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29 ABSTRACT

30 High levels of halogenated organic contaminants (HOCs) have been found in the marine
31 predatory seabird great skua (*Stercorarius skua*) from breeding colonies in the Northeastern
32 Atlantic, with large unexplained inter-colony variation. The present study aimed at analyzing
33 if the HOCs occurrence in breeding great skuas in remote colonies was explained by local
34 baseline food web exposure determined by long-range transport, or by ecological factors such
35 as diet specialization and relative trophic position in the breeding area. The occurrence of
36 organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), and polybrominated
37 diphenyl ethers (PBDEs) was analyzed in plasma of 204 adult great skuas collected over two
38 years (2008 and 2009) and 5 colonies across the North-Atlantic from Shetland to Svalbard.
39 The Σ HOCs levels in plasma ranged across two orders of magnitude, from 40 to 7600 ng/g
40 (wet weight) and differed significantly across the great skua colonies. The variation in
41 contaminant occurrence among colonies did not reflect long-range transport through a
42 latitudinal or remoteness gradient, as the second northernmost colony (Bjørnøya), had the
43 highest contaminant concentrations. No latitudinal or remoteness gradient was evident in the
44 contaminant pattern among the colonies. The contaminant levels increased significantly with
45 increasing $\delta^{15}\text{N}$ values, and regurgitated pellets of undigested prey suggested that great skuas
46 with higher $\delta^{15}\text{N}$ values had a higher proportion of bird prey in their diet, mostly seabirds. In
47 contrast, great skuas from colonies with lower $\delta^{15}\text{N}$ and lower contaminant level fed mostly
48 on fish. The enrichment of $\delta^{13}\text{C}$ increased with decreasing $\delta^{15}\text{N}$ and lower contaminant levels.
49 Therefore, individual behavior of great skuas, such as migration strategies and diet
50 specialization, rather than long-range transport and thus baseline food web exposure, explain
51 among and within colony variance in contaminant occurrence.

52

53 **Keywords:** Great skua, *Stercorarius skua*, long-range transport, Arctic, global fractionation,
54 remoteness index
55
56 **Capsule:** Trophic level and diet specialization, not long-range transport, explains the large-
57 scale variation in contaminant occurrence in the avian predator great skuas breeding in the
58 Northeastern Atlantic.

59 INTRODUCTION

60 High levels of halogenated organic contaminants (HOCs) have been found in the marine top
61 predator great skua (*Stercorarius skua*) from remote breeding colonies in the Northeastern
62 Atlantic with large unexplained inter-colony variation (Leat et al. 2013). Adult birds are
63 exposed to HOC through the diet, and lipid soluble recalcitrant HOCs biomagnify through the
64 food web with increasing HOC concentrations with trophic position (Borgå et al. 2001; Ruus
65 et al. 2002; Borgå et al. 2004). As great skuas are migratory, a central question is whether the
66 source of HOCs is predominantly from the local food web near the breeding colonies or from
67 their wintering areas. The great skuas' wintering areas have previously been shown to explain
68 up to 22% of the variation in HOC concentrations in some of the breeding colonies, leaving
69 much unexplained variance in HOC occurrence to other factors such as local sources during
70 the breeding period (Leat et al. 2013). Local HOC sources, both the baseline exposure and
71 ecological factors like trophic position and diet specialization, may influence the HOC
72 occurrence, as they affect the trophic magnification in the food web (Borgå et al. 2012).

73
74 A principal source of spatial variation in contaminants in the abiotic environment is the
75 location relative to primary emission sources. Primary emission sources of HOCs have been
76 concentrated in industrialized- and agricultural areas mostly in the mid-latitudes of the
77 northern hemisphere (Breivik et al. 2004). However, long-range transport of HOCs in the
78 atmosphere has distributed these pollutants to remote environments, including the Arctic
79 (Muir and de Wit 2010). Although long-range transport is considered the main source of
80 contaminants to the Arctic, local sources such as settlements and military sites may contribute
81 as well (Brown et al. 2009). One of the physical processes governing long-range atmospheric
82 transport is cold condensation; that is, semi-volatile HOCs shift from gas phase to condensed

83 phases with decreasing temperature (Wania and Mackay 1993). Differences in chemical's
84 volatility lead to compositional shifts in pollutant pattern along latitudinal and altitudinal
85 temperature gradients, a process often referred to as global distillation (Wania and Mackay
86 1993). As an alternative to global distillation, the remoteness hypothesis suggests that it is the
87 differential removal of pollutants from the air with distance from emission sources, rather
88 than a temperature gradient, that leads to environmental gradients of pollutants (von Waldow
89 et al. 2010a, von Waldow et al. 2010b). Comparisons of HOCs in wildlife between eastern
90 and western Greenland at similar latitude appear to support the remoteness hypothesis, with
91 higher HOC concentrations in eastern Greenland closest to European emission sources
92 (Vorkamp et al. 2004). However, observational data show that latitudinal gradients often
93 coincide with remoteness from emission sources, particularly in Europe (von Waldow et al.
94 2010a, von Waldow et al. 2010b).

95
96 Most studies of spatial distribution and long-range transport of pollutants focus on abiotic
97 compartments such as soils water and air (Agrell et al. 1999, Meijer et al. 2003, Gioia et al.
98 2006, Gioia et al. 2008, Lohmann et al. 2009). Few studies have been carried out in biota, but
99 a study of great black-backed gull (*Larus marinus*) along the Norwegian coast recognized the
100 pattern of contaminant occurrence predicted by global distillation (Steffen et al. 2006). Also,
101 studies of eagles suggested that at remote sites without local point sources, long-range
102 transport of contaminants is important (Elliott et al. 2009, Fort et al. 2014).

103
104 The effect of diet on biomagnification of HOCs is well known and established for food webs
105 using stable isotopes of nitrogen ($\delta^{15}\text{N}$) as a measure of relative trophic position (Fisk et al.
106 2001, Borga et al. 2012). Even within a seabird colony, a positive association between trophic
107 position and contaminant concentrations may be significant (Sagerup et al. 2002), although

108 such intraspecific relationship is rarely documented in wildlife, probably due to restrictions in
109 sample sizes and lack of sufficient dietary variance among individuals. Whilst most great skua
110 pairs are generalist predators, some specialize on specific prey types and in some cases on
111 individual species (Furness 1987, Phillips et al. 1997). Therefore, some of the variation in
112 HOC occurrence in top predators such as great skuas may be explained by variation in diet,
113 either among or within colonies.

114

115 The distribution of great skua breeding colonies across the Northeast Atlantic cover a large
116 spatial scale (60°N to 80°N), and thus provides the opportunity to analyze if the spatial
117 variance of HOCs reflects the pattern caused by long range transport, or if trophic level and
118 diet specialization overrides this food web baseline signal. Concentrating on a single species
119 controls for some aspects of biological variability, which can affect HOC concentrations such
120 as species-specific biotransformation rates, and physiological adaptations, for example
121 metabolic rates. The great skua feed opportunistically on a wide range of prey, including
122 adults and chicks of seabirds, terrestrial mammals, pelagic fish, demersal fish discarded from
123 fishing boats, prey gained through kleptoparasitism of other seabirds, and some caught in
124 surface waters (Furness 1987). Indigestible prey remains are regurgitated in the form of
125 pellets, from which prey can be identified to taxon or in some cases species level. The diet of
126 individual skua pairs can be studied as they are highly territorial, and regurgitated pellets are
127 thus highly likely to be produced by the birds occupying that territory.

