1   **Potential effect of migration strategy on pollutant occurrence in eggs of**

2   **Arctic breeding barnacle geese (*Branta leucopsis*)**

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Abstract

Arctic-breeding geese acquire resources for egg production from overwintering and breeding grounds, where pollutant exposure may differ. We investigated the effect of migration strategy on pollutant occurrence of lipophilic polychlorinated biphenyls (PCBs) and protein-associated poly- and perfluoroalkyl substances (PFASs) and mercury (Hg) in eggs of herbivorous barnacle geese (*Branta leucopsis*) from an island colony on Svalbard. Stable isotopes (δ^{13}C and d^{15}N) in eggs and vegetation collected along the migration route were similar. Pollutant concentrations in eggs were low, reflecting their terrestrial diet ($\sum$PCB = 1.23 ± 0.80 ng/g ww; $\sum$PFAS = 1.21 ± 2.97 ng/g ww; Hg = 20.17 ± 7.52 ng/g dw). PCB concentrations in eggs increased with later hatch date, independently of lipid content which also increased over time. Some females may remobilize and transfer more PCBs to their eggs, by delaying migration several weeks, relying on more polluted and stored resources, or being in poor body condition when arriving at the breeding grounds. PFAS and Hg occurrence in eggs did not change throughout the breeding season, suggesting migration has a greater effect on lipophilic pollutants. Pollutant exposure during offspring production in Arctic-breeding migrants may result in different profiles, with effects becoming more apparent with increasing trophic levels.

Introduction

Migratory birds utilize resources from multiple locations to fuel energetic costs associated with reproduction.[1,2] However, resources can also be geographically isolated, particularly for terrestrial bird species that fly overseas. Individuals may therefore be limited by where they acquire energy for both flight and reproduction.[3] In highly seasonal environments such as the Arctic, terrestrial birds often follow a "green wave" of spring resources,[4] where individuals optimize timing between high quality resources and reproductive success.[5,6] An individual's timing depends on many factors including body condition and resource availability and conditions along the flyway and at the breeding grounds.[7,8]

In female birds, reproduction includes egg production. Given feeding sites are geographically isolated, then energy directed towards egg production will range from exclusive reliance on distant wintering ground resources, to energy obtained during migration, or to reliance on local breeding resources, but is typically a mix.[3,9] Energy often represents nutrients available to an individual in the form of lipids and protein, with lipids being energetically richer and less costly to transport over long distances than protein.[10-12]

Avian eggs reveal how females both acquire and utilize energy,[13] and are useful in the biomonitoring of environmental pollutants.[14] During egg production, females maternally transfer various lipophilic pollutants including polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB), and protein-associated pollutants such as per- and polyflouroalkyl substances (PFASs) and mercury (Hg).[15-17] These contaminants are known for their persistent, bioaccumulative and toxic properties.[18-20]

58   In migratory birds such as geese, the consumption of vegetation contaminated via

59   atmospheric deposition represents a source of exposure to certain pollutants.[21,22] Several studies

60   in migratory birds have identified a spatial relationship between latitudinal position and

61   pollutant exposure.[23,24] With increasing latitude, atmospheric and soil deposition of lighter

62   chlorinated PCBs and HCB increases, whereas heavier chlorinated PCBs decreases.[25-27] The

63   exposure profile of many bird species is dominated by heavier, more persistent PCBs,[28] and this

64   profile should also reflect spatial trends in migratory birds that feed at different sites during egg

65   formation. Ecological tracers such as stable isotopes have been used in large part to identify

66   energy sources utilized during egg production,[13,29] however, ecotoxicological studies

67   combining stable isotopes and pollutants as chemical tracers have received much less attention.

68

69   The purpose of this study was to investigate how migration strategy, both in terms of timing

70   and spatial dietary energy source, affects pollutant occurrence in eggs of Svalbard-breeding

71   barnacle geese (*Branta leucopsis*). Female geese acquire and utilize terrestrial resources along

72   their migration route relative to their breeding grounds including: resources from distant

73   overwintering grounds (United Kingdom), staging areas (northern Norway), and local bird cliff

74   and island tundra (Svalbard, Norway).[13,30] Stable isotopes and observational data indicate that

75   early arriving females utilize distant resources for egg production before local breeding ground

76   resources reach peak availability,[13] while late arriving females are better suited at utilizing local

77   resources before laying eggs.[31] To our knowledge, no attempt has been made to combine stable

78   isotopes and pollutants as ecological and chemical tracers in this migratory species.

79   Additionally, storage and transport of lipids are also less costly than proteins, meaning

80   migration strategies may have a greater effect on pollutants associated with lipids than proteins.

81   Given latitudinal differences exist in the PCB and HCB profile in air and soil, then geese serve

82   as a model species to track the movement of environmental pollutants.

84   To determine where geese acquire their energy for egg production and whether this reflects

85   pollutant exposure, we collected vegetation along the flyway of the goose and eggs at the

86   Svalbard breeding grounds. We also quantified nest hatch date for the breeding population as a

87   proxy for migration timing and energy source. We hypothesized that: 1) early egg laying

88   females fuel reproduction using either stored body reserves or distant wintering ground

89   resources (UK and/or northern Norway), leading to pollutant remobilization or higher exposure

90   in females and maternal transfer to eggs; 2) late egg laying females feed on local breeding

91   ground resources (Svalbard), and are exposed to lower concentrations of pollutants than

92   individuals relying on distant resources; and 3) migration strategy has a greater effect on

93   concentrations of lipophilic pollutants (PCBs and HCB) in eggs than protein-associated

94   pollutants (PFASs and Hg).

