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1 Serum POP concentrations are highly predictive of inner blubber concentrations at
2 two extremes of body condition in northern elephant seals
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ABSTRACT

28 Long-lived, upper trophic level marine mammals are vulnerable to bioaccumulation of persistent
29 organic pollutants (POPs). Internal tissues may accumulate and mobilize POP compounds at
30 different rates related to the body condition of the animal and the chemical characteristics of
31 individual POP compounds. Collection of samples from multiple tissues is a major challenge to
32 ecotoxicology studies of free-ranging marine mammals, yet the ability to predict POP
33 concentrations in one tissue from another tissue remains rare. Northern elephant seals (*Mirounga*
34 *angustirostris*) forage on mesopelagic fish and squid for months at a time in the northeastern
35 Pacific Ocean, interspersed with two periods of fasting on land, which results in dramatic
36 seasonal fluctuations in body condition. Using northern elephant seals, we examined commonly
37 studied tissues in mammalian toxicology to describe relationships and determine predictive
38 equations among tissues for a suite of POP compounds, including Σ DDTs, Σ PCBs, Σ chlordanes,
39 and Σ PBDEs. We collected paired blubber (inner and outer) and blood samples from adult
40 female and male seals in 2012 and 2013 at Año Nuevo State Reserve (California, USA). For
41 females (N = 24), we sampled the same seals before (late in molting fast) and after (early in
42 breeding fast) their approximately seven month foraging trip. For males, we sampled different
43 seals before (N = 14) and after (N = 15) their approximately four month foraging trip. We
44 observed strong relationships among tissues for many, but not all compounds. Serum POP
45 concentrations were strong predictors of inner blubber POP concentrations for both females and
46 males, while serum was a more consistent predictor of outer blubber for males than females. The
47 ability to estimate POP blubber concentrations from serum, or vice versa, has the potential to
48 enhance toxicological assessment and physiological modeling. Furthermore, predictive equations
49 may illuminate commonalities or distinctions in bioaccumulation across marine mammal species.

50 **Keywords:** pinniped, marine mammal, tissue-correlation, predictive-equations, fasting,
51 partitioning coefficients

52 **Capsule:** Relationships among serum and blubber layers in elephant seals, after fasting and
53 foraging periods, can enhance contaminant studies and enable comparisons among species.

54

55

HIGHLIGHTS

- 56 • Serum was a strong predictor of inner blubber for DDTs, PCBs, CHLs, and PBDEs
- 57 • Relationships between serum and outer blubber were stronger for males than females
- 58 • Higher blubber/serum partitioning coefficients in early breeding than late molting
- 59 • Predictive equations provided for seals at two extremes of body condition

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INTRODUCTION

63 Persistent organic pollutants (POPs) are lipophilic environmental contaminants that are
64 pervasive in marine food webs and bioaccumulate in organisms, which presents particular
65 concern for long-lived, upper trophic level marine mammals. Although close proximity to POP
66 sources can result in higher POP concentrations (Frouin et al. 2011), even foraging strategies that
67 place animals far from contaminant sources do not insulate them from POP exposure (Peterson et
68 al. 2015). Elevated concentrations of POPs are associated with endocrine, immune, and
69 reproductive effects in marine and terrestrial wildlife (Tanabe 2002, Debier et al. 2005,
70 Desforges et al. 2016). Among marine mammals, pinnipeds and odontocetes are vulnerable to
71 biomagnification of POPs due to their high trophic position, and therefore are often the target
72 species for POP biomonitoring efforts (Weijs et al. 2010, Yordy et al. 2010b, Lopez et al. 2012).

73 The internal tissues of marine mammals accumulate and mobilize POPs at varying rates,
74 which presents challenges for interpretation. Blood and blubber are commonly studied tissue
75 compartments because they can be sampled non-lethally and are relatively accessible to
76 researchers. Blood is advantageous for study because it is in direct contact with internal tissues
77 of toxicological concern, including the liver and other organs, and it is responsive to recent
78 foraging (De Swart et al. 1996) or fasting (Louis et al. 2014). Therefore, blood can serve as a
79 relevant indicator of recent contaminant exposure or a reflection of changes in physiological
80 state that may liberate contaminants from storage tissues into circulation. Blood collection from
81 live pinnipeds is generally less invasive than blubber collection, and many studies may store
82 blood samples long term, often the serum or plasma compartments, that could be utilized to
83 compare contaminant exposure over time; however, the relationship between blood and blubber
84 layers may be inconsistent (Lydersen et al. 2002). In contrast, blubber is a lipid-rich tissue used
85 for energy storage in both pinnipeds and odontocetes (Koopman et al. 1996, Strandberg et al.
86 2008). Within the vertical profile of the blubber layer, metabolic activity and fatty-acid
87 mobilization vary, with inner blubber more metabolically active than outer (Strandberg et al.
88 2008, Fowler et al. 2014). While recent events such as foraging or fasting may impact
89 contaminant concentrations in blood and inner blubber (Louis et al. 2014), outer blubber or full
90 thickness blubber cores may provide a more relevant indicator of longer-term bioaccumulation
91 (Randhawa et al. 2015). Blubber is often collected in pinnipeds and cetaceans, and in some cases
92 it is the only tissue available to study (*e.g.*, Barón et al. 2015, Hunt et al. 2015).