128

129 The aim of the present study was to unravel if the occurrence of HOCs in great skuas across
130 the North Atlantic was explained by local baseline food web exposure determined by long-
131 range transport, or by relative trophic positions and local diet habits. We hypothesized that if
132 long-range atmospheric transport is the dominant process affecting HOC concentrations in

133 great skuas, plasma concentrations would decrease either with increasing latitude or
134 remoteness from emission sources. In addition, the pattern of HOCs was predicted to show
135 increasing proportion of less halogenated (thus more volatile) compounds in the northern
136 colonies or with increasing remoteness. If the HOC levels were predominantly described by
137 the great skuas trophic position and diet specialization we expected the HOC levels to be
138 related to the stable isotope signatures of nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$), and to prey
139 remains in regurgitated pellets. None of the breeding colonies included in this study are close
140 to large urban areas, therefore it is presumed that sources of HOCs are dominated by long-
141 range atmospheric transport rather than local emission sources.

142

143 MATERIALS and METHODS

144 *Sampling*

145 Blood was sampled using heparinized syringes from 204 incubating adult great skuas in five
146 colonies in 2008; Ny-Ålesund 78°55'N, n=5; Bjørnøya 74°29'N, n=15; Hjelmsøya 71°03'N,
147 n=4; South-East Iceland 63°52'N, n=21; and Runde 62°23'N, n=5 (listed with decreasing
148 latitude), and in three colonies in 2009; Bjørnøya n=51; Iceland n=56, and Shetland 60°09'N,
149 n=47 (Table 1, Table S1). Plasma and red blood cells were separated by centrifuging and
150 frozen at -20°C. Birds were sexed by molecular methods using DNA extracted from red blood
151 cells following a modified protocol (Griffiths et al. 1998). Whilst all birds sampled in 2009
152 were successfully sexed, some of the individuals sampled in 2008 were not due to insufficient
153 quantity of red blood cells for analyses.

154 *Remoteness indices*

155 Remoteness of the colonies was estimated using remoteness indices with night-time light
156 emissions and cropland areas as proxies for HOC emissions from industrial and agricultural
157 activities respectively (von Waldow et al. 2010b) (see supporting information for details). For

158 the two emission scenarios, the colonies increased in remoteness as follows; Shetland and
159 Runde < Hjelmsøya and Iceland < Bjørnøya < Ny-Ålesund using night-time light emissions
160 and Shetland and Runde < Hjelmsøya < Iceland < Bjørnøya < Ny-Ålesund using cropland
161 area. Recently quantification of remoteness was modulated to take into account the
162 contaminants' physicochemical properties (Göktaş and MacLeod 2016). As these results were
163 highly correlated with von Waldow's remoteness index, the colonies were ranked according
164 to the original remoteness index.

165

166 *Contaminant analyses*

167 OCs (HCB, Dieldrin, Mirex, QCB, OCS, β -HCH, *trans*-nonachlor, oxychlordane, *cis*-
168 nonachlor, *cis*-chlordane, *pp'*-DDT, *pp'*-DDE, *pp'*-DDD, and PCB congeners: 31/28, 52, 49,
169 74, 70, 101, 99, 110, 149, 118, 153, 105/132, 138, 158, 187, 183, 128, 177, 156/171, 180,
170 191, 170, 201, 195/208, 194, 205, 206, 209) and PBDEs (congeners 47, 99, 100, 153, 154)
171 were extracted and measured in a total of 204 plasma samples at the Great Lakes Institute for
172 Environmental Research (GLIER) at the University of Windsor Ontario Canada by gas
173 chromatography electron capture detection (GC-ECD) (Lazar et al. 1992). Lipid content
174 (extractable organic content) in plasma was gravimetrically determined. The details of the
175 analyses are described in Leat et al. (2013). Recoveries of the internal recovery standard PCB-
176 30 were 77.4 +10% (mean + SD) in 2008 and 69.5 + 9.6 % in 2009. The data were recovery
177 corrected. Method detection limit was 0.08 $\mu\text{g}/\text{kg}$ for PCBs and 0.05 $\mu\text{g}/\text{kg}$ for all other OCs.
178 Method detection limits for PBDEs varied by congener; PBDE-47 0.373 $\mu\text{g}/\text{kg}$, PBDE-99
179 0.361 $\mu\text{g}/\text{kg}$, PBDE-100 0.471 $\mu\text{g}/\text{kg}$, PBDE-153 0.420 $\mu\text{g}/\text{kg}$, PBDE-154 0.488 $\mu\text{g}/\text{kg}$.

180

181 *Dietary descriptors*

182 Stable isotopes of nitrogen and carbon ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ respectively) were measured in red
183 blood cells as described in detail elsewhere (Leat et al. 2013). The analytical precision based
184 on the standard deviation of a NIST 8414 standard (bovine muscle) for 2008 and 2009 data
185 was 0.07 and 0.18‰ for $\delta^{15}\text{N}$ and 0.08 and 0.09‰ for $\delta^{13}\text{C}$, respectively. An internal
186 laboratory fish muscle standard was included for 2009 data for which the standard deviations
187 were 0.22‰ and 0.08‰ for $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$, respectively. The variation in spatial baseline
188 isotopic signature among remote sites (Green et al. 2016) is assumed to be negligible
189 compared to signature variation caused by differences in diet in the great skuas.

190 In 2009, breeding territories of blood-sampled birds in each colony were searched for
191 regurgitated pellets every 2-3 days, until the nest failed or fieldwork ceased. In 2008 and
192 2009, additional sampling of regurgitated pellets from great skuas breeding territories also
193 took place on an *ad hoc* basis in the Bjørnøya colony. Pellets were examined and classified by
194 prey type; bird, fish, mammal or other, and identified to species level whenever possible.
195 Carcasses of large birds, e.g. ~20 days old great skua chicks and rabbits (Shetland) were
196 counted as equivalent to 1 pellet. Sagittal otoliths recovered from the regurgitated pellets were
197 used to identify the fish species consumed (Härkönen 1986). A minority of all pellets
198 contained more than one prey type and were divided proportionally between the identified
199 groups of prey i.e. 0.5 fish, 0.5 bird. Vegetation in pellets was excluded from the dietary data
200 analysis as it was considered to have been ingested incidentally along with animal dietary
201 items. Territories with > 5 regurgitated pellets were classified as specialists if $\geq 70\%$ of pellets
202 came from one dietary prey group (bird fish or mammal) (Votier et al. 2004). Otherwise, they
203 were classified as generalists. Regurgitated pellets were destroyed once sampled, and
204 identified on site to prevent double counting, or they were removed for further examination.
205 Food regurgitated whilst handling the birds was not included in calculations of diet
206 composition, but was used to identify prey items from the pellets.

207 *Data analysis*

208 Data from 2008 and 2009 were analyzed separately, as not all colonies were sampled, as not
209 all variables were included in both years. That is, whilst the sex of all individuals sampled in
210 2009 was identified using DNA, the majority of individuals sampled in 2008 were not sexed
211 due to insufficient material for DNA analysis. Treating the years separately allows sex to be
212 included as an explanatory variable in the analysis of the 2009 data. Twelve individuals, three
213 from Bjørnøya and nine from Iceland, sampled in both 2008 and 2009 were included in the
214 analysis. Since males do the majority of foraging during incubation and often regurgitate food
215 to the female at the nest (Catry and Furness 1999, Caldow and Furness 2000) the registered
216 data (HOCs, stable isotopes, regurgitated pellets) in males and females of the same nest (pair)
217 may not be independent. To ensure independence of data, only the first bird sampled per
218 territory/nest was included in the analysis.

219

220 OCs were included in the data analysis if concentrations were above method detection limits
221 in $\geq 70\%$ across the colonies. In total 40 organochlorines were included in the analysis: 28
222 PCBs ranging from tri- to deca- homologues, and 12 organochlorine pesticides including
223 HCB, OCS, β -HCH, Mirex, trans-nonachlor, cis-nonachlor, oxychlordane, pp'-DDT, pp'-
224 DDD, and pp'-DDE. Non-detects in the included contaminants were replaced by modelled
225 values below the detection limit, using the distribution of concentrations above the detection
226 limit of an individual HOC as described in Leat et al. (2013) and in supplementary
227 information (Helsel 2006). Some of the 2009 data are published in Bourgeon et al. (2012) and
228 Leat et al. (2013).

