diomedea exulans* and in two *Catharacta*  
56 *skua* species (Goutte et al., 2014a,b). Apparent survival rate was lower in glaucous gulls  
57 *Larus hyperboreus*, bearing the highest levels of oxychlordan, a metabolite of the chlordane  
58 mixture, which is regarded as one of the most toxic POPs (Erikstad et al., 2013). However,  
59 adult survival rate was not related to POPs or Hg in tree swallows (*Tachycineta bicolor*), king  
60 eiders (*Somateria spectabilis*), white-winged scoters (*Melanitta fusca*), wandering albatrosses  
61 and two *Catharacta skua* species (Wayland et al., 2008; Hallinger et al., 2011; Goutte et al.  
62 2014a,b).

63 Some seabird species appear as ideal models for assessing the demographic  
64 consequences of environmental pollution. Firstly, individual detection probabilities of  
65 seabirds at breeding colonies are generally high because of high overall site fidelity (e.g.  
66 Gauthier et al., 2012). Secondly, large sample sizes and accurate measures of breeding outputs  
67 are relatively easy to obtain in seabird's colonies. Thirdly, these long-lived top predators are  
68 particularly exposed to contaminants, because of bioaccumulation process and  
69 biomagnification along the trophic web (Rowe, 2008; Letcher et al., 2010).

70           The present study focusses on black-legged kittiwakes *Rissa tridactyla* breeding in  
71 Svalbard, a Norwegian archipelago in the north-western part of the Barents Sea. The  
72 Norwegian Arctic is recognized as a final sink for organic and metallic pollutants, which are  
73 transported by atmospheric and oceanic currents and by large rivers (Gabrielsen and  
74 Henriksen, 2001). Previous studies in this population of Svalbard kittiwakes have reported  
75 deleterious effects of Hg and POPs on endocrine mechanisms (Nordstad et al., 2012; Tartu et  
76 al., 2013, 2014). The estimated number of breeding pairs in the Svalbard archipelago is  
77 270 000 in 215 colonies (Strøm, 2006). The status of black-legged kittiwakes is near  
78 threatened, with a pronounced population decline from 1995 to 2002 and a slight increase  
79 from 2002 to 2012 (Barrett et al., 2012). This study aims at detecting whether breeding  
80 probability the year of sampling and demographic traits the following year (apparent adult  
81 survival rate, breeding probability, probability of successfully raising at least one chick and  
82 probability of successfully raising two chicks) were correlated with individual blood levels of  
83 Hg or POPs. According to the few available long-term studies on polar seabird species  
84 (Erikstad et al., 2013; Goutte et al., 2014a,b), we predicted deleterious effects of Hg or POPs  
85 on breeding probability and breeding success during the year of sampling and during the  
86 following year and deleterious effects of the chlordane mixture and metabolites on survival  
87 rate in black legged kittiwakes.

88

## 89 **2. Materials and methods**

### 90 *2.1. Study area and birds*

91           Our study was conducted in a colony of black legged kittiwakes at Kongsfjorden,  
92 Svalbard (78°54'N, 12°13'E), seven kilometers southeast of Ny-Ålesund, Norway. Kittiwakes  
93 are colonial seabirds that breed on cliffs throughout the northern parts of the Pacific and

94 Atlantic, including the Barents Sea region up to the Svalbard Archipelago (Anker-Nilssen et  
95 al., 2000). Kittiwakes were studied in one plot of around 150 pairs breeding on cliff ledges at  
96 heights of 5–10 m. Male and female kittiwakes were sampled once, between 2007 to 2010  
97 years, during the pre-laying stage (arrival, nest building, courtship and mating period) from  
98 23<sup>rd</sup> of April to 16<sup>th</sup> of June. Table 1 summarizes sampling information: a total of 105  
99 kittiwakes were sampled for measurement of Hg and 138 kittiwakes for POPs. We chose to  
100 focus our study on the pre-laying period, because sampling kittiwakes during the incubating  
101 or chick-rearing period would have biased our demographic study towards good-quality birds  
102 (breeders) and would have missed possible effects in non-breeders.

103

## 104 *2.2. Capture and blood sampling*

105 Male and female kittiwakes were caught on the nests with a noose at the end of a 5 m  
106 fishing rod. Blood samples were collected from the alar vein with a 2 ml heparinized syringe  
107 and a 23-gauge needle. Kittiwakes were individually marked with metal rings and PVC  
108 plastic bands engraved with a three-digit code and fixed to the bird's tarsus for identification  
109 from a distance without perturbation.

110

## 111 *2.3. Laboratory analyses*

112 Blood samples were centrifuged. Plasma and red blood cells were separated and stored  
113 at – 20°C. Molecular sexing was performed on red blood cells as detailed in Weimerskirch et  
114 al. (2005). Total Hg was measured at the laboratory Littoral Environnement et Sociétés  
115 (LIENSs) from lyophilized red blood cells with an Advanced Mercury Analyzer  
116 spectrophotometer (Altec AMA 254). At least two aliquots ranging from 5 to 10 mg dry  
117 weight were analyzed for each individual until having a relative standard deviation <5 %. As  
118 described by Bustamante et al. (2006), accuracy was checked using a certified reference

119 material (CRM, Tort-2 Lobster Hepatopancreas, NRC, Canada; certified Hg concentration:  
120  $0.27 \pm 0.06 \mu\text{g g}^{-1}$  dry mass; with recoveries of 98 to 102%). Mass of CRM was adjusted to  
121 represent the same amount of Hg introduced in the AMA compared to that in blood samples.  
122 Blanks were analysed at the beginning of each set of samples and the detection limit of the  
123 method was  $0.005 \mu\text{g g}^{-1}$  dry mass. Mean values of replicates were used in statistical  
124 analyses.

125 POPs were analysed from whole blood samples at the Norwegian Institute for Air  
126 Research (NILU) in Tromsø. The following compounds were analysed: polychlorinated  
127 biphenyl (CB, -99, -118, -138, -153, -180, -183 and -187) hereafter referred as  $\Sigma$  PCBs, p,p'-  
128 DDE (p,p'-dichlorodiphenyldichloroethylene, HCB (hexachlorobenzene), and the chlordane  
129 mixture (trans-chlordane, trans-, cis-nonachlor) and metabolites (oxychlordane), hereafter  
130 referred as CHL. To a blood sample of 0.5 to 1.5 ml, an internal standard solution was added  
131 ( $^{13}\text{C}$ -labelled compounds from Cambridge Isotope Laboratories: Woburn, MA, USA). The  
132 sample was extracted twice with 6 ml of *n*-hexane, after denaturation with ethanol and a  
133 saturated solution of ammonium sulphate in water. Matrix removal on florisil columns,  
134 separation on an Agilent Technology 7890 GC and detection on an Agilent Technology  
135 5975C MSD were performed as described by Herzke et al. (2009). The limit for detection was  
136 threefold the signal-to-noise ratio, and for the compounds investigated the limit ranged from  
137  $0.4$  to  $122 \text{ pg.g}^{-1}$  wet weights (ww). For quality assurance, blanks (clean and empty glass  
138 tubes treated like a sample) were run for every 10 samples similar to standard reference  
139 material (1589 a human serum from NIST). The accuracy of the method was within the 70  
140 and 108% range.