Materials and methods

Barnacle goose biology

98   The barnacle goose population in the present study overwinter on the Solway Firth (UK) and

99   migrate to the high Arctic archipelago of Svalbard, Norway (Figure S1, Supporting

100   Information). Most individuals stopover in spring staging areas along the coast of northern

101   Norway for several weeks,[32] but a small number of birds skip these sites during their northward

102   migration.[33,34] Geese typically depart from the Solway Firth between late April and early May,

103   spending several weeks in mainland Norway before arriving at the Svalbard breeding grounds

104   in late May.[35] The geese also utilize additional pre-breeding sites on Bjørnøya and along the

105   west coast of Svalbard, which include tundra vegetation fertilized by marine birds at cliff-

106   breeding colonies.[30] When female geese arrive at the breeding grounds, they commence egg

107   laying in as little as three days.[36] Females typically lay a clutch of four eggs and only lay once

108 per breeding season.[37,38] The egg laying period for the breeding colony typically spans

109 approximately two weeks, and eggs of a clutch hatch synchronously.[38,39]

110

111 **Study sites and sampling effort**

112 In 2016, our study included three main areas along the migration route of the Svalbard-

113 breeding population of the barnacle goose, including: UK (Solway Firth); northern Norway

114 (Helgeland and Vesterålen); and Svalbard (Kongsfjorden). Barnacle geese breed on several

115 islands in the fjord,[38] and our study population represented the Storholmen Island colony

116 (78°56'N, 12°14'E).

117

118 *Sighting and nest data*

119 Intensive sightings of ringed barnacle geese were carried out in northern Norway from 29

120 April to 21 May 2016 in Vesterålen (municipalities of Andøy, Hadsel, Sortland and Øksnes),

121 and from 22 April to 21 May 2016 in Helgeland (municipalities of Herøy and Træna). On

122 Svalbard, we registered all nests on Storholmen Island and recorded ring codes of nesting

123 individuals. At least one member of each nesting pair from our sampling effort was ringed, and

124 we assumed any unringed individuals in nesting pairs represented the partner. For each

125 registered nest, we also recorded the hatching date of the clutch, defined as the first day when

126 an egg in each clutch hatched.

127

128 *Vegetation and egg sampling*

129 Vegetation was collected on the Solway Firth and Vesterålen in May 2016, and on Svalbard

130 June–July 2016. Sites on Svalbard included both island colony and marine bird cliff tundra,

131 referred to as island tundra and cliff tundra respectively. Vegetation represented a mix of

132 graminoid and forb species, reflecting the diet of the geese (see Table S1, Supporting

133 Information). Diet was sampled in areas where geese had been observed grazing and where

134 fresh droppings were present. Only the top layers of vegetation were used for subsequent

135 analysis, as geese predominantly graze at this level.[35]

136

137   For eggs, an intensive sampling effort took place during the main incubation period on

138 Storholmen Island from 9 June to 20 June. We had originally planned to sample from early and

139 late arriving females, however almost all individuals had commenced egg laying prior to our

140 sampling period. Instead, we sampled a single egg at random from 61 nests, to reduce the

141 potential effects of intra-clutch variation. Although egg laying sequence may affect pollutant

142 concentration in avian species,[40] several studies have demonstrated that mother-egg or inter-

143 clutch variation is greater than intra-clutch variation.[15,41] Thus, we assumed that each egg

144 sampled was representative of a female's entire clutch.

145

146   We prioritized sampling from nesting pairs where at least one parent was ringed and

147 observed at the staging areas in northern Norway. We attempted to sample eggs from females

148 utilizing early or late migration strategies based on sighting data from northern Norway as well

149 as egg incubation stage.[42] Eggs were stored overnight at 4°C. Embryonic age (defined as

150 incubation stage) varied greatly across all eggs, so samples were homogenized to obtain a signal

151 representing whole egg content. Homogenates were aliquoted to polypropylene tubes and stored

152 at –20 °C. Samples were analyzed for protein content and stable isotopes of carbon (δ^{13}C) and

153 nitrogen (δ^{15}N), the details of which are described in Supporting Information.

154

155 **Pollutant analysis**

156   Egg homogenates were analyzed for PCBs, HCB and PFASs at Norwegian Institute for Air

157 Research (NILU) at the Fram Centre in Tromsø, Norway. Mercury in eggs was analyzed at the

University of Oslo, Norway. PCBs and HCB were measured in vegetation from the Solway Firth, Vesterålen and Svalbard, and PFASs from one site on the Solway Firth and bird cliff tundra from Svalbard. A total of 43 compounds were analysed, including 19 PCB congeners, HCB, 22 PFAS compounds and mercury.

PCBs and HCB analysis

For eggs, approximately 1.5 g of pre-weighed homogenate was freeze-dried for moisture content removal in approximately 1:5 weight/weight (w/w) of anhydrous sodium sulfate (burnt at 600 °C). For vegetation, approximately 10 g of pre-weighed material was pulverized with liquid nitrogen and freeze-dried in 1:3 (w/w) sodium sulfate. Samples were spiked with 2.7 ng/µl of ^{13}C-labelled internal standards: PCB-28, -31, -52, -47, -37, -74, -66, -101, -99, -149, -118, -153, -105, -138, -187, -183, -180, -170, -194 and -209, and HCB. Sample homogenate was extracted three times with cyclohexane/acetone (3:1) (40/30/30 ml) in an ultrasonic bath. Supernatant from each step was combined, and then 10% of the combined supernatant was aliquoted into a pre-weighed vial for gravimetric lipid determination. The remaining supernatant was evaporated to dryness and reconstituted in 0.5 ml of isooctane and transferred to EZ-POP NP cartridges (Supelco®) for clean-up purposes. PCBs and HCB were eluted from the cartridges with 3×5 ml of acetonitrile and the eluent was evaporated and reconstituted in 0.5 ml of isooctane. An additional clean-up step was performed using automated solid phase extraction where extract was eluted with 1 g of activated Florisil® (burnt at 450 °C) with 12 ml of 1:10 dichloromethane/hexane. The collected extract was evaporated to approximately 0.1 ml and quantitatively transferred to a GC vial, evaporated to 100 µl, and spiked with ^{13}C-labelled PCB-159 volume correction standard. See Supporting Information for details on instrument analysis.