229

230 In all analyses, PCBs were grouped according to homologue group as volatility and long-
231 range transport depends upon the degree of halogenation. Detection rates of the five PBDEs

232 analyzed (PBDE-47, 99, 100, 153, and 154) varied between congeners, colonies and years
233 (Figure S2). PBDE 47 was detected in 97% of the samples and in samples where all five
234 PBDEs were above detection limit (n=89, four individuals sampled in both years were
235 excluded from the 2009 dataset), PBDE 47 was positively correlated with the other PBDEs
236 (Spearman's rank correlations: rho = 0.81, S = 22236, rho = 0.84, S = 18349, rho = 0.61, S =
237 46348, rho = 0.67, S = 38931, p <0.001 in all cases) (Figure S3). Therefore PBDE 47 was
238 used as a proxy for commercial penta-BDEs in analysis of spatial variation in PBDE
239 concentration.

240

241 Lipid content in plasma did not differ significantly between colonies (2008: $F_{447} = 0.39$, p =
242 0.81; 2009: $F_{249} = 2.4$, p = 0.098, two low outliers were removed to meet assumption of
243 normality of residuals) and all HOC concentrations are presented on a wet weight basis.

244

245 Contaminant concentrations and patterns among colonies were investigated using the
246 multivariate methods redundancy analysis (RDA) and principal component analysis (PCA)
247 using the *vegan* package in R (R Development Core Team 2015). Colony, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ and
248 lipid content were included as explanatory variables (active in RDA, passive in PCA) for both
249 years. The percentage of variation attributable to each explanatory variable was assessed by
250 RDA. This was done by comparing the residual variance of the full model with models
251 constrained to each explanatory variable in turn, separately (by conditioning out the other
252 explanatory variables). Sex was included as explanatory variable in 2009, only. HOC
253 concentrations were logarithmically transformed to reduce variance heterogeneity and
254 skewness, whilst to investigate HOC pattern the data were standardized by norm to analyze
255 the relative contribution of each contaminant to the total.

256

257 The sum of the OCs, and PBDE 47, were used to examine the overall relationships between
258 diet, mainly carbon source ($\delta^{13}\text{C}$), and relative trophic level ($\delta^{15}\text{N}$), and HOCs. Relationships
259 between stable isotopes and HOCs concentrations were investigated using general linear
260 models with backwards stepwise selection to remove non-significant variables. HOC
261 concentration data were log transformed to reduce heterogeneity and skewness (Full model
262 for 2009 in Equation 1).

263

$$264 \quad \log \Sigma\text{OC or } \log \Sigma\text{PBDEs} = \delta^{15}\text{N} + \delta^{13}\text{C} + \% \text{lipid} + \text{sex} + \varepsilon \quad (\text{Eq. 1})$$

265

266 The relationship between diet measured by regurgitated pellets and stable isotopes was
267 investigated by linear models whereas the relationship between pellets and HOCs was
268 examined visually due to small sample sizes of pellets.

269

270 RESULTS AND DISCUSSION

271 Overall, the data show that great skuas feeding at higher trophic levels and specializing on
272 bird prey (inferred from stable isotopes and regurgitated pellets) have higher concentrations of
273 pollutants, and that this is the main explanation for variation in HOC occurrence among
274 colonies, rather than global distillation and remoteness (colonies spanning nearly 19 degrees
275 latitude) to primary sources and wintering sites (Leat et al. 2013). Large sample sizes were
276 obtained from Bjørnøya, Runde, Iceland and Shetland (15 – 56 individuals), whereas small
277 sample sizes from Hjelmsøya, Runde and Ny-Ålesund (4 – 5 individuals) limit the
278 interpretation of results from these colonies. Contaminant concentrations decreased in the
279 following order: Bjørnøya (2009) > Bjørnøya (2008) > Hjelmsøya (2008) > Runde (2008) >
280 Iceland (2009) > Ny Ålesund (2008) > Iceland (2008) > Shetland (2009). *pp'*-DDE and PCB
281 153 were the contaminants of highest concentrations in all colonies for both years, followed

282 by oxychlordane, mirex, and trans-nonachlor (Table 1).

283

284 *HOCs occurrence in great skua plasma and long-range transport*

285 To meet the predictions of global distillation or remoteness hypotheses, Runde/Shetland and

286 Ny-Ålesund should be extremes, representing least remote/lowest latitude and most

287 remote/highest latitude, respectively. However, great skuas from Ny-Ålesund and Runde had

288 intermediate contaminant concentrations compared to those of the other colonies (Table 1).

289 Great skuas from Shetland had lowest contaminant concentrations, despite being the least

290 remote/lowest latitude colony. The HOC concentrations were higher in the remote and high

291 latitude colony Bjørnøya, compared to the other colonies both in 2008 and 2009. Skuas from

292 Bjørnøya had twice as high mean Σ OC concentrations as the second most contaminated

293 colony in 2008 (Hjelmsøya), and more than 3.5 times the mean concentrations from Iceland in

294 both years (Table 1).

295

296 Lipophilic HOC concentrations in the blood of birds are known to increase with decreasing

297 body mass and lipid reserves (Bustnes et al. 2012b, Bustnes et al. 2017). Therefore,

298 differences among colonies in the average body condition of great skuas offer a potential

299 explanation for spatial differences in HOC concentrations. However, the present study is

300 biased towards the selection of breeding birds. Great skuas are long-lived and may offset

301 breeding seasons if conditions are poor (Cstry et al. 1998). Hence, birds in poor condition are

302 unlikely to have been sampled. There was, however, no significant difference in body mass or

303 wing length of great skuas among colonies sampled in 2009 (Bourgeon et al. 2012). Thus,

304 body condition was not considered an important explanatory factor for variation in HOC

305 occurrence in the present study, and therefore not included in the statistical analyses.

306

307 PCA plots of HOC concentrations and patterns are presented in supporting information
308 (Figure S1). No clear latitudinal gradient was found (selected representative HOCs are
309 presented in Table 1). The direct multivariate redundancy analyses (RDA) accounted for 32%
310 and 60% of the variance in HOC concentrations in 2008 and 2009, respectively (Table 2).
311 Significant explanatory variables accounting for most of the variance in concentrations were,
312 in decreasing order: colony, lipid%, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in 2008; and colony, $\delta^{15}\text{N}$, $\delta^{13}\text{C}$ and lipid
313 in 2009 (Figure S1, Table 2). The direct multivariate redundancy analyses accounted for 52%
314 and 18% of the variance in HOC patterns in 2008 and 2009, respectively (Table 2).
315 Significant explanatory variables accounting for most of the variance in patterns were, in
316 decreasing order: colony, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in 2008; and colony, sex, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ in 2009
317 (Figure S1, Table 2). Male great skuas had higher HOC concentrations compared to females,
318 which is in accordance with findings in other avian wildlife (Bustnes et al. 2003, Bustnes et
319 al. 2005, Bustnes et al. 2007, Bustnes et al. 2017). However, neither the lipid content or sex
320 were found to be major contributors to the inter-colony difference in HOC occurrence in great
321 skuas. There was no clear latitudinal trend in the relative pattern of HOCs in general, or in
322 PCB homologue patterns in either year. The contaminants with the largest variance in relative
323 contribution among the samples were penta- to nona-CB homologue groups, *pp'*-DDE, *pp'*-
324 DDT, oxychlorane and Mirex. If long-range transport would explain the contaminant pattern
325 found in great skuas, the pattern in the Bjørnøya colony should have had proportionally more
326 volatile HOCs, and the pattern in the Iceland colony would have been intermediate to
327 Shetland and Bjørnøya, which is not the case.