141

142 *2.4. Life history traits*

143 From 2007 to 2012, individuals were individually identified, through PVC plastic  
144 bands reading. Using a mirror at the end of an 8 m fishing rod, we checked the whole plot  
145 (about 120 nests) every two days to monitor breeding status (at least one egg is laid or no egg  
146 laid). Then, we checked the nest content every 2 or 3 days to monitor the number of chicks  
147 that reached at least 12 days of age per nest.

148

## 149 *2.5. Statistical analyses*

150 We used R software (R Development Core Team 2012) and generalized linear models  
151 (GLMs) with normal distribution and a link function to test whether log-transformed Hg,  $\Sigma$   
152 PCBs, DDE, HCB or CHL levels were linked to sex, year and the interaction sex  $\times$  year.  
153 GLMs with binomial error distribution and a logit link function were then used to test whether  
154 breeding probability (will breed or will skip) the year of sampling was linked to pre-laying  
155 Hg,  $\Sigma$  PCBs, DDE, HCB or CHL levels.

156

## 157 *2.6. Estimating the effect of Hg and POPs on demographic parameters*

158 The effects of Hg and POPs concentrations on the demographic parameters were  
159 evaluated through the capture-recapture data of sampled kittiwakes. A MSMR (Multi-State  
160 Mark Recapture, Lebreton and Pradel, 2002) model was constructed by distinguishing five  
161 states: non-breeder (NB, defined as an individual that was not observed with an egg), failed  
162 breeder (FB, defined as an individual that was observed with one or two eggs, or one or two  
163 chicks but that failed to raise a chick), successful breeder with one chick (SB1, defined as an  
164 individual that raised one chick), successful breeder with two chicks (SB2, defined as an  
165 individual that raised two chicks), and dead. The state dead ( $\dagger$ ) was an absorbing state  
166 representing death or permanent emigration from the study area. Kittiwakes that were ringed  
167 and observed the years before sampling for Hg or POPs were considered as non-observed, in

168 order to test the effect of contaminants (at year  $t$ ) on future (year  $t+1$ ) survival and breeding  
169 performances. Models were parameterized in terms of the probability of survival ( $S$ ), the  
170 probability of breeding ( $\beta$ ), the probability of breeding successfully ( $\gamma$ ), the probability of  
171 successfully raising two chicks ( $\delta$ ), and the detection probability ( $p$ ). Transition probabilities  
172 between states were thus modeled with a four-step procedure where  $S$ ,  $\beta$ ,  $\gamma$  and  $\delta$  were  
173 considered as four successive steps in transition matrices. Figure 1 presents a multinomial tree  
174 diagram describing the probability structure for multistate observations, and parameters of the  
175 model are defined in Table 2. We chose a MSMR approach since this allows taking into  
176 account the probability of detecting individuals given their return to the study sites. It also  
177 allows taking into account the previous breeding state of individuals which might be  
178 important to obtain unbiased estimates of demographic parameters (Lebreton and Pradel  
179 2002).

180 Several constraints were made to ensure that the parameters of the model were  
181 estimable. The state “dead” being explicitly included in the model but being never  
182 encountered, transition probabilities from the state dead were fixed to 0 and capture  
183 probability was fixed to 0 (Pradel 2005, Choquet et al. 2009a). Because our capture-recapture  
184 analyses relied on a limited number of individual capture histories, parameters  $S$ ,  $\beta$ ,  $\gamma$ ,  $\delta$  and  $p$   
185 were constrained to be constant over time but state and sex dependent. With this constraint the  
186 initial model was full-rank. Note that we ran a model where all demographic parameters were  
187 time, sex and state dependent but this model was highly rank deficient.

188 This MSMR model was parameterized by the survival–transition probabilities matrix:

189

	NB	FB	SB1	SB2	†
NB	$S(1 - \beta)$	$S\beta(1 - \gamma)$	$S\beta\gamma(1 - \delta)$	$S\beta\gamma\delta$	*
FB	$S(1 - \beta)$	$S\beta(1 - \gamma)$	$S\beta\gamma(1 - \delta)$	$S\beta\gamma\delta$	*
SB1	$S(1 - \beta)$	$S\beta(1 - \gamma)$	$S\beta\gamma(1 - \delta)$	$S\beta\gamma\delta$	*
SB2	$S(1 - \beta)$	$S\beta(1 - \gamma)$	$S\beta\gamma(1 - \delta)$	$S\beta\gamma\delta$	*
†	–	–	–	–	*

190

191 Because we were interested to test for sex-specific effects of Hg and POPs on  
192 demographic parameters we started from an initial model including an effect of sex ( $g$ ) on  
193 each parameter. Model selection was first performed on detection probability by testing state-  
194 dependency (difference between all states, between breeders and non-breeders, or no  
195 difference). We then tested for sex difference and state-dependency (difference between all  
196 states, difference between breeders and non-breeders or no difference) for  $S$ ,  $\beta$ ,  $\gamma$  and  $\delta$ . We  
197 tested for an effect of Hg,  $\Sigma$  PCBs, DDE, HCB, or CHL on demographic parameters the  
198 following year to test the hypothesis that contamination levels in one breeding season may  
199 influence the survival and breeding success of an individual in the following season. We built  
200 MSMR models where each demographic parameter  $\theta$  was modeled as a function of  
201 contaminant  $C$  using a logit link function:  $\text{logit}(\theta) = a + b \times C_i$ , where  $a$  is an intercept,  $b$  is a  
202 slope and  $C_i$  is Hg or POPs concentration for individual  $i$ . The 95% confidence interval (CI)  
203 of the slope parameters  $b$  was used, as well as Akaike's Information Criterion corrected for  
204 small sample size (AICc, Burnham and Anderson, 2002) for inference. We considered an  
205 effect of contaminant as statistically supported when 0 was outside the 95% CI of the mean of  
206 the slope of the relationship (Grosbois et al., 2008). When  $b < 0$ , or  $b > 0$ , the covariate  $C$  has  
207 a negative or positive effect on the demographic parameter, respectively. We tested the  
208 goodness-of-fit (GOF) of the time dependent MSMR model using U-CARE (Choquet et al.  
209 2009b). All models were run under program E-SU RGE 1.8.5 allowing splitting transition  
210 probabilities between states (Choquet et al. 2009a).