183 *PFAS analysis*

184   1–1.5 g of pre-weighed homogenized egg material was extracted using 8 ml acetonitrile,

185 while 30 g of vegetation was extracted using ca. 40 ml of methanol following methods described

186 previously.[17] Egg and vegetation extracts were evaporated to 2 ml and 1.5 ml respectively. Prior

187 to extraction, all samples were spiked with 0.5 ng/µl of ^{13}C-labelled internal standards: PFBA,

188 PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnDA, PFDoDA, PFTeDA, PFBS, PFHxS,

189 PFOS, PFOSA, 6:2 FTS and 8:2 FTS. Prior to quantification, each 0.5 ml of solution was spiked

190 with 2 ng of 3,7-brPFDcA recovery standard and 0.1 ml was transferred to an autoinjector vial

191 containing 0.1 ml of 2 mM NH_4OAc in HLB-water. Full details of the instrumental analysis are

192 described elsewhere.[43] Ten µl of extract was used to separate and analyses PFASs by ultrahigh

193 pressure liquid chromatography triple-quadrupole mass-spectrometry (UHPLC-MS/MS). Data

194 quantification was conducted with LCQuan software (Thermo Scientific). Unless specified, all

195 PFASs refer to linear isomers.

196

197 *Hg analysis*

198   Total mercury was analyzed by atomic absorption spectrometry using a Direct Mercury

199 Analyzer (DMA-80, Milestone). Approximately 0.03 g of freeze-dried egg homogenate was

200 analyzed. Samples were analyzed in parallel with sample blanks and certified reference material

201 (DORM-4 fish protein; DOLT-5 dogfish liver, National Research Council Canada). Samples

202 were analyzed in at least duplicate to ensure precision of measurements. Average recoveries of

203 the certified reference materials were within 10% of the reported values. The detection limit of

204 the instrument was 0.05 ng mercury.

205

206 *Quality assurance/control*

207   Concentrations reported for PCBs, HCB and PFASs were blank corrected based on the

208 average concentration detected within blank samples. Limits of detection (LOD) and

209 quantification were calculated as three and ten times the standard variation within blank

210 samples, respectively. LOD for PCBs ranged from 0.001 to 0.012 ng/g wet weight (ww); HCB

211 was 0.026 ng/g ww; and PFASs from 0.015 to 0.100 ng/g ww (Table 1). PCB and HCB

212 concentrations were only reported for analytes that had a quantification/qualifier ion ratio

213 within 20% of the ratio determined within the quantification standard. Reference material for

214 PCBs and HCBs (Contaminated fish reference material, EDF-2525) and PFASs (Pike-perch,

215 QM03-2) were also extracted in conjunction with sample material to assess method

216 performance. Internal standard recoveries for PCBs in eggs ranged between 40% and 60%; and

217 PFASs between 50% and 73% in eggs, and from 16% to 165% in vegetation (Table S8,

218 Supporting Information).

219

220 **Data treatment and statistical analyses**

221 *Pollutant datasets*

222 We used two datasets for statistical analyses, including: 1) lipophilic compounds with 19

223 PCB congeners and HCB; and 2) protein-associated compounds with six PFAS compounds and

224 mercury. Individual pollutants were included in datasets if they were detected in 60% or more

225 of our egg samples, to maximize statistical information and reduce random noise from non-

226 detect samples (see Table S7, Supporting Information for pollutants excluded). When

227 individual concentrations of each pollutant across all samples fell below the LOD, we imputed

228 left-censored data by replacing missing values (53 values for PCB; 71 values for PFAS) with a

229 random number between 0 and the LOD assuming a beta distribution ($\alpha = 5$, $\beta = 1$). We also

230 calculated pattern or relative contribution of PCBs and PFASs, expressed as the proportion of

231 each PCB congener or PFAS family to the sum total (*e.g.* $[PCB_i]/\sum PCB$ or $[PFAS_i]/\sum PFAS$).

232 For PCBs, we also summed concentrations according to the number of chlorine atoms as well

233 as metabolic group (Tables S9–10, Supporting Information).

235 *Statistical analyses*

236   We analyzed pollutant concentrations and patterns in R v. 3.4.1.[44] Multivariate analysis and

237 visualization of data was conducted by Principal Component Analysis (PCA) within the *vegan*

238 package v. 2.4-4.[45] We transformed pollutant concentrations ($\log_{10} x$) to normalize distributions

239 and reduce heterogeneity and/or skewness. We explored absolute concentrations of PCBs,

240 HCB, PFAS and Hg, as well as relative concentrations (*i.e.* patterns) of PCBs and PFASs by

241 PCA. Biological variables, which included hatch date, egg size (length), embryonic age, and

242 values of $\delta^{13}C$ and $\delta^{15}N$, were projected on the ordination space as passive variables. We

243 conducted a redundancy analysis (RDA) on both datasets in order to summarize the explanatory

244 power of relevant explanatory or biological variables, and quantified the percentage of variation

245 explained by each variable. Biological variables in our RDA included hatch date, egg size

246 (length), embryonic age, values of $\delta^{13}C$ and $\delta^{15}N$ and lipid and protein content. Hatch date was

247 positively correlated with lipid content (Pearson's R = 0.29, P = 0.02), and PCB concentrations

248 also increased with later hatch date, independent of lipid content (see Results and Discussion).