328

329 The lack of a latitudinal or remoteness gradient in concentrations and pattern of HOCs in
330 great skuas is in contrast to abiotic studies (Agrell et al. 1999, Meijer et al. 2003, Gioia et al.
331 2006) and the few previous studies of geographic distribution of HOCs in biota (Ter Schure et

332 al. 2002, Vorkamp et al. 2004, Steffen et al. 2006, Bourgeon et al. 2012, Bustnes et al. 2012a).
333 Most of the species in which latitudinal or remoteness gradients in HOCs have been found are
334 cold blooded species occupying lower trophic levels compared to the great skua (Ter Schure
335 et al. 2002, Vorkamp et al. 2004, Bourgeon et al. 2012, Bustnes et al. 2012a). The HOC
336 pattern in the skuas across the colonies is dominated by recalcitrant and biomagnifying HOCs.
337 Therefore, biotransformation of HOCs in the trophic levels preceding that of the great skua,
338 and in the great skua itself, seem to obscure long-range abiotic transport patterns. In general,
339 biotransformation is efficient in warm blooded species compared to cold blooded lower
340 trophic level species where the contaminant pattern to a higher degree resembles that of the
341 abiotic environment (Borga et al. 2004). The dominance of highly chlorinated PCBs
342 congeners in the pattern of HOCs in Bjørnøya birds in 2008 is indicative of a pattern
343 dominated by biomagnification, rather than long-range transport (Borgå et al. 2001).

344

345 *Relative trophic position and HOC concentrations in great skua*

346 The stable isotope signatures differed among colonies (Figure 1, Table 1, MANOVA 2008:
347 Pillai = 0.87 $F_{448} = 10$ $p < 0.001$; 2009: Pillai = 0.96 $F_{2150} = 70$ $p < 0.001$) and there were no
348 differences between sexes (2009: Pillai = 0.018 $F_{1150} = 1$ $p = 0.26$). In 2008, great skuas from
349 Bjørnøya had higher $\delta^{15}\text{N}$ than Iceland, Ny-Ålesund and Runde skuas ($r^2 = 0.42$, $F_{448} = 10.5$,
350 $p < 0.001$, Tukey $p < 0.005$). Hjelmøy, with similar $\delta^{15}\text{N}$ to Bjørnøya, had higher $\delta^{15}\text{N}$ values
351 than Iceland ($p = 0.042$). In 2009, skuas from Bjørnøya had higher $\delta^{15}\text{N}$ values than Iceland
352 and Shetland skuas (Kruskal-Wallis $\chi^2 = 121$, $df = 2$, $p < 0.001$, post hoc comparisons
353 Wilcoxon Signed rank tests $p < 0.001$), and Iceland skuas had higher $\delta^{15}\text{N}$ than Shetland (p
354 < 0.001). As stable isotopes and colony could not be judged as independent variables, colony
355 was excluded from models investigating the relationship between HOC concentrations and
356 stable isotopes.

357
358 Across colonies there was a positive linear relationship between $\delta^{15}\text{N}$ and ΣHOC
359 concentrations (2008: $R^2=0.11$, slope=-0.55, $p=0.007$, 2009: $R^2=0.59$, slope=0.93, $p<0.0001$,
360 Figure 2). Although lipophilic recalcitrant HOCs show increasing concentrations with trophic
361 position in the food web (Borgå et al. 2001, Fisk et al. 2001, Ruus et al. 2002, Lavoie et al.
362 2010), such relationships are rarely shown within one species. However, $\delta^{15}\text{N}$ levels were
363 found to be positively related to OC concentrations in both bald eagles (*Haliaeetus*
364 *leucocephalus*) (Elliott et al. 2009) and white-tailed eagles in Norway (*H. albicilla*) (Bustnes
365 et al. 2013), and to HCB, DDTs and PCBs (from muscle) in Bjørnøya glaucous gulls (*Larus*
366 *hyperboreus*) (Sagerup et al 2002.), and weak positive relationships were found between $\delta^{15}\text{N}$
367 and HOCs in liver of northern fulmars (*Fulmarus glacialis*) (Knudsen et al. 2007). Even
368 though there are no measurements of baseline $\delta^{15}\text{N}$ from the study area, we assume that $\delta^{15}\text{N}$
369 is an indicator of relative trophic level. Green et al. (2016) reported that blue mussels (*Mytilus*
370 *edulis*) occupy the same trophic level along the whole Norwegian coast (range: 5-8‰), with
371 some stations appearing to be outliers (particularly low or high $\delta^{15}\text{N}$). However, the $\delta^{15}\text{N}$
372 levels in these stations was attributed to anthropogenic influence, i.e. increased anthropogenic
373 input. Thus, we assume that the difference in isotopic signal across the remote colonies in the
374 present study, reflects differences in diet specializations rather than differences in baseline
375 isotopic signal. In the present study, positive linear relationships between $\delta^{15}\text{N}$ and log sum
376 HOCs within the colonies were found for the Iceland colony in 2009 but not 2008 ($R^2=0.07$,
377 $p=0.05$, and $R^2=0.17$, $p=0.5$, respectively), in Shetland in 2009 ($R^2=0.10$ $p=0.03$), and the
378 Bjørnøya colony in 2008 but not 2009 ($R^2=0.26$, $p=0.03$, and $R^2=0.004$, $p=0.67$,
379 respectively). These results suggest variation in individual diet specialization or prey taxon
380 availability between breeding seasons.
381

382 The regurgitated pellets indicated differences in diet and occurrence of specialist and
383 generalist predators among colonies, with trophic position increasing from Shetland to Iceland
384 to Bjørnøya ($\chi^2 = 296$, $df = 4$, $p < 0.001$, respectively). Great skuas from Bjørnøya had a
385 higher proportion of bird in their diet (2008: $\chi^2 = 229$, $df = 1$, $p < 0.001$, 2009: $\chi^2 = 132$, $df = 1$,
386 $p < 0.001$), whilst great skuas in Iceland and Shetland had a higher proportion of fish ($\chi^2 = 86$
387 and $\chi^2 = 95$, $p < 0.001$) (Table 4). Great skuas from Shetland also had a higher proportion of
388 other diet items. Of the regurgitated pellets in Shetland, 19% ($n = 71$) contained mammal
389 remains, mostly rabbit (*Oryctolagus cuniculus*), whilst only one regurgitated mammal pellet
390 was recorded in Iceland. At Bjørnøya, the arctic fox (*Vulpes lagopus*) is the only mammal
391 present. The regurgitated mammal pellets from Shetland were concentrated in 14 territories
392 out of 42, of which only 7 had more than one regurgitated mammal pellet. Of the territories at
393 Bjørnøya where 5 or more pellets were collected ($n = 56$, a mean of 13 pellets per nest), the
394 majority were bird specialists (2008: 67%, 2009: 77%, Table 4). In contrast, 47% of great
395 skua territories from Shetland were occupied by fish specialists and 43% by generalists. In
396 Iceland, only three territories had more than 5 pellets, and recorded two fish specialists and a
397 generalist. Details of identified bird and prey species can be found in Supplementary
398 Information.