93 Northern elephant seals (*Mirounga angustirostris*) are upper trophic level predators that
94 bioaccumulate POPs as they forage in the northeastern Pacific Ocean (Peterson et al. 2015). The
95 life history of northern elephant seals includes two foraging migrations per year interspersed

96 with fasting periods for breeding and molting on land, which makes them relatively accessible
97 for study among marine mammals (Robinson et al. 2012). Additionally, foraging and fasting life-
98 history phases create dramatically different seasonal body conditions. For example, during the
99 molting fast females spend roughly 4-6 weeks fasting and lose approximately 25% of their mass,
100 of which 41% comes from fat stores (Worthy et al. 1992). Females then go to sea and forage for
101 approximately 7 months, during which time they undergo a 95% mean mass gain (Robinson et
102 al. 2012). At the end of their foraging trip, females return to land and undergo a breeding fast,
103 lasting approximately 4-6 weeks, which occurs as they give birth to a pup and lactate, and results
104 in loss of approximately 40% of their body mass and 57% of their fat stores (Costa et al. 1986,
105 Crocker et al. 2001). After breeding, females once again return to sea for a shorter 2-3 month
106 foraging trip, which ends when they return to land for molting (Robinson et al. 2012). Males
107 undergo a similar proportional loss of body mass (34-41%) and fat stores (54-59%) while on
108 shore for fasting (Deutsch et al. 1990, Crocker et al. 2012). The male breeding fast is longer than
109 females, lasting approximately 2-3 months, while the male molting fast is approximately 4-6
110 weeks in duration (Le Boeuf et al. 2000). The two male foraging trips at sea each year are similar
111 to each other in duration, lasting approximately 4 months each (Le Boeuf et al. 2000). Because
112 body condition is an important determinant of POP concentrations in blood and blubber
113 (Peterson et al. 2014), effective toxicological risk assessment relies upon understanding POP
114 concentrations at extremes in body condition.

115 The naturally occurring extremes of body condition of northern elephant seals present an
116 opportunity to examine how body condition impacts the relationships of contaminant
117 concentrations among tissues. We examined commonly studied tissues with higher (serum, inner
118 blubber) and lower (outer blubber) metabolic activity to describe relationships and determine

119 predictive equations among tissues for POPs in northern elephant seals. Our primary focus was
120 to investigate relationships for polychlorinated biphenyls (PCBs),
121 dichlorodiphenyltrichloroethane (DDT) and metabolites of DDT, chlordanes (CHLs), and
122 polybrominated diphenyl ethers (PBDEs), although we also included hexachlorobenzene (HCB),
123 hexachlorocyclohexane (α -HCH and β -HCH), and the naturally produced 6-MeO-BDE 47.
124 Specifically, our objectives were to 1) assess serum POP concentrations as a predictor of inner
125 and outer blubber POP concentrations, and inner blubber POP concentrations as a predictor of
126 outer blubber POP concentrations at two body condition extremes, 2) compare serum to blubber
127 relationships between males and females, and 3) use partitioning coefficients to compare
128 blubber/serum POPs and inner blubber/outer blubber POPs relationships between females and
129 males at two naturally occurring extremes of elephant seal body condition.

130

131 METHODS

132 *Animal Sampling*

133 We collected paired blubber and blood samples from adult female and male northern
134 elephant seals in 2012 and 2013 at Año Nuevo State Reserve (California, USA, 37.11° N,
135 122.33° W). The same known-age females (N = 24), ranging in age from four to twelve years,
136 were sampled before (late in the molting fast) and after (early in the breeding fast) an
137 approximately seven month long foraging trip. Due to the challenges associated with repeatedly
138 sampling males, blubber cores and blood samples were collected from 29 unique male northern
139 elephant seals at two points in their life history: 14 seals were sampled at the end of the molting
140 fast and 15 seals were sampled approximately four months later at the start of the breeding fast.

141 We used standard procedures for chemical immobilization and collection of blubber and

142 blood from northern elephant seals (Le Boeuf et al. 2000, Robinson et al. 2012), and these
143 procedures have been described previously in Peterson et al. (2015). In brief, a full-thickness
144 blubber core was collected from the lateral pelvic area of each seal from a sterile scalpel incision
145 with a 6 mm biopsy punch (Miltex, Inc., York, Pennsylvania, USA) and stored in aluminum foil.
146 Blood samples were collected from the extradural vein and stored on ice in the field. Upon return
147 to the lab within several hours, samples were centrifuged and serum aliquots were transferred to
148 glass vials. Blubber cores and serum samples were stored at -20°C until analysis.

149

150 ***Laboratory analysis***

151 We targeted 29 PCB congeners (IUPAC numbers: CB 28, 49, 52, 74, 95, 99, 101, 105, 110,
152 118, 128, 138, 146, 149, 153, 156, 170, 171, 174, 177, 180, 183, 187, 194, 196/203, 199, 206,
153 and 209), seven PBDE congeners (IUPAC numbers: BDE 28, 47, 99, 100, 153, 154, and 183),
154 three DDTs (*p,p'*-DDD, *p,p'*-DDE, *p,p'*-DDT), five chlordanes (CHLs: OxC (oxychlordanes), CC
155 (*cis*-chlordanes), TC (*trans*-chlordanes), TN (*trans*-nonachlor), CN (*cis*-nonachlor)),
156 hexachlorobenzene (HCB), hexachlorocyclohexane (α -HCH, β -HCH), and the naturally-
157 produced methoxylated PBDE, 6-MeO-BDE 47, in all samples.