249 This prompted us to conduct partial RDA (pRDA) by treating PCB and HCB concentrations on

250 wet weight basis and lipid content as a covariable. A pRDA fits the biological variables to the

251 residual variation that is not attributable to the covariables.[46] The relationship between

252 significant biological variables and pollutant concentrations are depicted using linear

253 regressions. Unless specified, PCB, HCB and PFAS concentrations are reported on wet weight

254 (ww), and mercury on dry weight (dw) basis.

255

256 **Results and Discussion**

257 **Breeding population of Storholmen**

In 2016, a total of 272 breeding pairs were registered on the Storholmen island breeding colony. Nest hatching commenced 10 June and concluded 20 July, with a peak hatch date between 24 and 25 June (Figure 1). Hatching dates of the subsampled population (N = 61 pairs) were similar to the colony as a whole, with a peak hatch date of 25 June (range: 18 June to 20 July; Figure 1; Table 1).

The spread in hatch dates is twice as large as in 1993 and 1994 (range = 15 days),[47] as well as in 2006 and 2007 (range = 15 days).[13] This new timeframe suggests that geese are responding to a warmer climate, due to increased availability of resources at the staging areas in northern Norway and Svalbard breeding grounds, and has resulted in a broadening of the time window for reproduction. The mean hatch date for the Svalbard population has also advanced by approximately one week since the 1990s,[39,47] and 2016 represented the earliest hatch date on record for the island population.

From the 61 nesting pairs which eggs were sampled, at least one individual from 23 nesting pairs was resighted at the staging areas in northern Norway previously in the same breeding season (Vesterålen N = 18; Helgeland N= 5), meaning that we could not account for the migratory behavior of the remaining geese. It is likely that several non-sighted geese utilized staging areas in northern Norway before arriving in Svalbard, but were either not observed during the sighting period or were feeding outside sighting areas. Lipid content in eggs was $17.0 \pm 2.4\%$, and was 2-5 times greater than protein content ($4.9 \pm 1.0\%$; Table 1). Lipid content in eggs also increased with hatch date, which was contrary to expectation. We expected that earlier arriving females would utilize stored body reserves, resulting in increased lipid availability during egg production. Instead, later arriving females were remobilizing a greater proportion of lipids, which could be due to differences in foraging behavior for geese that

283   migrate late to the Svalbard breeding grounds,[7] or energetic differences in vegetation along the

284   migration route.[48] We found no relationships between all other biological variables measured

285   (Tables S2–4, Supporting Information).

286

287   **Spatial contribution of resources for egg production**

288   Stable isotope signatures in vegetation overlapped between the wintering, staging and island

289   colony sites (Figure 2). Vegetation sampled from cliff tundra contained lower $\delta^{13}C$ and higher

290    $\delta^{15}N$ values compared to all other sites ($\delta^{13}C$: t-test: $t_{13} = -2.75$, $P = 0.02$; $\delta^{15}N$: t-test: $t_{13} = 4.26$,

291    $P < 0.01$; Figure 2). We expected to find a unique isotopic composition along the flyway of the

292   goose following a previous study on goose droppings collected from each site.[13] However,

293   stable isotope signatures between vegetation and droppings may not be comparable given

294   fractionation between diet and droppings takes place during digestion.[49,50]

295

296   Egg stable isotope signatures ($\delta^{13}C = -28.4 \pm 0.8$ ‰, range: -30.3, -26.6; $\delta^{15}N = 10.1 \pm 1.7$

297   ‰, range: 7.7, 19.4; Figure 2) were unrelated to lipid and protein content or sighting of

298   individuals in northern Norway (Tables S4–6, Supporting Information). Values of $\delta^{13}C$ in eggs

299   increased with later hatch date (Pearson's $R = 0.30$, $P = 0.02$; Figure 3b), but $\delta^{15}N$ values did

300   not. $\delta^{13}C$ and $\delta^{15}N$ values were higher in eggs compared to vegetation from all sites (t-test: $\delta^{13}C$

301    $t_{72} = -2.75$, $P < 0.001$; $\delta^{15}N$ $t_{68} = 6.15$, $P < 0.001$), except for cliff tundra where $\delta^{15}N$ was higher

302   than in eggs (t-test: $t_{61} = -6.54$, $P < 0.001$; Figure 2).

303

304   Carbon and nitrogen isotope signatures in the eggs of barnacle geese from Storholmen Island

305   were similar to a neighboring island colony in 2006 and 2007.[13] The high $\delta^{13}C$ values in eggs

306   compared to vegetation suggest that geese also utilize stored body reserves for egg production

307   such as breast muscle and abdominal fat, which is often enriched in ^{13}C.[11,29] A previous study

308   on Greater Snow Geese (*Chen caerulescens atlantica*) found a strong positive relationship

309   between $\delta^{13}C$ values in maternal storage tissues and eggs,[29] suggesting that an increasing

310   reliance on stored body reserves corresponds to an enriched ^{13}C signal in eggs.

311

312      $\delta^{15}N$ values in eggs did not change throughout the breeding season, suggesting that most

313   females either did not utilize local Svalbard resources from bird cliff in 2016, or utilized this

314   resource in similar proportions. Due to an overlapping $\delta^{13}C$ signal across most sites, we could

315   not calculate the contribution of resources towards egg production under a stable isotope mixing

316   model.[51] A previous model has shown that the Svalbard goose population can allocate 50% of

317   resources from vegetation in the UK and northern Norway for egg production, assuming a

318   limited number of sites along the flyway.[13] However, reliance of resources from UK and

319   northern Norway decreases with later egg laying date; and this relationship has only been

320   observed in the lipid-free yolk component of eggs.[13] Even though we measured stable isotopes

321   in whole eggs, the positive relationship between $\delta^{13}C$ signal in eggs and nest hatch date

322   remained, suggesting that egg energy source varies throughout the breeding season.