211

### 212 **3. Results**

213

#### 214 *3.1 Associations between Hg or POPs and breeding probability in year of blood sampling*

215 Table 1 summarizes the values of Hg,  $\sum$  PCBs, DDE, HCB and CHL in males and  
216 females. Appendix 1 gives the concentrations of each POP congener and appendix 2 presents  
217 the relationships between levels of Hg,  $\sum$  PCBs, DDE, HCB and CHL.

218 Hg levels were significantly higher in males than in females ( $F_{1,103} = 3.993$ ,  $p =$   
219  $0.048$ ), but did not differ between the two sampling years (year:  $F_{1,102} = 3.339$ ,  $p = 0.071$ ; sex  
220  $\times$  year:  $F_{1,101} = 1.102$ ,  $p = 0.296$ ). Breeding probability during the sampling year was  
221 influenced by Hg levels ( $df = 103$ ,  $\chi^2 = 12.983$ ,  $p < 0.001$ ): kittiwakes that would skip (mean  
222  $\pm$  SD:  $2.284 \pm 0.417 \mu\text{g}\cdot\text{g}^{-1}$ ) had higher pre-laying Hg levels than kittiwakes that would breed  
223 ( $1.962 \pm 0.470 \mu\text{g}\cdot\text{g}^{-1}$ ).

224 Levels of  $\sum$  PCBs, DDE, HCB, or CHL did not differ between males and females  
225 (sex:  $p > 0.07$  for all tests: sex  $\times$  year:  $p > 0.09$  for all tests). Levels of  $\sum$  PCBs ( $F_{3,134} = 4.935$ ,  
226  $p = 0.003$ ), HCB ( $F_{3,134} = 37.035$ ,  $p < 0.001$ ),  $\sum$  CHL ( $F_{3,134} = 12.818$ ,  $p < 0.001$ ), but not  
227 DDE ( $F_{3,134} = 2.519$ ,  $p = 0.061$ ) differed among years. Breeding probability was not  
228 influenced by levels of  $\sum$  PCBs, DDE, HCB, or CHL during the sampling year ( $p > 0.61$  for  
229 all tests).

230

### 231 *3.2. Associations between Hg and demographic parameters in year after blood sampling*

232 The GOF of the MSMR model was overall not significant (males:  $\chi^2 = 48.913$ ,  $df = 69$ ,  
233  $p = 0.968$  and females:  $\chi^2 = 47.435$ ,  $df = 71$ ,  $p = 0.986$ ). The best model according to AICc  
234 (model 16, Appendix 3) indicated that breeders in the previous year had higher breeding  
235 probabilities and detection probabilities than non-breeders in the previous year. However  
236 birds captured as breeders or non-breeders did not differ in survival rate, probabilities of  
237 successfully raising one or two chicks (Appendix 3 and Table 3). Demographic parameters  
238 did not differ between males and females (Appendix 3 and Table 3).

239 Model selection and slope estimates suggested no effect of Hg on demographic  
240 parameters. Model Hg3 had a  $\Delta\text{AICc}$  lower than 2 compared to the null model, but the effect  
241 of Hg on breeding probability the following year was not supported, since the 95% CI of the  
242 slope parameter included 0 (Table 4).

243

### 244 *3.3. Associations between POPs and demographic parameters in year after blood sampling*

245 Model selection was based on  $\Delta\text{AICc}$  higher than 2 compared to the intercept model  
246 and the 95% CI of the slope of the relationship that did not include zero. Hence, in spite of  
247 good AICc, several models suggesting an effect of  $\Sigma$  PCBs, DDE, HCB or CHL on  
248 demographic parameters were not retained. Only six models met these requirements (Table  
249 5). Models HCB5 and HCB6 suggested a negative effect of HCB on breeding probability the  
250 following year for individuals and especially males observed as breeders (Fig.2A, 2B).  
251 Model CHL6 suggested a negative effect of CHL on breeding probability the following year  
252 for males observed as breeders (Fig. 2C). Model HCB1 suggested a positive effect of HCB on  
253 the probability of successfully raising two chicks the following year (Fig. 3). Model HCB8  
254 suggested a negative effect of HCB on survival rate of females (Fig. 4A). Model CHL7  
255 suggested a negative effect of CHL on survival rate (Fig. 4B). We could also notice a  
256 tendency towards a negative effect between survival rates and levels of  $\Sigma$  PCBs (model  
257 PCB7,  $\Delta\text{AICc} = 1.24$ , mean slope and 95% CI = -0.44 [-0.82 ; -0.03]), DDE (Model: DDE7,  
258  $\Delta\text{AICc} = 0.88$ , slope = -0.42 [-0.82 ; -0.01]), HCB for males and females (Model HCB7,  
259  $\Delta\text{AICc} = 1.73$ , slope = -0.47 [-0.88 ; -0.06]), or CHL for females only (Model CHL8,  $\Delta\text{AICc}$   
260 = 1.50, slope = -0.73 [-1.29 ; -0.17]).

261

## 262 **4. Discussion**

263 Using a long-term data set and MSMR models, this study explores the demographic  
264 effects of Hg or families of legacy POPs (7 PCB congeners, p-p' DDE, HCB, and the  
265 chlordane mixture and metabolites (trans-chlordane, trans-, cis-nonachlor, oxychlordane)) in a  
266 free-living Arctic seabird species. It should be noticed that differences in toxicity among POP  
267 congeners were not taken into account in these analyses, because toxic equivalent factors  
268 (TEFs) were only available for PCB-105 and PCB-118. Moreover interactions among families  
269 of pollutants may occur within an organism to induce synergistic effects, but they are difficult  
270 to demonstrate within a field study.

271

#### 272 *4.1. Survival and contaminants*

273 Estimated demographic parameters were similar to those previously estimated in other  
274 populations of black legged kittiwakes (Frederiksen et al., 2005). Adult survival rate in this  
275 study (85% [82 – 88%]) was within the range of estimated survival rates in north Atlantic  
276 populations (80-92%, Danchin and Monnat, 1992; Erikstad et al., 1995; Oro and Furness,  
277 2002; Frederiksen et al., 2005).