158 Extraction, clean-up, and concentration measurement methods for blubber and serum
159 followed protocols described in Vanden Bergh et al. (2012) and Peterson et al. (2015). In brief,
160 for blubber analyses, the skin layer and hair (Schwarz et al. 2015), was removed from the outer
161 portion of the biopsy core, and the remaining blubber layer was cut into inner and outer segments
162 of approximately equal mass. Inner and outer blubber segments were analyzed separately due to
163 differences in metabolic activity and stratification in fatty acid profiles among layers (Strandberg
164 et al. 2008, Fowler et al. 2014). Serum samples were split for separate determination of target

165 contaminants and lipids. Four lipid classes in serum (total cholesterol, phospholipids,
166 triacylglycerides, and non-esterified fatty acids) were determined with enzyme kits from Diasys
167 Diagnostic Systems (Holzheim, Germany) and Wako Chemicals (Neuss, Germany), with the
168 concentrations of each lipid class calculated on the basis of standard equivalents. Total lipid
169 concentrations were calculated as the sum of the four lipid classes (Debier et al. 2006, Vanden
170 Berghe et al. 2012). All POP concentrations in serum were lipid-normalized before statistical
171 analyses.

172 PBDEs, MeO-PBDEs, CHLs, HCB, and HCHs were measured by gas chromatography-
173 electron capture negative ion/mass spectrometry (GC-ECNI/MS) on a 30 m × 0.25 mm × 0.25
174 μm DB-5 column (J&W Scientific, Folsom, CA, USA) by monitoring two ions $m/z = 79$ and 81
175 (for PBDEs and MeO-PBDEs) and two specific ions for each pesticide. DDTs and PCBs were
176 measured by gas chromatography-electron ionization/mass spectrometry (GC-EI/MS) on a 25 m
177 × 0.22 mm × 0.25 μm HT-8 column (SGE, Zulte, Belgium) by monitoring 2 ions for each
178 homologue group.

179

180 *Quality control*

181 For quality control (QC), we randomly analyzed procedural blanks, solvent blanks, and
182 standards throughout the extraction process. Recoveries for individual PCB and PBDE congeners
183 ranged between 75 and 104% (RSD < 12%). For each analyte, the mean procedural blank value
184 was used for subtraction to determine final analyte concentrations. After blank subtraction, the
185 limit of quantification (LOQ) was set at 3 × SD of the procedural blank. For analytes that were
186 not detected in procedural blanks, LOQs were calculated for a ratio S/N (signal to noise) equal to
187 10. A standard reference material SRM 1945 (PCBs, OCPs, and PBDEs in whale blubber) was

188 used to test the accuracy of the method. Measured values did not deviate more than 15% from
189 the certified values.

190

191 *Statistical analysis*

192 We examined the relationship between each pair of tissues (inner blubber:serum, outer
193 blubber:serum, outer blubber:inner blubber) individually for each class of contaminants (Σ DDTs,
194 Σ PCBs, Σ CHLs, and Σ PBDEs), for individual contaminants (HCB, α -HCH, β -HCH), and the
195 naturally-produced 6-MeO-BDE 47, before elephant seals left for a foraging trip (late in the
196 molting fast), and upon return from foraging (early in the breeding fast). In order to quantify the
197 relationships between concentrations of POPs in paired tissues, we used general linear models in
198 the statistical program R, version 3.0.2 (R Development Core Team 2012). We first ran a global
199 model for each pair of tissue POP concentrations with sex as a factor and an interaction between
200 the predictor tissue POP concentration \times sex. If the interaction was significant, we conducted
201 subsequent analyses for each sex separately. All POP concentrations were natural-log
202 transformed prior to analysis to meet the assumptions of general linear models. When sex was
203 not a significant predictor, we removed the term for the predictive equation.

204 In addition, we calculated partitioning coefficients for outer blubber/serum, inner
205 blubber/serum, and outer blubber/inner blubber for each contaminant in female and male
206 elephant seals. Partitioning coefficients were calculated for each individual as the ratio between
207 concentrations in the two tissues. We used mixed effects models to compare partitioning
208 coefficients by sampling period (late molting, early breeding), sex, and a sex \times sampling period
209 interaction, with individual as a random effect. If the interaction was not significant, we removed

210 it and reran the model. We conducted post-hoc pairwise comparisons on least-squares means
211 with a Tukey adjustment on p -values. For statistical significance, α was set at $p = 0.05$.

212

213

214

RESULTS

215 Serum, inner blubber, and outer blubber samples from all elephant seals had detectable
216 concentrations of Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs. The concentrations of POPs used in
217 this study for northern elephant seal serum, inner blubber, and outer blubber have been reported
218 previously (Peterson et al. 2015), although a summary of medians and ranges of POP
219 concentrations can be found in Table 1. The specific equations to predict POP concentrations in
220 one tissue from POPs concentrations in another tissue for all compounds are found in Table 2
221 and specific statistical results are found in Table 3.