323

324   **Low levels of pollutants in vegetation**

325   Low concentrations of HCBs were detected for vegetation at all sites, but PCBs were not

326   (Solway Firth: 0.02 ± 0.01 ng/g ww, N = 2; Vesterålen: 0.05 ± 0.03 ng/g ww, N = 3; Svalbard

327   cliff tundra: 0.09 ± 0.04 ng/g ww, N = 4; Svalbard island tundra: 0.03 ± 0.02 ng/g ww, N = 2).

328   PFASs were detected at cliff tundra (0.03 ng/g ww, N = 1), but not on the Solway Firth (N =

329   1). We could not sample a larger quantity of vegetation across all sites due to intensive goose

330   grazing activity, meaning these values should be treated with caution.

331

332   **Low levels of lipophilic pollutants in eggs**

333   Total PCB concentrations in eggs ranged between 0.46 and 4.42 ng/g ww (Table 1). PCB-

334   153 accounted on average for 32% of the total PCB concentration in eggs, followed by PCB-

335   118 (16%), PCB-138 (12%) and PCB-180 (10%). HCB was the dominating chemical in all

336   eggs, where concentrations ranged between 0.99 and 5.65 ng/g ww (Table 1).

337

338   Average concentrations of PCBs and HCB in barnacle goose eggs indicate low levels of

339   exposure in adult female geese. Pollutant concentrations are several orders of magnitude lower

340   than in eggs of piscivorous and predatory Arctic seabird species.[52,53] The low concentrations in

341   goose eggs reflects a terrestrial diet, and is similar to levels in other Arctic terrestrial species

342   occupying low trophic levels including caribou (*Rangifer tarandus*) and hare (*Lepus*

343   *arctica*).[54,55] Average lipid normalized concentrations of PCBs and HCB in eggs from this study

344   ($\sum_{12}$PCB = 7.2 ng/g lipid weight; HCB = 14.4 ng/g lw) are lower than in eggs from a

345   neighboring Svalbard barnacle goose colony sampled in 2006 ($\sum_{12}$PCB = 53.5 ng/g lw; HCB =

346   27.4 ng/g lw; n = 6).[56] The temporal decrease in PCB and HCB concentrations in biota is also

347   consistent with decreasing trends in air and monitoring data.[57,58]

348

349   **Effect of migration on PCB and HCB in eggs**

350   With later hatching date, both lipid content and wet weight concentrations of PCB in eggs

351   increased (Figures 3a and 3c). Hatch date contributed to 10.9% of the total variation in wet

352   weight pollutant concentrations (RDA$_{\text{Hatch date}}$: $F_{1,54} = 6.62$, $P = 0.001$) and 6.9% when lipid

353   content was treated as a covariable (pRDA$_{\text{Hatch date}}$: $F_{1,53} = 4.37$, $P = 0.01$; Figure S3, Supporting

354   Information). HCB contributed little to pollutant variation across eggs (Figure 3d). When

355   exploring differences in PCB patterns across eggs, hatch date explained 4% of the total variation

356   in PCB patterns (RDA$_{\text{Hatch date}}$: $F_{1,54} = 2.26$, $P = 0.05$). The relative contribution of tri- and tetra-

357   chlorinated PCBs to the total PCB load was higher in late hatching eggs (75th percentile = 8.3

± 4.3%) than in early hatching eggs (25th percentile = 6.8 ± 1.5%; $RDA_{Hatch\ date}$: %variation = 6.7; $F_{1,54}$ = 3.86, P = 0.02; Figure S4, Supporting Information). We also found a weak relationship between the enrichment of ^{13}C and increasing concentrations of tri- and tetra-chlorinated PCBs in eggs ($RDA_{\delta 13C}$: %variation = 4.5; $F_{1,54}$ = 2.54, P = 0.07). Hatch date was unrelated to substitution patterns of PCBs when arranged by metabolic group.[59]

The finding that absolute PCB concentrations were higher in late hatching eggs (75th percentile = 1.74 ± 0.66 ng/g ww) than in early hatching eggs (25th percentile = 1.06 ± 0.15 ng/g ww) was contrary to our expectations. We expected the earliest hatching eggs to contain the highest concentrations of PCBs, as these represent females that arrive at the Svalbard breeding grounds prior to snowmelt,[60] thereby relying on resources from wintering grounds, staging areas, cliff tundra and/or stored body reserves for egg production. However, HCB concentrations in eggs did not change throughout the breeding season, suggesting that consumption of Svalbard resources was similar across females, assuming that HCB concentration in vegetation increases at higher latitudes.[61,62]

Late hatching eggs may instead represent a small number of females that delay their departure from the wintering grounds by several weeks, skip or have a very brief stopover at staging areas in northern Norway. The exact proportion of individuals that utilize this strategy is unclear, but these females arrive later or around the same time as individuals that utilize staging areas in northern Norway.[33-35] In addition, the pre-nesting period between arrival at the breeding grounds and egg laying may be shorter for late arriving females than early ones.[31] Thus, late arriving females may rely more on overwintering ground resources instead of breeding ground resources. Other relevant biological variables that could contribute to the total variation in pollutant occurrence across eggs include timing of departure from the overwintering

383   grounds, duration spent at staging areas, timing of arrival at the breeding grounds, and

384   proportion of resources utilized at different sites.

385

386   When breeding females utilize stored body reserves for egg production, PCBs becomes

387   remobilized and translocated within the body. The rate of diffusion depends on chlorine atom

388   placement and degree of the chlorination for a given PCB congener. For example, less

389   chlorinated PCBs translocate more quickly from stored body reserves than more chlorinated

390   PCBs due to their lower lipophilicity (*i.e.* lower K_{ow}).[63-65] Indeed, we observed the latest

391   hatching eggs to contain a significantly higher relative contribution (1.5% greater) of tri- and

392   tetra-chlorinated PCBs compared to the total PCB load when compared to the earliest hatching

393   eggs. The substitution pattern of PCBs should also affect their diffusion rates,[66] however this

394   pattern was similar across all eggs.