278 The adult survival rate of kittiwakes was not jeopardized by Hg, which corroborates  
279 most of the previous studies in free-living birds (Wayland et al., 2008; Hallinger et al., 2011;  
280 Goutte et al., 2014a,b). Apparent survival rate was negatively linked to HCB levels in  
281 females, to mixture of chlordane and oxychlordane, and tended to be negatively correlated  
282 with  $\sum$  PCBs or DDE levels. Only one study (Erikstad et al. 2013) highlighted a negative  
283 effect of oxychlordane on adult survival rate in the glaucous gull breeding in the Bjørnøya  
284 Island (blood levels of oxychlordane: 1.3 to 128.8 ng.g<sup>-1</sup> wet weight, median: 13.2 ng.g<sup>-1</sup> ww)  
285 and this effect was the most pronounced among the most contaminated females. Even if  
286 kittiwakes were more than 10-time less contaminated than glaucous gull (blood levels of  
287 oxychlordane: 0.007 to 6.0 ng.g<sup>-1</sup> wet weight), this study reveals that high levels of the

288 chlordanes mixture and metabolites or HCB could negatively affect adult survival rate, and  
289 especially in female kittiwakes.

290 The correlation between POP levels and survival rate could be a by-product of age-  
291 dependent mechanisms, with older kittiwakes having the highest POP burden and the lowest  
292 survival probability. Age of kittiwakes was unknown in this study and we could not control  
293 for age. However, blood levels of PCB-153, p,p'-DDE, HCB, and oxychlordanes were  
294 unrelated to age in glaucous gulls (Bustnes et al., 2003b). Similarly, blood levels of PCBs or  
295 organochlorine pesticides (HCB, lindane, chlordanes mixture, mirex, DDT and metabolites)  
296 were unrelated to age in wandering albatrosses (Carravieri et al., 2014). Therefore, it seems  
297 unlikely that age was a confounding factor in the correlation between POP levels and survival  
298 rate. In addition, as we did not monitor long-distance dispersal, our findings on apparent  
299 survival rate could also include the effects of POPs on long-term emigration of the most  
300 polluted birds.

301 This study suggests that HCB or the chlordanes mixture and metabolites may weaken  
302 the general health of kittiwakes and may increase their vulnerability to harsh environmental  
303 pressures in the Arctic (Letcher et al., 2010). In that context, it is conceivable that the effect  
304 of POPs on survival rate is only detected during harsh environmental events. Because our  
305 sample size did not allow taking into account an effect of years, we could not have tested  
306 whether harsh environmental conditions during a specific year would exacerbate the effects of  
307 pollutants on demographic parameters the following year.

308

#### 309 *4.2. Long-term fecundity and contaminants*

310 A previous study on this population of kittiwakes has highlighted that total blood Hg  
311 load during the pre-laying period predicted the likelihood of breeding, with non-breeders  
312 having higher Hg levels than breeders, but not the timing of breeding, clutch size, and

313 breeding success (Tartu et al., 2013). Moreover experimentally elevated Hg levels (total Hg in  
314 blood, mean  $\pm$  SD: from  $0.73 \pm 0.09$  to  $3.95 \pm 0.68$  mg.kg<sup>-1</sup> fresh weight) led to an altered  
315 pairing behaviour in white ibises *Eudocimus albus* (Frederick and Jayasena, 2011). In the  
316 present study, Hg levels were higher in kittiwakes that would skip breeding than in birds that  
317 would breed, as previously shown (Tartu et al., 2013). Hg levels did not affect breeding  
318 probability and breeding success the following year, which differed from previous studies in  
319 the south polar skua *Catharacta maccormicki* (Hg levels in blood: mean  $\pm$  SE:  $2.15 \pm 0.17$   
320  $\mu\text{g.g}^{-1}$  dry mass), in the brown skua *C. lonnbergi* ( $8.22 \pm 0.24$   $\mu\text{g.g}^{-1}$  dry mass) and in the  
321 wandering albatross ( $7.7 \pm 3.6$   $\mu\text{g.g}^{-1}$  dry mass) (Goutte et al., 2014 a,b). However, Hg levels  
322 in these species were measured during the incubation and the chick-rearing period, while Hg  
323 levels in the present study were measured in pre-laying kittiwakes. Furthermore, breeding  
324 success was monitored on chicks that reached at least 12 days of age and did not allow testing  
325 an effect of contaminants on late developmental stage.

326 POPs burden did not influence the breeding probability the year of sampling, which  
327 was consistent with a previous study on the same population of kittiwakes (Tartu et al. 2014).  
328 Breeding probability the following years was reduced by high HCB levels in breeders and  
329 especially in males, or by high levels of the chlordanes mixture and metabolites in male  
330 breeders. A negative correlation between POP levels and breeding probabilities the following  
331 year has been highlighted in the wandering albatross (Goutte et al., 2014b). Male breeders  
332 seemed to be the most sensitive to POPs. Energetic and time-dependent costs of reproduction  
333 have been shown to induce downstream consequences on reproductive investment during the  
334 following breeding season (carry over effect, Catry et al., 2013). One may suggest that POPs  
335 burden may intensify these carry over effects, but studies are needed to either rebut or confirm  
336 this hypothesis.

337           Levels of  $\Sigma$  PCB, DDE, HCB, and the chlordane mixture and metabolites did not  
338 influence the probability of successfully raising one chick the following year, which was  
339 consistent with a previous study on the same population of kittiwakes and during the year of  
340 sampling (Tartu et al., 2014). We detected a positive relationship between the probability of  
341 successfully raising two chicks the following year and HCB levels, but not PCBs, DDE or the  
342 chlordane mixture. This positive relationship between HCB and breeding performance  
343 appears surprising, as contaminants are believed to induce deleterious effects on reproductive  
344 traits. Previous studies have pointed out that female kittiwakes and gulls with higher levels of  
345 organochlorine pesticides laid their eggs earlier in the season (Bustnes et al., 2008; Tartu et  
346 al., 2014). As laying early is related to high breeding success (Lack, 1968), this could explain  
347 the positive relationship between HCB and the probability of successfully raising two chicks.  
348 In another hand, this relationship may not be causal and may be enhanced by confounding  
349 factors: for instance, kittiwakes succeeding in raising two chicks may be of higher quality,  
350 rely on higher trophic level organisms and hence be more exposed to pollutant.

351

352           It appears that some families of POPs may be more prone to trigger damaging effects  
353 the following year. Specifically, high levels of HCB or the chlordane mixture and metabolites  
354 were correlated to lower survival rate and lower probability to breed the following year. These  
355 findings corroborate a previous study: despite their lower concentrations, HCB and  
356 oxychlordane tended to be more often related to adverse effects than PCB and DDE in  
357 glaucous gull (Bustnes, 2006). Although levels of these “legacy” POPs are expected to  
358 decline, as shown in Canadian Arctic seabirds from the 1970s to the late 1990s (Braune et al.,  
359 2005), they appear to represent a potential threat for adult survival rate and thus for  
360 population dynamics.