399

400 *Carbon source and HOC occurrence*

401 Bjørnøya, Hjelmsøya and Runde were depleted in $\delta^{13}\text{C}$ compared to Iceland and Ny-Ålesund
402 in 2008 ($r^2 = 0.74$, $F_{448} = 38$, $p < 0.001$, Tukey $p < 0.005$) (Table 1). Likewise, in 2009 skuas
403 from Bjørnøya were depleted in $\delta^{13}\text{C}$ compared to Iceland and Shetland ($r^2 = 0.52$, $F_{2151} = 83$,
404 $p < 0.001$, Tukey $p < 0.001$) (Table 1). Across all colonies, there was a negative linear
405 relationship between carbon source ($\delta^{13}\text{C}$) and ΣHOC concentrations (2008: $R^2 = 0.19$, slope=
406 0.73, $p = 0.0004$, 2009: $R^2 = 0.24$, slope= -1.34 , $p < 0.0001$, Figure 2). Most reported studies have

407 found few or no significant relationships between $\delta^{13}\text{C}$ values and HOCs in avian tissues
408 (Ruus et al. 2002, Sagerup et al. 2002, Knudsen et al. 2007), however, Lavoie et al. (2010)
409 found sum brominated flame retardants (BFRs) to increase with $\delta^{13}\text{C}$. Studies of coastal
410 breeding eagles report that PCBs, DDE and PBDEs in chicks increase with marine input and
411 increasing $\delta^{13}\text{C}$ levels (less negative values) (Elliott et al., 2009), and thus is opposite to the
412 findings from the present study. The study by Elliott et al., (2009) also included some
413 individual eagles breeding in estuaries or further up river and therefore reflect more riverine
414 and freshwater systems. For white-tailed eagles in Norway, there was a negative relationship
415 between $\delta^{13}\text{C}$ levels and POPs (Bustnes et al. 2013) similar to this study. $\delta^{13}\text{C}$ measurements
416 from the present study varied only from -19.8 to -17.7‰, which is at the end of scale
417 compared to the study by Elliott et al. (2009), which might explain the contrasting findings.
418

419 Spatial patterns in $\delta^{13}\text{C}$ are well known in the Southern Ocean with $\delta^{13}\text{C}$ decreasing with
420 increasing latitude (Quillfeldt et al. 2010), but less is known from the northern hemisphere. In
421 the present study, $\delta^{13}\text{C}$ in great skuas does not simply decrease with increasing latitude, as
422 Ny-Ålesund $\delta^{13}\text{C}$ is comparable to Iceland rather than Bjørnøya. Runde skuas are grouped
423 with Bjørnøya and Hjelmøy in $\delta^{13}\text{C}$ values. We hypothesized that the Shetland skuas feed to
424 a greater degree on the terrestrial (less seabird prey, more mammal prey) system as reflected
425 in the regurgitated pellets, and that this further contributes to lower HOC exposure, as HOC
426 levels in the terrestrial system are generally lower than the marine system for the North
427 Atlantic and Arctic region (AMAP 2004). If so, Ny-Ålesund and Iceland would also reflect a
428 more enriched $\delta^{13}\text{C}$ accompanied with lower HOC levels compared to e.g. Bjørnøya. Remains
429 of mammals were found in regurgitated pellets from Iceland, and also the Ny-Ålesund skuas
430 are known predators on eggs and chicks of terrestrial feeding geese (Bustnes, Personal
431 communication). However, none of these colonies had a distinct terrestrial $\delta^{13}\text{C}$ signal.

432 Bjørnøya was the colony with the lowest $\delta^{13}\text{C}$ values indicating a marine signal, and the
433 highest levels of HOCs, which support the general assumption that marine food webs are
434 more contaminated than terrestrial food webs. However, further research is needed on the
435 baseline levels of $\delta^{13}\text{C}$ along a northern-Arctic, latitudinal gradient, as has been done for
436 southern-Antarctic areas.

437

438 *Measuring diet – combining stable isotopes and pellet data*

439 When combined with regurgitated pellets, the indication for colony differences in HOCs
440 being mostly driven by the trophic position at breeding site is even stronger. Across both
441 years, birds from Bjørnøya had the highest HOC concentrations and the highest $\delta^{15}\text{N}$ values,
442 which probably resulted from Bjørnøya skuas having the highest proportion of bird prey in
443 their diet. Bjørnøya had the highest proportion of individuals specializing on bird prey, and
444 these skuas had consistently higher concentrations of both OCs and PBDEs compared to
445 generalist skuas in Bjørnøya. This has also been found for glaucous gulls (*Larus hyperboreus*)
446 at Bjørnøya (Bustnes et al. 2000). Shetland at the other extreme had a more mixed diet
447 dominated by fish, with only one bird prey specialist, several skuas feeding on terrestrial
448 mammals (rabbit), and the lowest $\delta^{15}\text{N}$ values and HOC concentrations across colonies.
449 Iceland had intermediate HOC concentrations and $\delta^{15}\text{N}$ values, and a higher proportion of bird
450 prey in their diet, than birds at the Shetland colony. No pellets were collected from the three
451 smaller colonies, Ny-Ålesund, Hjelmsøya and Runde, holding less than 60 pairs at each
452 location (around 10, 5 and 50 pairs for both years, respectively). These three colonies are
453 located near to large colonies of other species including geese and eider ducks, providing
454 potential prey either through kleptoparasitism or by preying directly on eggs, chicks or adult
455 birds. Small great skua colonies have been shown to have a higher incidence of seabird

456 predation (Votier et al. 2007). However, HOC concentrations in Ny-Ålesund, Hjelmsøya and
457 Runde were lower than birds from Bjørnøya, where seabird prey dominate.

458
459 Stable isotopes and regurgitated pellets both provided information on the diet of great skuas in
460 the breeding season, and can be used to analyze the relationship between diet and HOC
461 concentrations. One obvious difference is that the regurgitated pellets only reflect the
462 momentary diet and give specific information on prey items, whereas the stable isotopes
463 integrate the carbon source and relative positioning in the food web over time. In the present
464 study, red blood cells were analyzed for stable isotopes, which integrates the stable isotopes
465 over the past 2-3 weeks (Bearhop et al. 2002). Pellets tend to over represent the proportion of
466 mammal and bird in the diet due to their amount of indigestible material, and the longevity of
467 pellets composed of fur or feathers in the environment (Furness and Hislop 1981, Votier et al.
468 2001). Regurgitates may also be biased in the other direction as a bird diet may be more
469 difficult to regurgitate than a fish diet (Furness 1987). However, presuming that the bias in
470 pellet data towards birds and mammals is constant across colonies, pellets still reflect
471 differences in diet among colonies.

472

473 *Contaminant levels compared to other studies*

474 The organochlorine concentrations in great skua plasma were amongst the highest found in
475 plasma of seabirds in contemporary studies. Glaucous gulls (*Larus hyperboreus*), particularly
476 those breeding in Bjørnøya, are known to have high concentrations of HOCs associated to
477 negative effects (Verreault et al. 2010). Great skua from Bjørnøya in 2009 had twice the DDT
478 concentrations found in glaucous gulls from Bjørnøya in 2002 and 2004 (Verreault et al.
479 2005). Higher OC concentrations in great skua compared to seabirds of a similar ecological
480 niche is not confined to Bjørnøya. Also, in Ny-Ålesund on Svalbard, OC concentrations in

481 great skuas exceeded those of glaucous gulls (Verreault et al. 2006), although both great skuas
482 and glaucous gulls have lower levels in Ny-Ålesund than Bjørnøya (Verreault et al. 2005,
483 Verreault et al. 2006). OC concentrations in great skuas from Hjelmsøya are higher than in
484 whole blood of great black-backed gulls at Hornøya in northern Norway (Bustnes et al. 2005).
485 There are no close comparisons across species available for the least contaminated colony
486 Shetland, however, plasma concentrations there exceeded those of great black-backed gulls
487 whole blood concentrations in Norway (Bustnes et al. 2005).

488
489 As in the present study of great skuas, PBDEs are generally found at low concentrations
490 compared to many of the most persistent OCs in the above-mentioned avian top predators
491 (Verreault et al. 2006, Chen and Hale 2010). As with OC, PBDE concentrations in great skuas
492 exceeded those of glaucous gulls (Verreault et al. 2007). Although the dietary niches of
493 glaucous gulls, great black-backed gulls, and great skuas overlap (Cramp and Simmons 1983)
494 the exact composition of their diets may differ both during the breeding and non-breeding
495 season. Differences in migration route and wintering areas may also contribute to both inter-
496 and intra-species differences (Steffen et al. 2006, Magnusdottir et al. 2012, Leat et al. 2013).