395

396   The body condition of females offers an alternative explanation for the higher lipid content

397   and PCB concentrations in late hatching eggs. Females that arrive late at the breeding grounds

398   may be in poorer body condition, and thus will depend more on stored body reserves (*e.g.* lipids)

399   to maintain body condition.[7] A remobilization of lipids will thus lead to increased circulating

400   levels of pollutants in blood,[67] thereby increasing the potential for pollutants to be transferred

401   during egg production. Additionally, a greater reliance on distant resources may result in

402   exposure to higher concentrations of PCBs, as these areas are closer to potential point sources

403   of pollution compared to remote polar regions.[23,24] The high lipid content and PCB

404   concentration in late hatching eggs is likely to be due to a combination of factors, including

405   females foraging predominantly at distant overwintering grounds, followed by the direct flight

406   to the Svalbard breeding grounds resulting in a greater reliance on stored body reserves and/or

407   poorer body condition. However, we were unable to assess the exact contribution of each of

408 these factors to the overall pollutant profile measured in eggs, and this uncertainty warrants

409 future research. This could include the use of tracking devices to determine each individual's

410 migration schedule.[68]

411

412 **Similar PFASs and Hg occurrence across eggs**

413 Total detectable PFAS concentrations in eggs ranged between 0.05 and 17.7 ng/g ww (Table

414 1). When detectable, linear-PFOS on average accounted for 29% of the total PFAS

415 concentration in eggs, followed by: PFUnDA (25%), PFNA (13%), PFTriDA (11%), PFDcA

416 (9%) and PFDoDA (8%). We detected mostly long-chained perfluorinated carboxylates

417 (PFCAs) in eggs, which are also common in other bird species and the marine ecosystem in

418 general.[69] Total mercury concentrations in eggs ranged between 9.76 and 40.99 ng/g dw (Table

419 1).

420

421 Occurrence of PFAS and Hg in relation to the protein content of eggs did not change

422 throughout the breeding season (Figures 3e–f), supporting our expectation that migration

423 strategy has a greater effect on pollutants associated with lipids than proteins. Proteins may

424 serve as a limiting resource during egg formation,[29,70] and energetic costs of transporting stored

425 protein during migration may be greater than for lipids.[11] Thus, the acquisition and allocation

426 of PFASs and Hg towards egg production should be limited by similar mechanisms.

427 Alternatively, a similar PFAS or Hg signal across eggs may be due to similar exposure profiles

428 at each site along the flyway. For example, fractionation of PFASs generally does not occur

429 along latitudinal gradients,[71] as the chemicals are mainly transported through oceanic currents.[72]

430 This could be validated by future or increased sampling efforts of vegetation at each site along

431 the migration route.

432

The present study reveals differences in exposure profiles of eggs of herbivorous geese, which may be a consequence of different migration strategies. Eggs laid later in the breeding season contained higher concentrations of PCBs. Barnacle geese are also responding to a warming climate by arriving earlier at the breeding grounds, which can affect the optimal timing between departure from overwintering grounds, arrival at the breeding grounds and peak food quality.[68] A shift in timing may also lead to a change in reproductive success of Arctic-breeding goose populations,[36] which may lead to further changes in the exposure profile of eggs. Recent evidence shows some polar bears (*Ursus maritimus*) have shifted their summer diet, which includes increased consumption of goose eggs.[73] This will not only impact the reproductive success the barnacle goose populations, but may cause changes in the distribution pollutants across Arctic food webs. The study of pollutants as chemical tracers in Arctic migrants yields insights into potential energy sources utilized during offspring production. Our study concerned an herbivorous migrant, and we expect stronger relationships for organisms that feed at higher trophic levels, where the effects of migration or reproductive strategies may become more apparent.

Supporting Information

The Supporting Information is free of charge on the ACS Publication website at DOI: xx.xxx/acs.est.xxxxxxx.

Schematic of migration timing and route of the barnacle goose (Figure S1); description of vegetation sampled (Table S1); summary statistics of relationships between measured biological variables (Tables S2–S6); summary of pollutant datasets (Tables S7); pollutant recoveries (Table S8); classification of PCBs according to chlorination and metabolic group (Tables S9–S10 and Figure S2); summary statistics of multivariate analyses (Tables S11–S14 and Figures S3–S6).

458

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675

676　**Table 1.** Biological and pollutant information (PCB, HCB, PFAS and Hg) in barnacle goose

677　eggs sampled on Svalbard in 2016. Unless specified, estimates refer to the egg contents.

Biological variable		Min-max	N	Mean ± SD
Mass (g)	Whole egg:	82.0–113.3	61	98.4 ± 7.1
	Content:	71.0–101.6		87.8 ± 6.6
Whole egg size (mm)	Length:	67.6–92.5	61	76.2 ± 4.2
	Width:	47.2–55.2		50.3 ± 1.6
Embryo age (d)		0–23	61	13.3 ± 5.7
Nest hatch date		18 June–20 July	61	28 June ± 5.6 days
Lipid (%)		10.6–26.0	59	17.0 ± 2.4
Water content (%)		60.2–72.7	61	68.3 ± 1.6
Protein (%)		2.8–8.7	50	4.9 ± 1.0