222

223 *Serum and inner blubber*

224 Late molting seals did not have a significant interaction between sex \times serum
225 concentrations of Σ DDTs, Σ PCBs, Σ CHLs, or Σ PBDEs on the concentrations of these POP
226 classes in inner blubber ($F \leq 1.80$, $p \geq 0.18$). Similarly, early breeding seals did not have a
227 significant interaction between serum POP concentrations \times sex on the POP concentrations in
228 inner blubber, except for PBDEs ($F_{1,35} = 6.45$, $p = 0.015$). Therefore, we removed the interaction
229 from all models except for early breeding PBDEs, where we analyzed males and females
230 separately.

231 Serum POP concentrations were positively related to inner blubber POP concentrations
232 for all major POP classes (Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs) both late in the molting fast

233 (Fig. 1) and early in the breeding fast (Fig. 2), while accounting for any potential effect of sex.
234 Early in the breeding fast, concentrations of Σ PBDEs in serum were positively related to
235 concentrations of Σ PBDEs in inner blubber for females and males. For Σ DDTs, Σ PCBs, and
236 Σ CHLs at both sampling periods, and Σ PBDEs at late molting, all relationships between serum
237 and inner blubber had R^2 values >0.7 , and relationships for all POP classes were stronger at early
238 breeding than at late molting (Table 2). In addition, the highest R^2 values at each life history
239 phase were found for Σ DDTs.

240 While Σ DDT, Σ PCB, Σ CHL, and Σ PBDE concentrations in serum were related to those
241 in inner blubber, we found varying relationships for the remaining POP compounds (Fig. 3; Fig.
242 4). During the late molt we did not detect any significant interactions between POP
243 concentrations in serum \times sex on POP concentrations in inner blubber for α -HCH, β -HCH, or
244 HCB ($F_{1,34} \leq 0.77$, $p \geq 0.35$). Serum concentrations were positively related to inner blubber
245 concentrations for α -HCH, β -HCH, and HCB. Males were excluded from tests for 6-MeO-BDE
246 47 due to low detectability of this compound in samples; no significant relationship between
247 serum and inner blubber was found for 6-MeO-BDE 47 in females (Fig. 3).

248 In early breeding, we did not detect a significant serum \times sex interaction for 6-MeO-BDE
249 47 ($F_{1,34} = 2.20$, $p = 0.15$) or β -HCH ($F_{1,34} = 0.11$, $p = 0.75$). For α -HCH, we detected a marginally
250 significant interaction ($F_{1,35} = 3.05$, $p = 0.09$) and analyzed female and male seals separately.
251 Serum concentrations of 6-MeO-BDE 47 and β -HCH were positively related to inner blubber
252 concentrations, while accounting for any potential effect of sex. For α -HCH we detected a
253 significant relationship between serum and inner blubber concentrations for females, but not
254 males. While the relationships between serum concentrations and inner blubber concentrations of
255 β -HCH at late molting and early breeding were strong ($R^2 \geq 0.58$), we observed weaker

256 relationships for α -HCH ($R^2 \leq 0.38$). In addition, there was no significant relationship between
257 serum and inner blubber for HCB, while accounting for sex.

258

259 *Serum and outer blubber*

260 We detected a significant interaction between POP concentrations in serum \times sex on POP
261 concentrations in outer blubber for Σ DDTs, Σ PCBs, and Σ CHLs during late molting ($F_{1,34} \geq 5.27$,
262 $p \leq 0.001$) and early breeding ($F_{1,35} \geq 7.48$, $p \leq 0.010$). In addition, we detected a serum \times sex
263 interaction in early breeding Σ PBDEs ($F_{1,35} = 12.58$, $p < 0.001$), but not late molting Σ PBDEs
264 ($F_{1,34} = 3.57$, $p = 0.067$). Therefore, we removed the interaction from the model for late molting
265 Σ PBDEs, but for all other POP classes we conducted separate statistical analyses on female and
266 male elephant seals.

267 For combined male and female seals late in the molting fast, concentrations of Σ PBDEs
268 in serum were positively related to concentrations of Σ PBDEs in outer blubber, when accounting
269 for sex. There was not a significant sex effect for Σ PBDEs, indicating that similar Σ PBDE
270 concentrations in serum corresponded to similar Σ PBDE concentrations in outer blubber for
271 males and females (Fig. 1). Early in breeding, serum Σ PBDE concentrations were positively
272 related to outer blubber concentrations for males and females. In addition, serum Σ DDT, Σ PCB,
273 and Σ CHL concentrations were positively related to outer blubber concentrations during both
274 sampling periods for females and males (Fig. 1; Fig. 2). Outer blubber:serum relationships were
275 stronger for males ($0.94 \geq R^2 \geq 0.79$) than females ($0.75 \geq R^2 \geq 0.23$) for Σ DDTs, Σ PCBs, Σ CHLs, and
276 Σ PBDEs at both sampling periods, but the differences between the sexes were more pronounced
277 for Σ PCBs and Σ CHLs at both periods than for Σ DDTs and Σ PBDEs (Fig. 1; Fig. 2). For
278 Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs, the slope of the relationship for males was steeper than

279 females, indicating that at higher serum concentrations individual males had proportionately
280 higher outer blubber contaminant concentrations than females (Table 2).