497
498 The HOCs that dominated the great skua pattern at all colonies (pp'-DDE, PCB-153,
499 oxychlorane, mirex, trans-nonachlor), also dominate the pattern in other seabird species
500 (Bustnes et al. 2005, Helberg et al. 2005, Borgå et al. 2007, Lavoie et al. 2010), due to their
501 high recalcitrance and lipid solubility. Trophic level and diet specialization are the most likely
502 explanation for the higher levels in great skuas than other species, in addition to physiological
503 differences such as metabolic rate, longevity, or biotransformation ability.

504

505 A strength of the present study was the large sample size covering the great skua breeding
506 population of the north-eastern Atlantic which allowed a broader spatial perspective. Large
507 sample sizes are needed to test relationships between dietary descriptors and contaminant
508 levels within the same species. These results show that large scale spatial variations in HOC
509 occurrence in great skua is dominated by ecological factors such as local diet and trophic
510 position. In combination with previous studies it can be concluded that these factors are more
511 important than both wintering area and long-range transport of contaminants from primary
512 sources. Condition, lipid content and sex differences are less important factors in the broader
513 spatial perspective. As such, for remote regions, long-range transport and predictions of
514 global abiotic distribution of contaminants is not sufficient to predict risk of contaminant
515 exposure in wildlife.

516

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527

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684 Wania, F., and D. Mackay. 1993. Global fractionation and cold condensation of low volatility organochlorine
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686

687 Table 1. Concentrations ($\mu\text{g}/\text{kg}$ ww) of selected organochlorines (OCs) and polybrominated diphenylethers (PBDEs) in plasma and stable
 688 isotope signatures of nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) in red blood cells (‰) of adult great skuas by breeding colony and year (arithmetic
 689 mean \pm SD, min-max of concentrations).

	2008					2009		
	Ny-Alesund (n=5)	NORTH Bjørnøya (n=15 ^a)	SOUTH Hjelmsøya (n=4)	Iceland (n=21)	Runde (n=5)	Bjørnøya (n=51)	NORTH Iceland (n=56)	SOUTH Shetland (n=47)
% Lipids	1 \pm 0.4 0.5-1.6	0.84 \pm 0.42 0.33-1.73	1.04 \pm 0.12 0.96-1.22	0.90 \pm 0.3 0.08-1.5	0.90 \pm 0.12 0.74-1.05	0.92 \pm 0.18 0.30-1.2	0.88 \pm 0.21 0.11-1.4	0.87 \pm 0.17 0.5-1.2
$\delta^{15}\text{N}$	13.2 \pm 0.3 12.8-13.7	13.9 \pm 0.4 13.4-14.9	13.7 \pm 0.4 13.3-14.2	13.1 \pm 0.4 12.5 - 13.6	13.3 \pm 0.3 13.0-13.7	14.6 \pm 0.4 13.7-15.3	13.3 \pm 0.4 12.0-14.0	12.7 \pm 0.4 11.0-13.6
$\delta^{13}\text{C}$	-18.2 \pm 0.2 -18.5 to -17.9	-19.1 \pm 0.3 -19.8 to -18.8	-19.3 \pm 0.4 -19.7 to -18.9	-18.2 \pm 0.3 -18.6 to -17.7	-18.8 \pm 0.2 -19.0 to -18.6	-19.1 \pm 0.3 -19.7 to -18.5	-18.5 \pm 0.3 -19.3 to -17.9	-18.5 \pm 0.3 -19.2 to -17.9
β -HCH	1.9 \pm 1.4 0.134-3.8	0.497 \pm 0.443 ND-1.8	1.03 \pm 1.35 0.205-3	0.532 \pm 0.628 ND-2.2	0.383 \pm 0.227 ND-0.56	2.4 \pm 2 0.381-11	1.1 \pm 1 ND-4.2	0.192 \pm 0.192 ND-1.2
Oxy-Chlordane	14 \pm 11 0.979-35	63 \pm 39 12-142	21 \pm 30 4.1-65	12 \pm 11 ND-44	15 \pm 22 2.9-54	98 \pm 69 7.7-335	17 \pm 16 1.8-92	3.9 \pm 7.6 0.424-51
HCB	12 \pm 7.4 1.3-20	10 \pm 8 2.6-28	11 \pm 9.3 2.9-25	4.7 \pm 3.1 0.972-14	6.5 \pm 4.5 2.6-14	23 \pm 11 4.5-57	4.6 \pm 2.4 0.741-11	1.9 \pm 1.8 0.6-12.5
Mirex	14 \pm 8.4 1.5-28	18 \pm 12 ND-39	9.9 \pm 8.7 3-22	12 \pm 18 ND-88	7.7 \pm 8.3 2.5-22	51 \pm 34 7.3-134	12 \pm 11 2-64	4 \pm 4.5 0.628-26
pp'-DDE	163 \pm 108 10-325	196 \pm 130 40-508	179 \pm 188 51-458	175 \pm 148 31-544	160 \pm 173 50-461	526 \pm 307 59-1286	164 \pm 158 19-782	55 \pm 60 7.7-347
PCB 153	127 \pm 69 8.9-222	383 \pm 181 93-676	143 \pm 102 64-284	113 \pm 106 19-515	122 \pm 91 40-269	485 \pm 301 107-1660	120 \pm 102 22-509	61 \pm 55 9-279
Σ OCs	496 \pm 248 39-757)	1657 \pm 820 404-3073	825 \pm 600 375-1694	445 \pm 458 81-2270	679 \pm 519 269-1557	2410 \pm 1452 532-7619	641 \pm 507 141-2754	316 \pm 249 81-1376
PBDE 47	11 \pm 7.6 ND-25	13 \pm 9.3 3.4-39	12 \pm 8.8 4.9-23	3.9 \pm 4 ND-16	15 \pm 21 2.7-52	17 \pm 25 ND-127	7.1 \pm 4.6 ND-21	3.9 \pm 2 0.935-10

690 ^a n=13 for % Lipids

691 Table 2 Multivariate redundancy analysis of HOC concentrations and patterns in great skua
 692 plasma collected in 2008 (5 colonies) and 2009 (3 colonies).

693 694		Explanatory variable	Variation explained (%)	p-value	Total variation explained by RDA (%)
695	<u>Concentration</u>				
696	2008 a	Colony	34%	0.001	32%
697		Lipid %	28%	0.003	
698		$\delta^{15}\text{N}$	5.3%	0.001	
699		$\delta^{13}\text{C}$	6%	0.004	
700					
701	2009	Colony	9.8%	0.001	61%
702		Lipid %	0.6%	0.04	
703		$\delta^{15}\text{N}$	3.1%	0.001	
704		$\delta^{13}\text{C}$	0.3%	0.001	
705		Sex ^a	3.2%	0.15	
706	<u>Patterns</u>				
707	2008 a	Colony	23%	0.001	48%
708		Lipid %	4.5%	0.12	
709		$\delta^{15}\text{N}$	0.6%	0.002	
710		$\delta^{13}\text{C}$	0.3%	0.001	
711					
712	2009	Colony	53%	0.001	18%
713		Lipid %	1.6%	0.98	
714		$\delta^{15}\text{N}$	7.9%	0.001	
715		$\delta^{13}\text{C}$	4.8%	0.001	
716		Sex ^a	12%	0.04	

717 a Sex was not included in the 2008 models as the sex of the majority of individuals was
 718 unknown

719 Table 3. Diet composition of breeding great skuas from three breeding colonies in 2008 and
 720 2009 estimated from regurgitated pellets. Number and type of diet specialists ($\geq 70\%$ of pellets
 721 belonging to one dietary group, Votier et al. 2004), and generalists from study territories
 722 where more than 5 pellets were recorded.