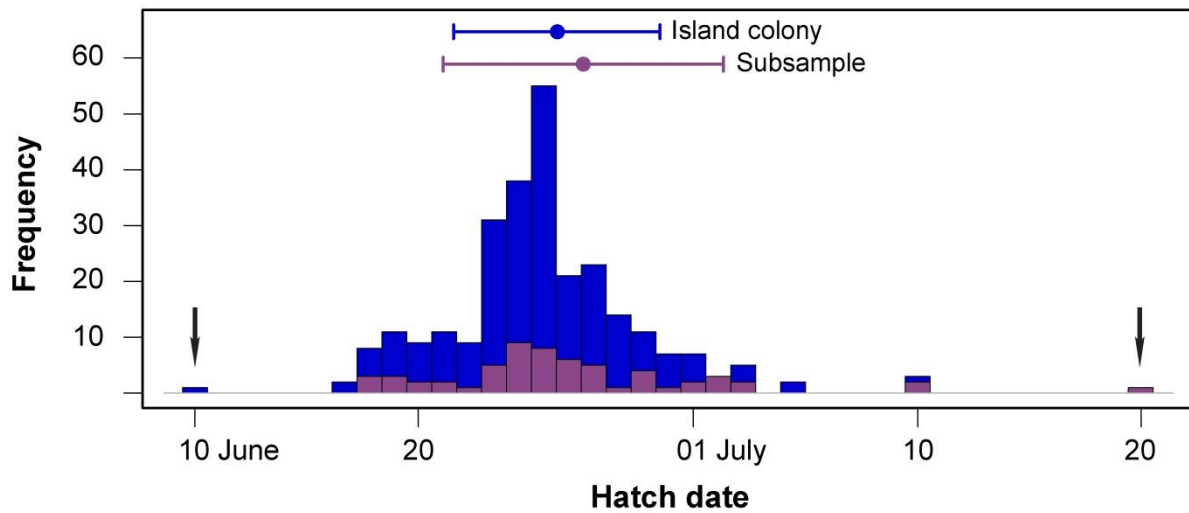| Pollutant	LOD	% detected[*]	Min-max	Median[‡]	Mean ± SD
PCBs (ng/g ww, N = 58)					
PCB-28/31	0.003	96	<LOD–0.025	0.009	0.010 ± 0.003
PCB-52	0.005	93	<LOD–0.044	0.008	0.010 ± 0.006
PCB-47	0.003	72	<LOD–0.008	0.004	0.005 ± 0.001
PCB-37	0.001	98	<LOD–0.401	0.013	0.023 ± 0.054
PCB-74	0.002	100	0.010–0.262	0.023	0.031 ± 0.035
PCB-66	0.003	98	<LOD–0.037	0.013	0.014 ± 0.006
PCB-101	0.004	70	<LOD–0.031	0.005	0.008 ± 0.006
PCB-99	0.001	100	0.005–0.101	0.022	0.028 ± 0.019
PCB-149	0.004	98	<LOD–0.040	0.010	0.011 ± 0.006
PCB-118	0.003	70	<LOD–1.104	0.127	0.197 ± 0.176
PCB-153	0.012	100	0.139–1.559	0.335	0.402 ± 0.244
PCB-105	0.002	91	<LOD –0.338	0.044	0.068 ± 0.058
PCB-138	0.010	100	0.056–0.422	0.126	0.150 ± 0.087
PCB-187	0.003[×]	100	0.029–0.156	0.058	0.066 ± 0.025
PCB-183	0.003[×]	100	0.009–0.076	0.022	0.025 ± 0.013
PCB-180	0.002	100	0.041–0.362	0.113	0.129 ± 0.064
PCB-170	0.007[×]	100	0.025–0.199	0.054	0.060 ± 0.033
PCB-194	0.007[×]	100	0.008–0.055	0.016	0.018 ± 0.008
PCB-209	0.007[×]	65	<LOD–0.017	0.010	0.010 ± 0.003
ΣPCB			0.462–4.418	1.006	1.227 ± 0.800
HCB	0.026	100	0.987–5.647	2.368	2.364 ± 0.697
PFASs (ng/g ww, N = 59)					
PFHpS	0.035	3	<LOD–0.289	0.268	0.268 ± 0.030
branched-PFOS	0.070	7	<LOD–4.247	2.114	2.195 ± 2.089
linear-PFOS	0.070	63	<LOD–11.304	0.314	0.930 ± 2.319
PFNS	0.065	5	<LOD–0.509	0.436	0.456 ± 0.046
PFNA	0.015	81	<LOD–0.326	0.062	0.080 ± 0.060
PFDcA	0.015	81	<LOD–0.367	0.058	0.078 ± 0.067
PFUnDA	0.015	98	<LOD–0.954	0.127	0.167 ± 0.153
PFDoDA	0.015	81	<LOD–0.230	0.056	0.066 ± 0.040
PFTriDA	0.020	75	<LOD–0.261	0.098	0.109 ± 0.055
PFTeDA	0.020	32	<LOD–0.110	0.054	0.053 ± 0.022
ΣPFAS			0.054–17.690	0.539	1.209 ± 2.972
Hg (ng/g dw, N = 61)	0.050	100	9.760–40.990	18.520	20.170 ± 7.520

678　Min = minimum; Max = maximum; N = sample size; SD = standard deviation; LOD = limit
679　of detection; ww = wet weight; dw = dry weight. [*]Percentage of eggs quantified above the
680　LOD. [‡]Calculated using values above LOD. [×]Values represent limits of quantification (LOQ).