361

362 **Acknowledgments**

363 The study was funded by the Institut Paul-Émile Victor (IPEV Programme 330,  
364 O Chastel), Agence Nationale de la Recherche (ANR PolarTop, O. Chastel), COPOL (G.W.  
365 Gabrielsen & J.O. Bustnes) and AVITOX (J. O. Bustnes). This study was approved by the  
366 French and Norwegian Ethic committees and by the Governor of Svalbard. The authors thank  
367 the numerous fieldworkers who helped with blood sampling and ring-reading: A. Lendvai, E.  
368 Noreen, T. Nordstad, K. Sagerup, S.A. Hanssen, C. Trouvé, and J. Welcker. At the LIENSs,  
369 the authors thank M. Brault-Favrou from the Plateforme Analyses Elementaires for her  
370 excellent technical assistance in laboratory analyses. AMA was funded by the CPER (Contrat  
371 de Projet Etat Région).

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509

510 Table 1: Levels (mean  $\pm$  SD) of  $\Sigma$  PCBs (CB, -99, -118, -138, -153, -180, -183 and -187),  
 511 p,p'-DDE, HCB, CHL (transchlordan, trans-, cis-nonachlor, oxychlordan,) and Hg  
 512 (mercury) in blood of male and female kittiwakes sampled during the pre-laying period.

	Year	Males	Females
$\Sigma$ PCBs (pg.g <sup>-1</sup> ww)	2007	14700 $\pm$ 9630	12640 $\pm$ 6421
	2008	14896 $\pm$ 11029	13399 $\pm$ 9197
	2009	9282 $\pm$ 7915	10375 $\pm$ 4705
	2010	12786 $\pm$ 10966	21168 $\pm$ 14390
DDE (pg.g <sup>-1</sup> ww)	2007	3622 $\pm$ 1730	3152 $\pm$ 1422
	2008	4025 $\pm$ 2642	4189 $\pm$ 3490
	2009	2618 $\pm$ 1660	2184 $\pm$ 890
	2010	3249 $\pm$ 2739	4725 $\pm$ 3584
HCB (pg.g <sup>-1</sup> ww)	2007	1616 $\pm$ 966	1600 $\pm$ 407
	2008	1616 $\pm$ 444	1691 $\pm$ 697
	2009	2416 $\pm$ 1493	2699 $\pm$ 451
	2010	2670 $\pm$ 877	3487 $\pm$ 1288
CHL (pg.g <sup>-1</sup> ww)	2007	1352 $\pm$ 782	1329 $\pm$ 508
	2008	1237 $\pm$ 510	1275 $\pm$ 765
	2009	1344 $\pm$ 1155	1353 $\pm$ 403
	2010	1766 $\pm$ 650	2482 $\pm$ 1602
Hg ( $\mu$ g.g <sup>-1</sup> dw)	2008	2.06 $\pm$ 0.44	1.97 $\pm$ 0.44
	2009	2.33 $\pm$ 0.55	2.01 $\pm$ 0.41

513

514

515 Table 2 Definition of parameters used in the multistate mark–recapture model

Parameter	Definition
$S_s^t$	Probability that an individual in state $s$ at time $t$ survives to time $t + 1$ and does not permanently emigrate from the study area
$\beta_s^t$	Probability that an individual in state $s$ at time $t$ breeds at time $t + 1$ given that it survives to $t + 1$
$\gamma_s^t$	Probability that an individual in state $s$ at time $t$ breeds successfully at time $t + 1$ given that it survives to and breeds at time $t + 1$
$\delta_s^t$	Probability that an individual in state $s$ at time $t$ raises successfully two chicks at time $t + 1$ given that it survives to and breeds successfully at time $t + 1$
$p_s^t$	Probability that an individual in state $s$ at time $t$ is encountered at time $t + 1$

516

517 Table 3: Estimation of parameters (mean and CI) calculated from the best model (model 16,  
 518 Appendix 3) for breeders and non-breeders.

	Non-breeders	Breeders
$S$ : apparent survival rate (%)	85 [82 ; 88]	85 [82 ; 88]
$\beta$ : Breeding probability (%)	47 [41 ; 53]	82 [78 ; 86]
$\gamma$ : Breeding success (%)	75 [71 ; 79]	75 [71 ; 79]
$\delta$ : Probability of raising 2 chicks (%)	40 [35; 45]	40 [35; 45]
$p$ : Detection probability (%)	78 [67 ; 85]	98 [90 ; 99]

519

520 Table 4: Modeling the effects of Hg levels and sex on demographic parameters of *Rissa*  
 521 *tridactyla* (N = 105). Models are arranged from lowest to highest  $\Delta\text{AICc}$ . The estimated slope  
 522 and 95% confidence intervals (CI) are given for the model (Hg3) that has a  
 523 lower AICc than the intercept model.  
 524

Hypothesis	# Model	Rank	Deviance	$\Delta\text{AICc}$	Slope	95% CI
Effect of Hg on $\gamma$	Hg3	12	1194.84	0	0.29	-0.84 ; 1.43 #
Intercept model	Hg0	10	1201.22	2.10		
Effect of Hg on $\delta$	Hg1	12	1197.40	2.56		
Effect of Hg and sex on $\gamma$	Hg4	14	1193.67	3.18		
Effect of Hg and sex on $\delta$	Hg2	14	1194.82	4.33		
Effect of Hg on $S$	Hg7	12	1201.11	6.28		
Effect of Hg on $\beta$	Hg5	14	1197.90	7.41		
Effect of Hg and sex on $\beta$	Hg6	18	1190.66	9.03		
Effect of Hg and sex on $S$	Hg8	14	1200.50	10.01		

525 # This effect is not supported because the 95% confidence intervals of the mean of the slope  
 526 of the relationship included zero.

527 Table 5: Modeling the effects of  $\Sigma$  PCBs, p,p'-DDE, HCB and CHL levels and sex on  
528 demographic parameters of *Rissa tridactyla* (N = 138). Models are arranged from lowest to  
529 highest  $\Delta$ AICc. The estimated slopes and 95% confidence intervals (CI) are given for models  
530 that have a lower AICc than the intercept model (NB: non-breeders, B: breeders).