281 For 6-MeO-BDE 47, α -HCH, β -HCH, and HCB compounds late in the molting fast, we
282 detected a significant interaction between POP concentrations in serum \times sex on POP
283 concentrations in outer blubber for α -HCH, β -HCH, and HCB ($F_{1,34} \leq 4.59$, $p \geq 0.039$).
284 Additionally, only 7 males had detectable concentrations of 6-MeO-BDE 47 at late molt, and
285 therefore, were not included in analyses. Although serum concentrations of α -HCH, β -HCH, and
286 HCB were positively related to outer blubber in males (Fig. 3), only β -HCH concentrations were
287 related in females. Concentrations of 6-MeO-BDE 47 in female serum samples were not
288 significantly related to concentrations in outer blubber samples (Fig. 3).

289 In contrast with samples from late in the molting fast, during early breeding we did not
290 detect a serum \times sex interaction for 6-MeO-BDE 47, α -HCH, β -HCH, or HCB ($F_{1,35} \leq 1.44$,
291 $p \geq 0.24$). Serum concentrations of 6-MeO-BDE 47, α -HCH, and β -HCH were positively related
292 to outer blubber concentrations (Fig. 4), while accounting for any potential effect of sex. Serum
293 concentrations of HCB were not significantly related to outer blubber concentrations.

294

295 *Inner blubber and outer blubber*

296 We observed a significant interaction between POP concentrations in inner blubber \times sex
297 on Σ PCB and Σ CHL concentrations in outer blubber during late molting ($F_{1,34} \geq 18.37$, $p \leq 0.001$)
298 and Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs during early breeding ($F_{1,35} \geq 7.80$, $p \leq 0.008$). In
299 addition, we found no detectable interaction for Σ DDTs and Σ PBDEs during late molting
300 ($F_{1,34} \leq 2.32$, $p \geq 0.14$). Therefore, in the case of late molting, we removed the interaction from the

301 models for Σ DDTs and Σ PBDEs, and for Σ PCBs and Σ CHLs we analyzed females and males
302 separately.

303 Inner blubber POP concentrations were positively related to outer blubber POP
304 concentrations for Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs during both sampling periods, for both
305 females and males (Fig. 2). For late molting seals, concentrations of Σ DDTs and Σ PBDEs in
306 serum were significantly related to concentrations of Σ DDTs and Σ PBDEs in outer blubber,
307 while accounting for sex (Fig. 1). The significant sex effect for Σ PBDEs in outer blubber showed
308 that males and females had the same slope, but males had higher concentrations of Σ PBDEs in
309 outer blubber than females for similar concentrations in inner blubber.

310 Additionally, the relationships between inner and outer blubber were stronger for males
311 than females for Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs at late molting (Fig. 1) and early
312 breeding (Fig. 2). For female concentrations of Σ CHLs in inner blubber and outer blubber, the
313 relationship was stronger in late molting ($R^2=0.66$) relative to early breeding ($R^2=0.19$), whereas
314 the relationships between concentrations of Σ PCBs and Σ PBDEs in inner blubber and outer
315 blubber were similar between the two life history stages (Fig. 1; Fig. 2). For Σ DDTs, Σ PCBs, and
316 Σ CHLs from both time periods, and early breeding Σ PBDEs, the slope of the relationship for
317 males was steeper than females, indicating that males with higher inner blubber concentrations
318 had proportionately higher concentrations than females in outer blubber when compared to males
319 and females with lower inner blubber concentrations.

320 Additionally, during the late molt we detected a significant interaction between POP
321 concentrations in inner blubber \times sex on POP concentrations in outer blubber for α -HCH, β -HCH
322 and HCB ($F_{1,34} \geq 5.51$, $p \leq 0.025$), but not for 6-MeO-BDE 47 ($F_{1,34} = 0.03$, $p = 0.87$). For 6-MeO-
323 BDE 47, inner blubber POP concentrations were positively related to outer blubber

324 concentrations (Fig. 3). Inner blubber concentrations of α -HCH and β -HCH were positively
325 related to outer blubber POP concentrations at late molting for females and males, separately
326 (Fig. 3). Inner blubber concentrations of HCB were only significantly related for males and not
327 females.

328 Early in the breeding fast, we did not detect significant interactions between inner
329 blubber \times sex on outer blubber concentrations of 6-MeO-BDE 47, α -HCH, β -HCH, or HCB
330 ($F_{1,35} \leq 1.43$, $p \geq 0.24$). Inner blubber concentrations for all four compounds were significantly
331 related to outer blubber concentrations, when accounting for any potential effect of sex (Fig 4).

332

333 *Partitioning coefficients*

334 Inner blubber/serum partitioning coefficients for Σ DDTs, Σ CHLs, and Σ PBDEs were
335 significantly lower at late molt than early breeding ($F_{1,45.9} = 15.64$, $p < 0.001$), but did not differ by
336 sex ($F_{1,60.0} = 0.61$, $p = 0.44$; Table 4). For Σ PCBs, we detected a significant sex \times time period
337 interaction ($F_{1,59.6} = 10.25$, $p = 0.002$). Females and males did not differ in Σ PCB inner
338 blubber/serum partitioning coefficients at late molting ($t = 1.15$, $p = 0.99$), but females had higher
339 inner blubber/serum partitioning coefficients than males at early breeding ($t = 4.73$, $p < 0.001$),
340 indicating that females had proportionally higher outer blubber concentrations relative to serum
341 than males at early breeding. Female partitioning coefficients for Σ PCBs were lower at late molt
342 relative to early breeding ($t = 5.75$, $p < 0.001$), while male coefficients for Σ PCBs did not differ
343 ($t = 0.413$, $p = 0.98$).