Figure legends

Figure 1. Histogram of nest hatching dates of barnacle geese breeding on Storholmen Island in 2016 (N = 272) and the subsampled population in the present study (N = 61). Arrows indicate the earliest and latest hatching dates. Mean ± SD above the plots.
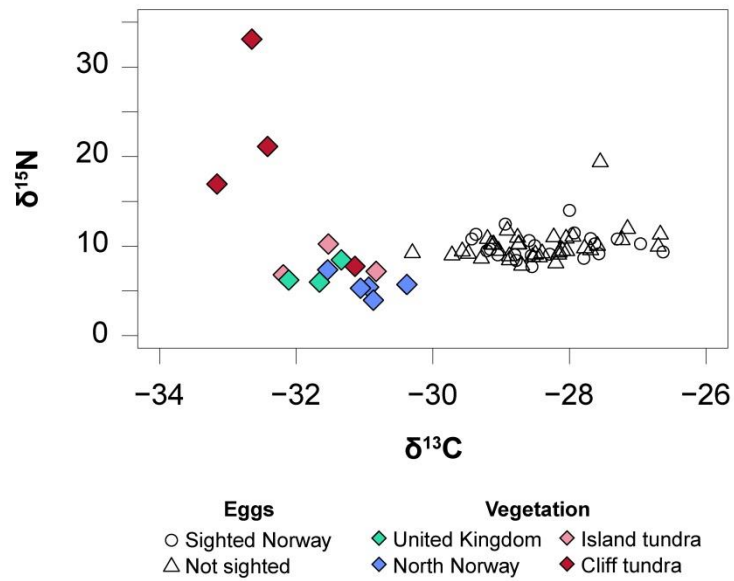
Figure 2. Stable isotope composition of $\delta^{13}C$ and $\delta^{15}N$ in eggs of barnacle geese sampled on Svalbard (N = 59) in 2016, as well as vegetation collected along the flyway including United Kingdom, northern Norway and Svalbard island and cliff tundra (N = 15) the same year. Circles represent eggs of geese sighted in Norway; triangles not sighted. Vegetation is denoted by diamonds.

Figure 3. Relationship between nest hatch date and: a) lipid content (%) ($R^2 = 0.07$, P = 0.02); b) $\delta^{13}C$ ($R^2 = 0.09$, P = 0.02); c) $\sum$PCB concentration (points and solid line on wet weight, $R^2 = 0.18$, P < 0.01); dashed line on lipid weight; $R^2 = 0.11$, P < 0.01); d) HCB concentration; e) $\sum$PFAS concentration; and f) mercury concentration in eggs of barnacle geese sampled on Svalbard in 2016. Circles represent eggs of geese sighted in Norway; triangles not sighted. Linear regressions presented when relationships were significant.
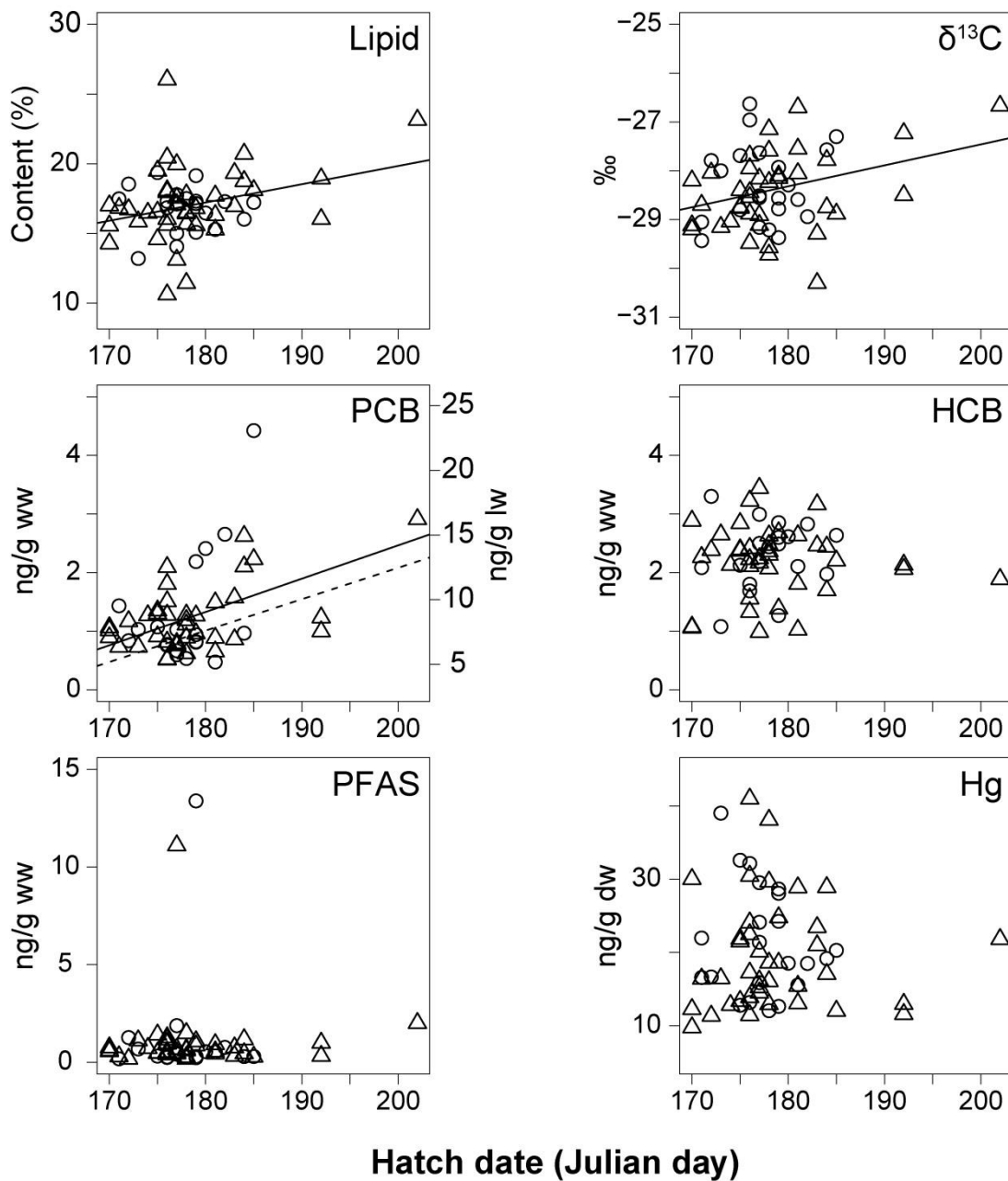
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