531

Hypothesis	# Model	Rank	Deviance	$\Delta$ AICc	Slope	95% CI	
Effect of $\Sigma$ PCBs on $\beta$	PCB5	14	1351.03	0	NB : -0.62	-1.48 ; 0.23	#
					B : -0.14	-0.82 ; 0.53	#
Effect of $\Sigma$ PCBs and sex on $\beta$	PCB6	18	1344.22	1.90	Male NB : -0.36	-1.50 ; 0.78	#
					Male B : -1.10	-2.43 ; 0.22	#
					Female NB : -0.87	-2.21 ; 0.46	#
					Female B : 0,83	-0.38 ; 2.06	#
Effect of $\Sigma$ PCBs on $S$	PCB7	12	1366.07	10.75	-0.44	-0.82 ; -0.03	##
Effect of $\Sigma$ PCBs on $\delta$	PCB1	12	1367.05	11.74	0.47	-0,36 ; 1,31	###
Intercept model	PCB0	10	1371.55	11.99			
Effect of $\Sigma$ PCBs and sex on $\delta$	PCB2	14	1364.06	13.03			
Effect of $\Sigma$ PCBs and sex on $S$	PCB8	14	1364.33	13.30			
Effect of $\Sigma$ PCBs on $\gamma$	PCB3	12	1368.92	13.61			
Effect of $\Sigma$ PCBs and sex on $\gamma$	PCB4	14	1368.69	17.66			
Effect of DDE and sex on $\beta$	DDE6	18	1339.67	0	Male NB : -0.26	-1.78 ; 1.26	#
					Male B : -1.17	-2.44 ; 0.10	#
					Female NB : -1.82	-3.79 ; 0.14	#
					Female B : 0.69	-0.64 ; 2.01	#
Effect of DDE on $\beta$	DDE5	14	1349.00	0.61	NB : -1.00	-2.08 ; 0.08	#
					B : -0.14	-0.70 ; 0.42	#
Effect of DDE on $S$	DDE7	12	1366.43	13.76	-0.42	-0.82 ; -0.01	##
Intercept model	DDE0	10	1371.55	14.64			
Effect of DDE on $\delta$	DDE1	12	1367.91	15.24			
Effect of DDE on $\gamma$	DDE3	12	1368.85	16.18			
Effect of DDE and sex on $S$	DDE8	14	1365.94	17.56			
Effect of DDE and sex on $\delta$	DDE2	14	1366.73	18.35			
Effect of DDE and sex on $\gamma$	DDE4	14	1368.19	19.80			
Effect of HCB and sex on $\beta$	HCB6	18	1339.36	0	Male NB : -1.50	-4.24 ; 1.25	#
					Male B : -1.86	-3.38 ; -0.34	#
					Female NB : -0.02	-0.87 ; 0.92	#
					Female B : 0.08	-0.74 ; 0.90	#

Effect of HCB on $\beta$	HCB5	14	1349.44	1.37	NB : -0.28 B : -0.53	-1.06 ; 0.50 -1.04 ; -0.01	#
Effect of HCB and sex on $\delta$	HCB2	14	1357.05	8.98	NB : -0.18 B : -2.27	-0.57 ; 0.21 -0.15 ; 4.69	# #
Effect of HCB on $\delta$	HCB1	12	1362.22	9.86	0.94	0.10 ; 1.79	
Effect of HCB and sex on $S$	HCB8	14	1360.54	12.47	Male : 0.41 Female : -0.82	-0.75 ; 1.57 -1.39 ; -0.25	#
Effect of HCB on $S$	HCB7	12	1365.58	13.22	-0.47	-0.88 ; -0.06	##
Intercept model	HCB0	10	1371.55	14.95			
Effect of HCB on $\gamma$	HCB3	12	1369.19	16.83			
Effect of HCB and sex on $\gamma$	HCB4	14	1367.08	19.01			
Effect of CHL and sex on $\beta$	CHL6	18	1338.80	0	Male NB : -0.59 Male B : -2.64 Female NB : -0.73 Female B : -0.07	-2.95 ; 1.77 -5.09 ; -0.18 -1.98 ; 0.51 -0.85 ; 0.70	# # #
Effect of CHL on $\beta$	CHL5	14	1347.61	0.10	NB : -0.73 B : -0.59	-1.81 ; 0.34 -1.20 ; 0.01	# #
Effect of CHL on $S$	CHL7	12	1363.00	11.20	-0.57	-1.00 ; -0.13	
Effect of CHL and sex on $\delta$	CHL2	14	1359.59	12.08	NB : 1.46 B : 1.85	-1.23 ; 4.15 -0.23 ; 3.93	# #
Effect of CHL on $\delta$	CHL1	12	1363.83	12.03	1.05	-0.15 ; 2.24	#
Effect of CHL and sex on $S$	CHL8	14	1361.52	14.01	Male : -0.04 Female : -0.73	-1.12 ; 1.04 -1.29 ; -0.17	### ##
Intercept model	CHL0	10	1371.55	15.51			
Effect of CHL on $\gamma$	CHL3	12	1368.41	16.61			
Effect of CHL and sex on $\gamma$	CHL4	14	1368.38	20.87			

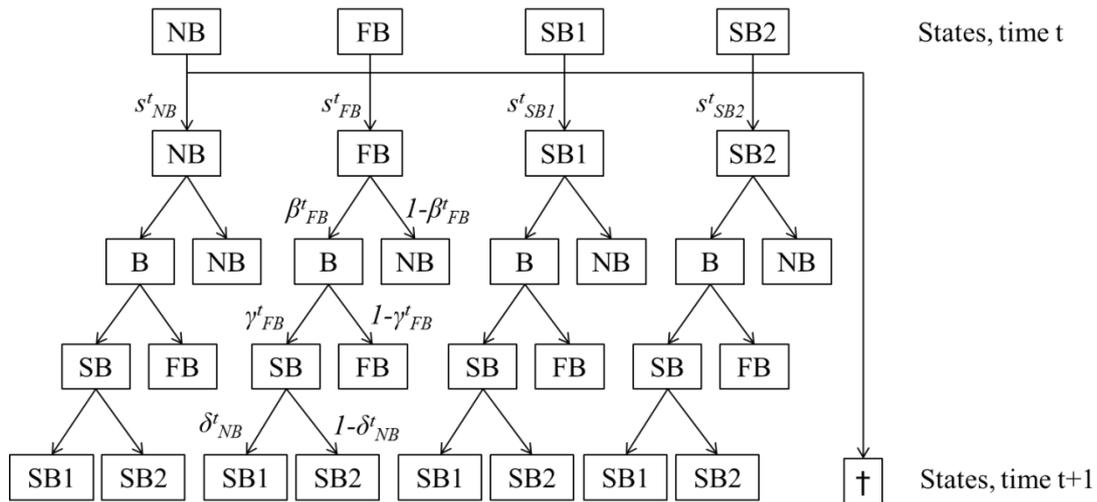
# This effect is not supported, because the 95% CI of the mean of the slope of the relationship included zero.

## This effect is not supported, because the model has a  $\Delta AICc < 2$  compared to the intercept model

532 ### This effect is not supported, because the 95% CI of the mean of the slope of the relationship  
533 included zero and the model has a  $\Delta AICc < 2$  compared to the intercept model.