Colony	Bjørnøya	Bjørnøya	Iceland	Shetland
Year	2008	2009	2009	2009
Date	07/07-19/07	12/07-18/08	04/06-11/07	09/06-28/07
No. of pellets	146	215	115	368
% Composition				
Fish	23	17	71	57
Bird	76	82	27	19
Mammal	N/A	0	1	19
Other	1	1	1	5
No of territories with more than 5 pellets				
Specialists	11	13	2	30
Fish	0	0	2	14
Bird	8	10	0	1
Mammal	0	0	0	2
Generalists	3	3	1	13

723

724

725 FIGURE CAPTIONS

726 Figure 1. Relationship between $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$

727 Figure 2. Relationship between isotopes and contaminant concentrations

728 Figure 3. Relationship between isotopes and diet inferred from pellet type

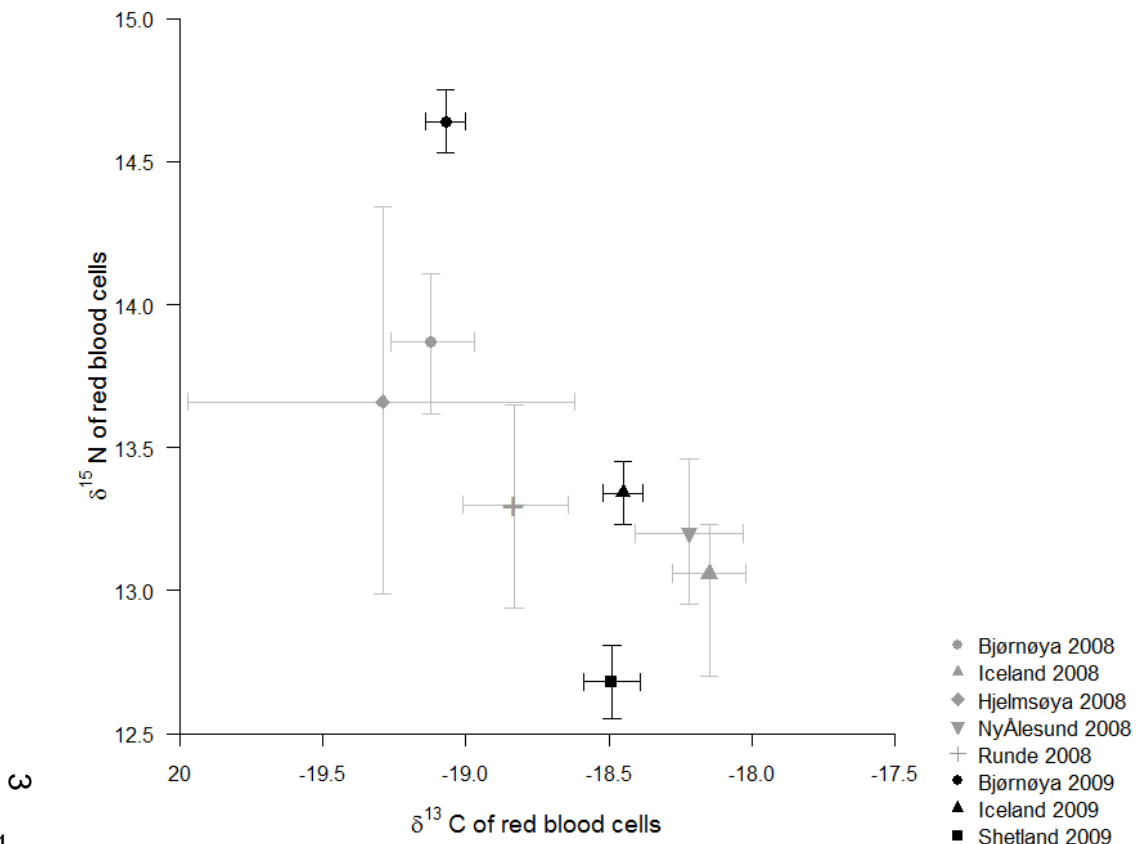
729 Figure 4. Contaminant concentrations between different specialists (diet inferred from pellets)

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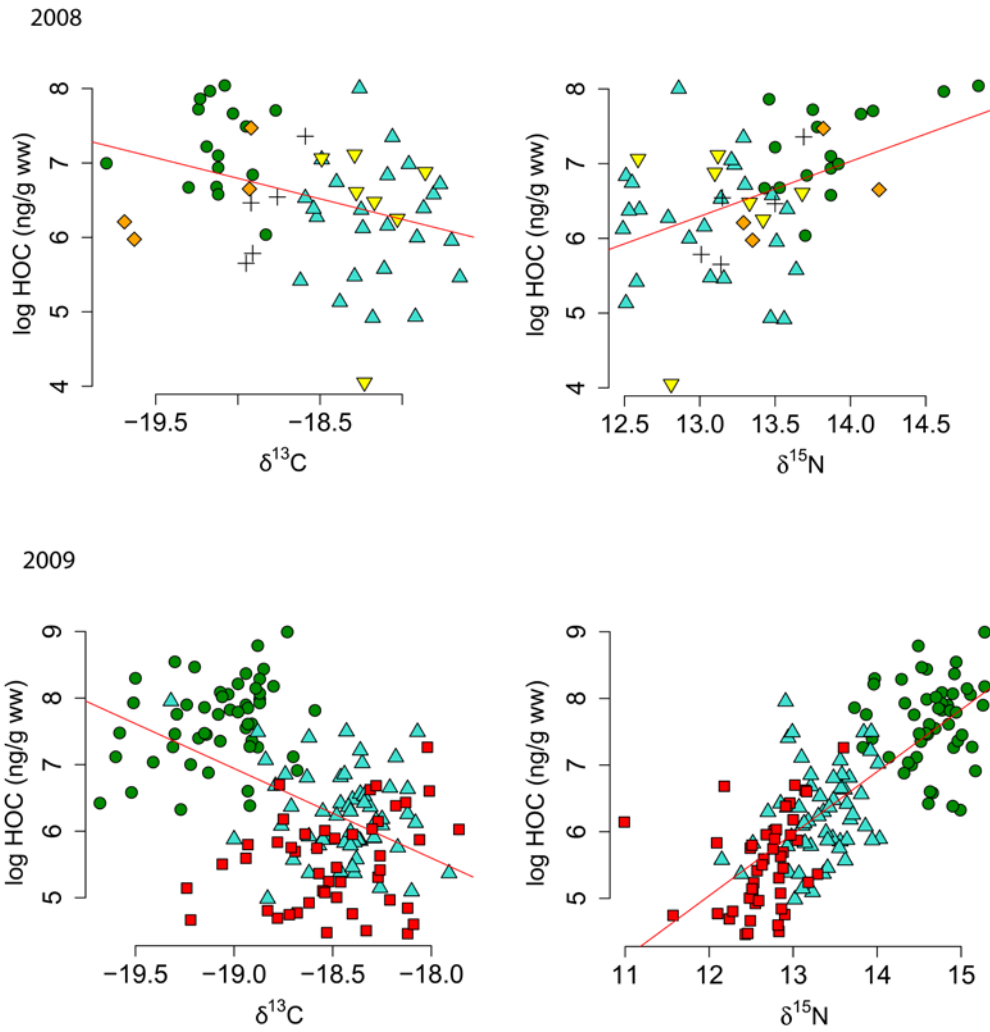


734

735 Figure 1. Great skua stable isotope values (‰) of nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) in red
736 blood cell (n = 214) by colony and year. Mean with 95% confidence intervals.