344 For outer blubber/serum partitioning coefficients, we detected a significant interaction
345 between sex and time period for Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs ($F \geq 6.57$, $p \leq 0.01$; Table
346 4). Females and males did not differ at late molting for Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs

347 ($t \leq 1.85$, $p \geq 0.26$), but partitioning coefficients for females were significantly higher than males
348 for each contaminant compound at early breeding ($t \geq 3.27$, $p \leq 0.01$). For males, partitioning
349 coefficients were lower at late molting than early breeding for Σ DDTs and Σ CHLs ($t \geq 3.75$,
350 $p \leq 0.002$), but not for Σ PCBs and Σ PBDEs ($t \leq 1.94$, $p \geq 0.22$; Table 4). For females, partitioning
351 coefficients were lower during late molting than early breeding for Σ DDTs, Σ PCBs, Σ CHLs, and
352 Σ PBDEs ($t \geq 7.39$, $p \leq 0.001$; Table 4).

353 For outer blubber/inner blubber partitioning coefficients, we detected a significant sex \times
354 time period interaction for Σ PCBs ($F_{1,59,6} = 15.10$, $p = 0.003$), Σ CHLs ($F_{1,59,6} = 5.80$, $p = 0.02$), and
355 Σ PBDEs ($F_{1,61,1} = 6.63$, $p = 0.01$), but not Σ DDTs ($F_{1,59,6} = 2.45$, $p = 0.12$). At late molting, outer
356 blubber/inner blubber coefficients were lower for females relative to males for Σ PCBs ($t = 3.05$,
357 $p = 0.02$; Table 4), but Σ CHLs and Σ PBDEs did not differ between males and females at this time
358 period ($t \leq 1.13$, $p \geq 0.67$). Early in breeding, females and males did not differ for Σ PCBs, Σ CHLs,
359 or Σ PBDEs (Table 4). For males, outer blubber/inner blubber partitioning coefficients were
360 lower at late molting than early breeding for Σ CHLs ($t = 2.86$, $p = 0.028$), but the time periods did
361 not differ for Σ PCBs or Σ PBDEs ($t \leq 1.48$, $p \geq 0.45$). Female outer blubber/inner blubber
362 partitioning coefficients were lower at late molting than early breeding for Σ PCBs, Σ CHLs, and
363 Σ PBDEs ($t \geq 5.05$, $p \leq 0.0001$). For Σ DDTs, outer blubber/inner blubber partitioning coefficients
364 were significantly lower at late molt than early breeding ($F_{1,45,9} = 42.37$, $p < 0.001$), but did not
365 differ by sex ($F_{1,60,0} = 2.96$, $p < 0.09$).

366

367

DISCUSSION

368 *Body condition fluctuations*

369 Free-ranging northern elephant seals demonstrated strong, predictive relationships among
370 serum, inner blubber, and outer blubber POP concentrations, even after the influence of lengthy
371 periods of foraging at sea or weeks of fasting on land. The ability to predict POP concentrations
372 between components of blood and blubber is important because POP concentrations can
373 fluctuate asynchronously among serum, inner blubber, and outer blubber in response to recent
374 fasting or foraging activities (Debier et al. 2012, Peterson et al. 2014, Louis et al. 2016). For
375 example, when northern elephant seals were sampled late in the molting fast after fasting for
376 several weeks, POP concentrations in serum and inner blubber likely reflected recent
377 mobilization of POPs from inner blubber to serum (Louis et al. 2014) and redistribution of POPs
378 among other internal tissues. Early in the breeding fast, elephant seals had recently returned from
379 an extensive, months-long foraging trip; therefore, serum and inner blubber POP concentrations
380 likely represented a combination of POPs acquired from food, as well as ongoing redistribution
381 of POPs from other internal tissues. Outer blubber, on the other hand, is less metabolically active
382 than inner blubber (Strandberg et al. 2008, Debier et al. 2012, Ellisor et al. 2013), and may
383 reflect a different, longer-term signal of POP bioaccumulation relative to inner blubber or serum.

384 Mobilization of fatty acids and POPs from blubber is complex, and is influenced by the
385 relative lipophilicity of individual fatty acids (Hall et al. 2008, Louis et al. 2016) and POPs
386 (Louis et al. 2016), as well as the vertical composition of the blubber itself (Koopman et al.
387 1996). Therefore, the relationship between POP concentrations in different tissues could change
388 as a consequence of preferential mobilization of some POP compounds over others. Of the POPs
389 investigated in this study, HCHs and HCB have the lowest lipophilicity and therefore likely
390 redistribute differently than the other POPs, which may explain why HCHs and HCBs
391 demonstrated poorer correlations among certain tissues.

392 Partitioning coefficients from the extremes of body condition corroborate the concept that
393 POP concentrations of different tissues do not fluctuate in parallel. For female seals, lower inner
394 blubber/serum, outer blubber/serum, and outer blubber/inner blubber partitioning coefficients for
395 Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs from late molting (pre-foraging), compared with early
396 breeding (post-foraging), showed that although both serum and blubber layers decreased in POP
397 concentrations during foraging (Peterson et al. 2015), the greatest decrease in magnitude
398 occurred in serum POP concentrations, followed by inner blubber and outer blubber,
399 respectively. Differences in partitioning coefficients between time periods were less consistent
400 for males, which may be a result of the lack of paired samples for males or may signal
401 differences in POP mobilization between males and females. However, in most cases males had
402 lower partitioning coefficients at late molt relative to early breeding.