534 Figure 1: A multinomial tree diagram describing the probability structure for multistate  
 535 observations. Solid boxes indicate the states alive in state NB (non-breeder), FB (failed  
 536 breeder), SB1 (successful breeder with one chick), SB2 (successful breeder with two chicks).  
 537 dead. State transition probabilities were decomposed in a four-step process. The state  
 538 transitions ( $S, \beta, \gamma, \delta$ ) are defined in Table 2 and states in the Methods section.

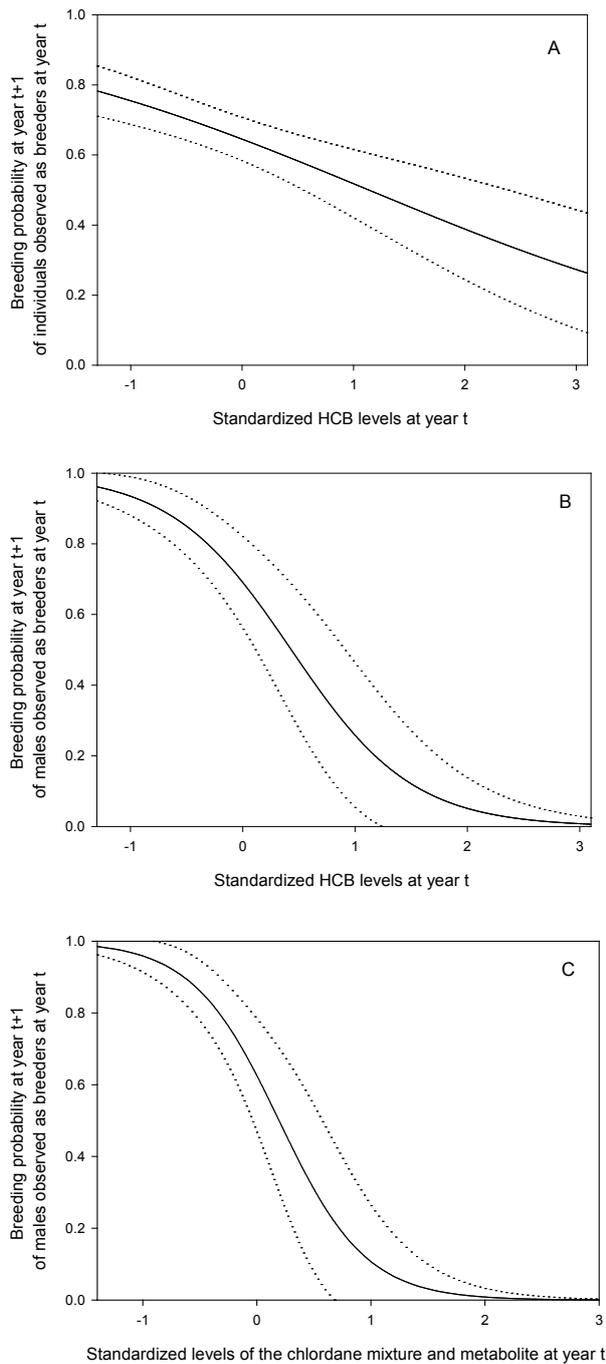
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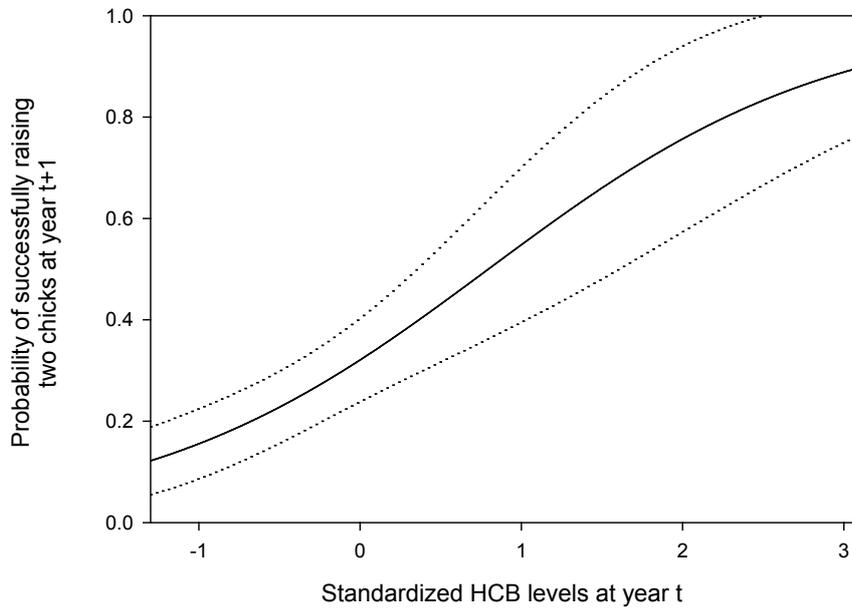
541

542 Figure 2: Relationship between breeding probability at year t+1 in black-legged kittiwakes  
543 and (A) standardized HCB levels in individuals observed as breeders at year t, (B)  
544 standardized HCB levels in males observed as breeders at year t and (C) standardized levels  
545 of the chlordane mixture and metabolites in males observed as breeders at year t . Dotted lines  
546 represent 95% CI.