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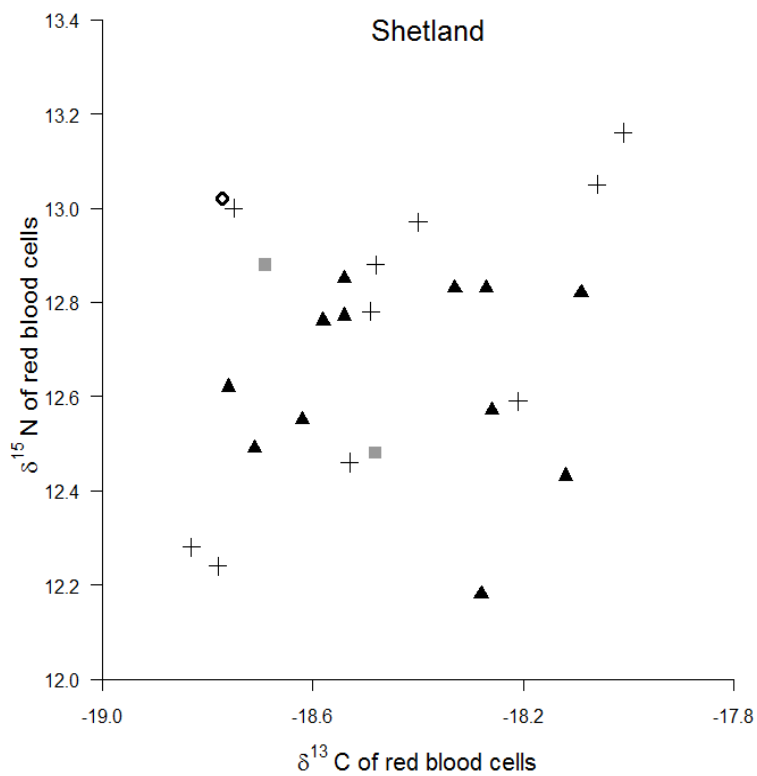
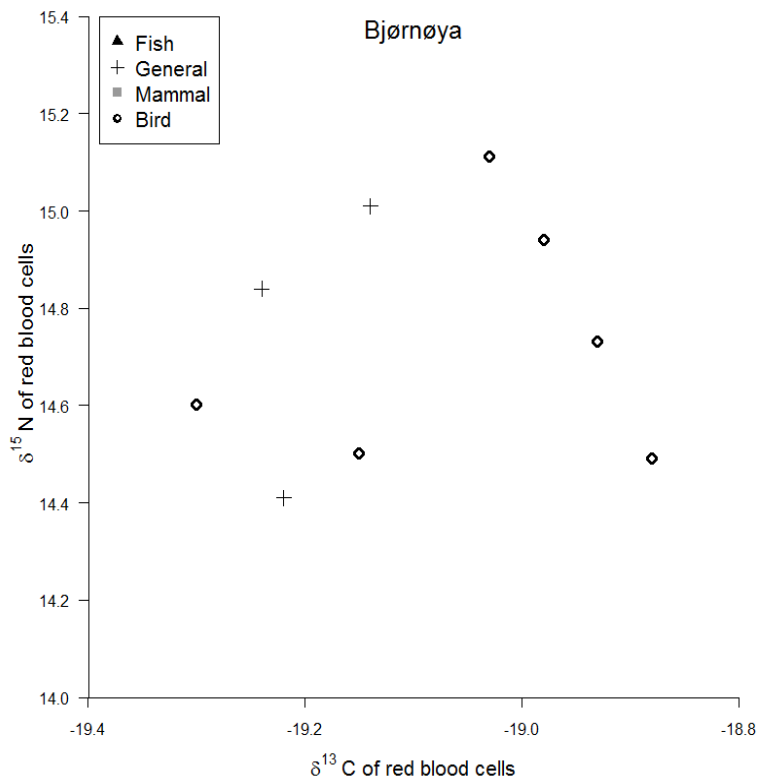
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742 Figure 2. Relationships between the sum of halogenated organic contaminants (HOCs),
 743 concentrations in plasma, and stable isotopes of nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) in red
 744 blood cells of great skuas by colony and year (top: 2008, bottom: 2009).



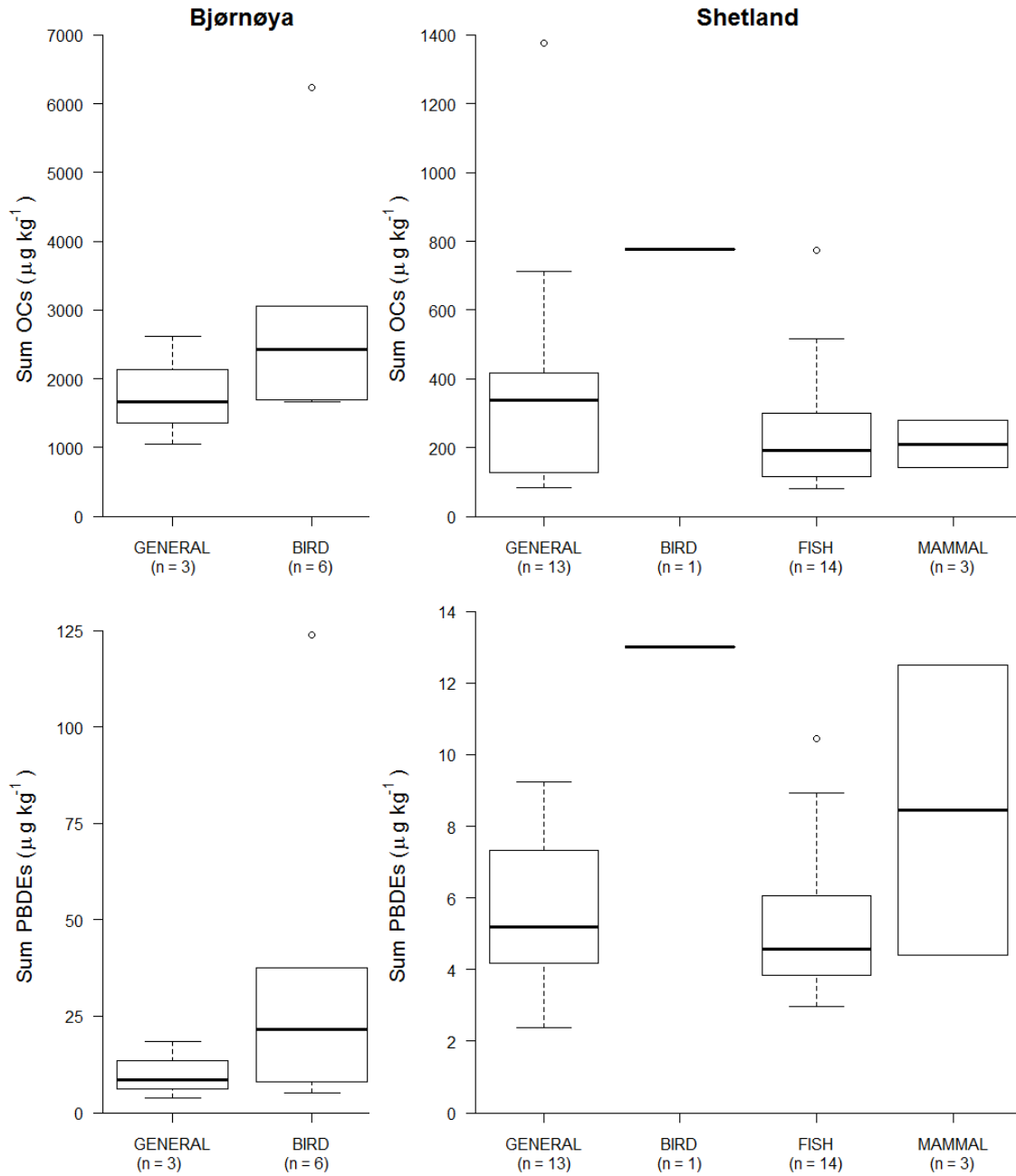
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745

746

747 Figure 3. Stable isotope values (‰) of red blood cells of great skuas from Bjørnøya (top) and

748 Shetland (below), classified from pellet type as specialists or generalists.



749

750 Figure 4. Sums of organochlorine and PBDE concentrations ($\mu\text{g/kg}$, ww) by territorial diet

751 specialisms classified by pellet type.

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