403

404 *Predictive equations*

405 Serum generally performed well as a predictor of inner blubber POP concentrations for
406 female and male elephant seals at two extremes in body condition. For example, we found
407 relatively strong predictive relationships between serum and inner blubber for Σ DDTs, Σ PCBs,
408 Σ CHLs, and Σ PBDEs at both body condition extremes. Among tissue comparisons, serum and
409 inner blubber were notable because both female and male seals could be included in the same
410 equation for nearly all POP compounds during both time periods. Serum and inner blubber have
411 higher turnover rates compared with outer blubber, which may be more important in these tissues
412 than the differences in offloading mechanisms between males and females. The lack of
413 detectable interactions between serum POP concentrations \times sex, or an effect of sex, except for
414 Σ PBDEs during early breeding, indicates that serum and inner blubber relate similarly in males

415 and females despite concurrent gestation of a pup by females and the highly probable
416 corresponding placental transfer of POPs, as well as transfer of POPs from mother to pup during
417 the first 5 days of lactation.

418 Unlike inner blubber:serum relationships, for outer blubber:serum and outer
419 blubber:inner blubber we detected a significant interaction between predictor tissue POP
420 concentration and sex for all major POP compounds, except Σ PBDEs. When there was a
421 significant interaction, and thus a difference in the slope of the relationships, male elephant seals
422 were always characterized by a steeper slope compared with females, resulting in greater
423 differences between male and female outer blubber concentrations at higher concentrations of
424 serum or inner blubber than at lower concentrations. Although this may be attributed to
425 differences in the impact of concurrent lactation on specific POP compounds during the breeding
426 fast (Debier et al. 2012, Vanden Berghe et al. 2012), the fact that significant interactions also
427 existed in late molting, when lactation and gestation should not be having immediate effects,
428 suggest that a lifetime of offloading opportunities for females may cause persistent differences in
429 bioaccumulation among tissue compartments.

430 Furthermore, while predictive equations for males performed well among all tissues and
431 nearly all POP compounds at both sampling periods, the strength of female tissue relationships
432 was inconsistent across POP compounds. Female serum and inner blubber were strong predictors
433 of outer blubber for Σ DDTs and Σ PBDEs, but weaker predictors for Σ PCBs and Σ CHLs. Female
434 lactation physiology results in disproportionate mobilization and transfer to milk of some POP
435 compounds over others (Debier et al. 2012, Vanden Berghe et al. 2013), which may explain the
436 weaker performance of predictive equations for Σ PCBs and Σ CHLs. For example, Σ PCBs
437 represents the sum of a suite of congeners, each with compound-specific log K_{ow} (octanol-water

438 partitioning coefficient; Hawker and Connell 1988). When females lactate during the breeding
439 fast, lower lipophilic congeners are more readily mobilized from inner blubber to serum and
440 from serum to milk during lactation (Debier et al. 2003, 2012a, Vanden Berghe et al. 2012),
441 which may make serum POP concentrations an unreliable predictor of inner or outer blubber
442 POP concentrations for some compounds in certain scenarios. In the current study, differences in
443 lipophilicity of specific congeners may distort the ability to predict outer blubber concentrations
444 from serum or inner blubber for Σ PCBs in both seasons ($R^2 \leq 0.44$) and for Σ CHLs in early
445 breeding ($R^2 \leq 0.25$).

446

447 *Interspecies comparisons*

448 Significant positive relationships between blood and fat compartments have been
449 observed and quantified for POPs in the tissues of polar bears (Bernhoft et al. 1997), turtles
450 (Keller et al. 2004), dolphins (Yordy et al. 2010c), and Hawaiian monk seals (Lopez et al. 2012).
451 Hawaiian monk seals and northern elephant seals are both pinnipeds, the taxonomic group that
452 includes seals, sea lions, and walrus. Similar to the elephant seal inner blubber:serum
453 relationships in the present study, Hawaiian monk seals of various ages and both sexes had
454 strong relationships between blubber and serum for Σ DDTs, Σ PCBs, and Σ CHLs (Lopez et al.
455 2012), while Σ PBDE relationships were not reported.

456 Our study of a pinniped species is similar in approach to a study on an odontocete, the
457 taxonomic group that includes toothed cetaceans, which determined predictive equations for
458 Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs in bottlenose dolphin (*Tursiops truncatus*) from Sarasota
459 Bay, Florida, USA (Yordy et al. 2010c). Bottlenose dolphin predictive equations were
460 established for male and female dolphins using plasma and full-thickness blubber; the blubber of

461 bottlenose dolphins varies with vertical profile (Ellisor et al. 2013). Partitioning coefficients
462 (blubber/plasma) were calculated for males, juveniles, and females (Yordy et al. 2010c).
463 Northern elephant seals differ from bottlenose dolphins in several aspects, namely that they come
464 ashore and undergo extreme fasting periods that mobilize POPs from blubber to serum (Debiec et
465 al. 2006). Additionally, the inter-offspring interval for dolphins is longer than for elephant seals,
466 which provides less frequent opportunities for contaminant elimination. Finally, dolphins have a
467 lipid-rich internal melon tissue that can accumulate POPs (Yordy et al. 2010a), while elephant
468 seals do not. Although different in certain life history aspects, both northern elephant seals and
469 bottlenose dolphins are relatively long-lived, top marine predators with relatively large fat
470 compartments (blubber) that can bioaccumulate POPs from marine prey.