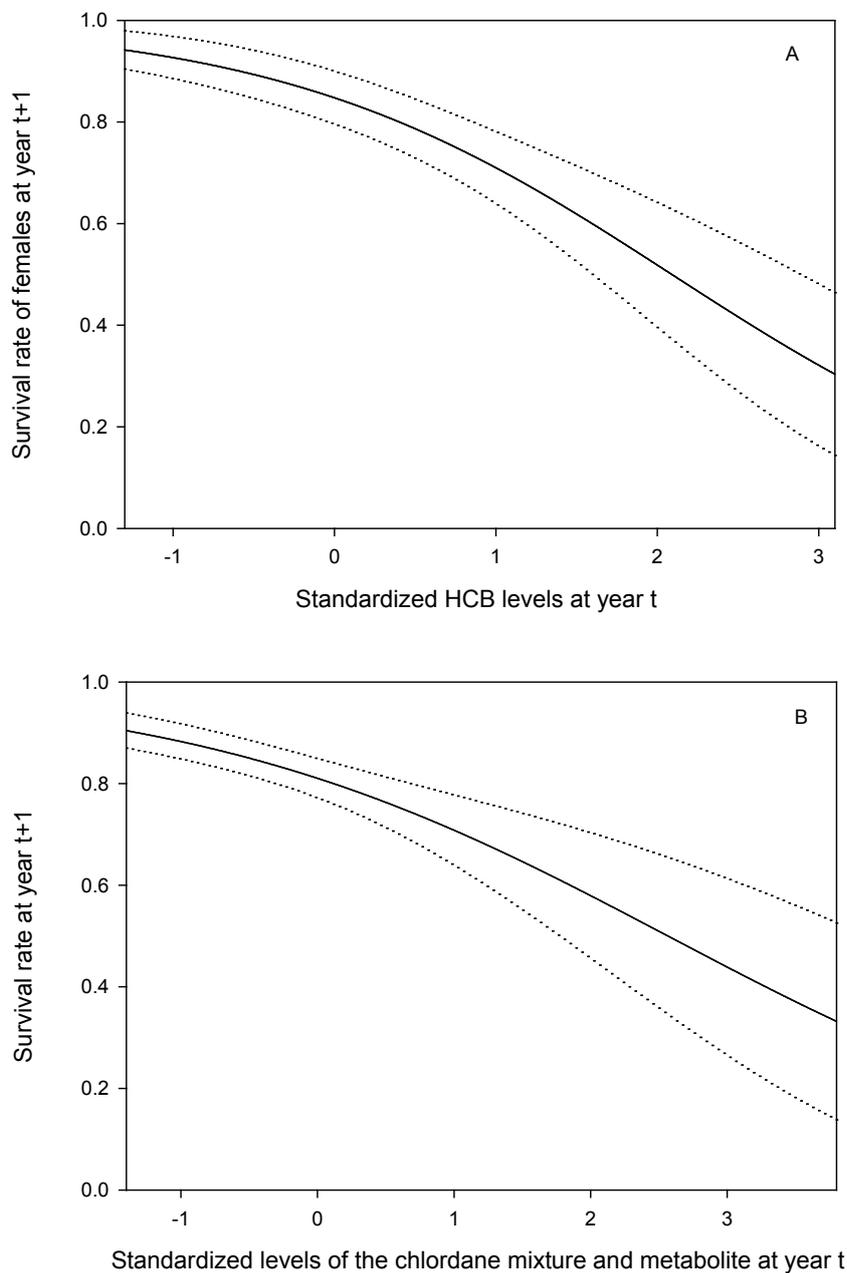
547

548 Figure 3: Relationship between probability of successfully raising two chicks at year t+1 in  
549 black-legged kittiwakes and standardized HCB levels at year t.



550

551 Figure 4: Relationship between survival rate at year t+1 in black-legged kittiwakes and (A)  
552 standardized HCB levels in females at year t, (B) standardized levels of the chlordane mixture  
553 and metabolites at year t.



554

555 Appendix 1: Concentrations (mean, median and standard deviation SD, in pg.g-1 ww)  
556 measured for each POP congener in 138 black-legged kittiwakes during the pre-laying period.  
557

Congener	Mean	Median	SD
PCB-99	1069	800	869
PCB-118	1638	1313	1217
PCB-138	4104	2856	3463
PCB-153	5498	4159	3979
PCB-180	1888	1513	1832
PCB-183	537	354	591
PCB-187	932	680	867
p,p' DDE	3892	2978	2900
HCB	2418	2026	1262
translordane	322	217	346
oxychlorane	1002	818	786
cisnonachlor	173	158	100
transnonachlor	216	196	142

558

559 Appendix 2: Relationships between Hg,  $\Sigma$  PCBs, DDE, HCB, and CHL levels

560

561

	$\Sigma$ PCB	DDE	HCB	CHL
DDE	$F_{1,136} = 180, p < 0.001$			
HCB	$F_{1,136} = 63, p < 0.001$	$F_{1,136} = 41, p < 0.001$		
CHL	$F_{1,136} = 129, p < 0.001$	$F_{1,136} = 104, p < 0.001$	$F_{1,136} = 201, p < 0.001$	
Hg	$F_{1,35} < 0.01 \quad p = 0.983$	$F_{1,35} = 0.04 \quad p = 0.839$	$F_{1,35} = 2.77 \quad p = 0.105$	$F_{1,35} = 1.33 \quad p = 0.136$

562

563

564 Appendix 3:

565 Initial model (Model1) considers sex-difference and state-difference on  $S$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ , and  
 566  $p$ . A. Modelling the effect of states on  $p$ . B. Modelling the effect of sex on  $s$ ,  $\beta$ ,  $\gamma$  and  $\delta$ , with  $p$   
 567 being different between breeders and non-breeders. C. Modelling the effect of states on  $s$ ,  $\beta$ ,  $\gamma$   
 568 and  $\delta$  ( $\delta$  is necessarily constant).

569

A. Hypothesis	# Model	Rank	Deviance	$\Delta AICc$
<b><math>p</math> differs between NB and B</b>	<b>Model2</b>	<b>31</b>	<b>3630,77</b>	<b>0</b>
$p$ is state-dependent	Model1	33	3630,23	3,73
$p$ is constant	Model3	30	3642,96	10,06

B. Hypothesis	# Model	Rank	Deviance	$\Delta AICc$
<b>No sex difference on <math>S</math>, <math>\beta</math>, <math>\gamma</math> and <math>\delta</math></b>	<b>Model8</b>	<b>18</b>	<b>3636,86</b>	<b>0</b>
Sex difference on $S$ , $\beta$ and $\delta$	Model5	27	3632,09	14,08
Sex difference on $S$ , $\gamma$ and $\delta$	Model6	27	3632,90	14,88
Sex difference on $\beta$ , $\gamma$ and $\delta$	Model7	27	3633,21	15,19
Sex difference on $S$ , $\beta$ and $\gamma$	Model4	30	3630,77	19,11
Sex difference on $S$ , $\beta$ , $\gamma$ and $\delta$	Model2	31	3630,77	21,24

C. Hypothesis	# Model	Rank	Deviance	$\Delta AICc$
$S$ and $\gamma$ are constant; $\beta$ is state-dependent	Model15	12	3640,14	0
<b><math>S</math> and <math>\gamma</math> are constant; <math>\beta</math> differs between NB and B</b>	<b>Model16</b>	<b>10</b>	<b>3644,82</b>	<b>0,59</b>
$S$ , $\beta$ and $\gamma$ are constant	Model17	11	3644,79	2,61
$\gamma$ is constant; $S$ and $\beta$ are state-dependent	Model10	15	3638,35	4,38
$S$ is constant; $\beta$ and $\gamma$ are state-dependent	Model14	15	3638,68	4,72
$S$ differs between NB and B; $\beta$ and $\gamma$ are state-dependent	Model13	16	3637,55	5,64
$S$ , $\beta$ and $\gamma$ are state-dependent	Model8	18	3636,86	9,10
$\beta$ differs between NB and B; $S$ and $\gamma$ are state-dependent	Model11	16	3641,59	9,69
$\gamma$ differs between NB and B; $S$ and $\beta$ are state-dependent	Model9	16	3678,02	46,12
$\beta$ is constant; $S$ and $\gamma$ are state-dependent	Model12	15	3725,68	91,71

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