471 Both studies found positive relationships among POP concentrations in male blood and
472 blubber. Predictive equations for bottlenose dolphins showed strong relationships for all POP
473 classes ($R^2 \geq 0.91$), but Yordy et al. (2010) did not quantify predictive relationships for females.
474 Bottlenose dolphin predictive equations for males and juvenile had slopes from 0.91-1.08,
475 similar to male elephant seals at early breeding (0.78-0.99), when they had recently completed
476 their foraging trip and thus were most directly comparable to continuously foraging bottlenose
477 dolphins. Male elephant seal equations showed a greater range of slopes (0.91-1.42) for POP
478 compounds late in the molting fast.

479 Additionally, blubber/plasma partitioning coefficients reported in Yordy et al. (2010) for
480 male bottlenose dolphins were higher for Σ DDTs (mean=1.95, 95% CI=1.73-2.17) and Σ CHLs
481 (2.52, 2.23-2.81), but lower for Σ PCBs (1.42, 1.24-1.60) and Σ PBDEs (1.24, 1.04-1.44), than
482 either inner blubber/serum or outer blubber/serum partitioning coefficients for male elephant
483 seals (Table 4). Further, female bottlenose dolphins (Yordy et al. 2010c) had lower partitioning

484 coefficients for Σ DDTs (1.93, 1.66-2.20), Σ PCBs (1.77, 1.42-2.10), and Σ CHLs (2.35, 1.14-3.55)
485 than female elephant seals early in the breeding fast, but higher partitioning coefficients than
486 female elephant seals late in the molting fast. Early in breeding, female elephant seals have just
487 returned from a long foraging trip, and have very recently given birth to a pup and begun
488 lactation. Although recent lactation could eliminate some POPs from serum, higher partitioning
489 coefficients in elephant seals early in the breeding fast are likely representative of greater
490 dilution of POPs in serum compared with blubber, in response to recent foraging.

491 The differences in partitioning coefficients between elephant seals and bottlenose
492 dolphins in this case may also be attributed to different compound-specific exposures related to
493 foraging location and prey. Elephant seal females during both time periods had higher
494 partitioning coefficients for Σ PBDEs than bottlenose dolphins, which may reflect lower exposure
495 during foraging, and consequently lower serum Σ PBDEs concentrations, than Sarasota Bay
496 bottlenose dolphins, which have higher concentrations for all POP compounds than northern
497 elephant seals (Yordy et al. 2010c).

498

499 *Implications for biomonitoring*

500 Measurements of POP concentrations in marine mammals are common in oceans around
501 the globe, but selection of tissues for sampling varies among studies. Therefore, the ability to
502 estimate POP blubber concentrations from serum, or vice versa, has the potential to enhance
503 toxicological assessment in marine mammals by providing a tool for expanding studies where
504 only one tissue is collected. In cases where blubber samples are more attainable than blood (Elfes
505 et al. 2010), the ability to estimate blood concentrations increases the capacity for toxicological
506 risk assessment, because blood interacts with vital organs. In other cases, blood samples are

507 more attainable or are banked from past collections and could be used to estimate blubber
508 concentrations, which may enable comparisons across space or time.

509 Further, the distinction between inner and outer blubber is important. The use of biopsy
510 darting to sample marine mammal blubber is increasingly used in the study of wild, free-ranging
511 marine mammals, yet the relationship of that sample to inner blubber or serum is unclear. For
512 example, outer blubber samples are commonly collected from large free-ranging mysticetes for
513 studies of hormones and contaminants because outer blubber is often the only sample that can be
514 obtained (*e.g.*, Hunt et al. 2015); therefore, the potential to link outer blubber to other tissues is
515 particularly important. Because blubber is the largest reservoir for POPs, the relationship
516 between blubber reserves and serum may enable estimations of peak serum concentrations
517 relative to life history phases and variability in body condition. The seals in the current study
518 were seemingly healthy animals that naturally fluctuate in body condition, but may represent the
519 processes that may occur in other mammals undergoing rapid changes in body condition.
520 Therefore, the relationship between contaminant concentrations in blood and blubber at such
521 extremes may be an important factor to assessments of animal health and relative vulnerability to
522 contaminant accumulation.

523 The detection of strong, positive relationships between concentrations of POP
524 compounds in blood and blubber of two marine mammals, in the current study and in Yordy et
525 al. (2010c), with clear behavior and life history differences raises the question: are predictive
526 equations species-specific or can we determine generalities among species? If future research
527 that involves direct handling of animals can produce additional blood-blubber relationships, then
528 it may be possible to construct more generalized equations for groups of similar mammals.
529 Generalized relationships, even for a few POPs, would enhance monitoring of contaminants in

530 wildlife, particularly in situations where free-ranging animals are difficult to sample.
531 Furthermore, comparisons among many species, including pinnipeds and odontocetes, could
532 identify differences in contaminant metabolism among species, and thereby further inform
533 models of potential health effects or risk assessment.

534

535

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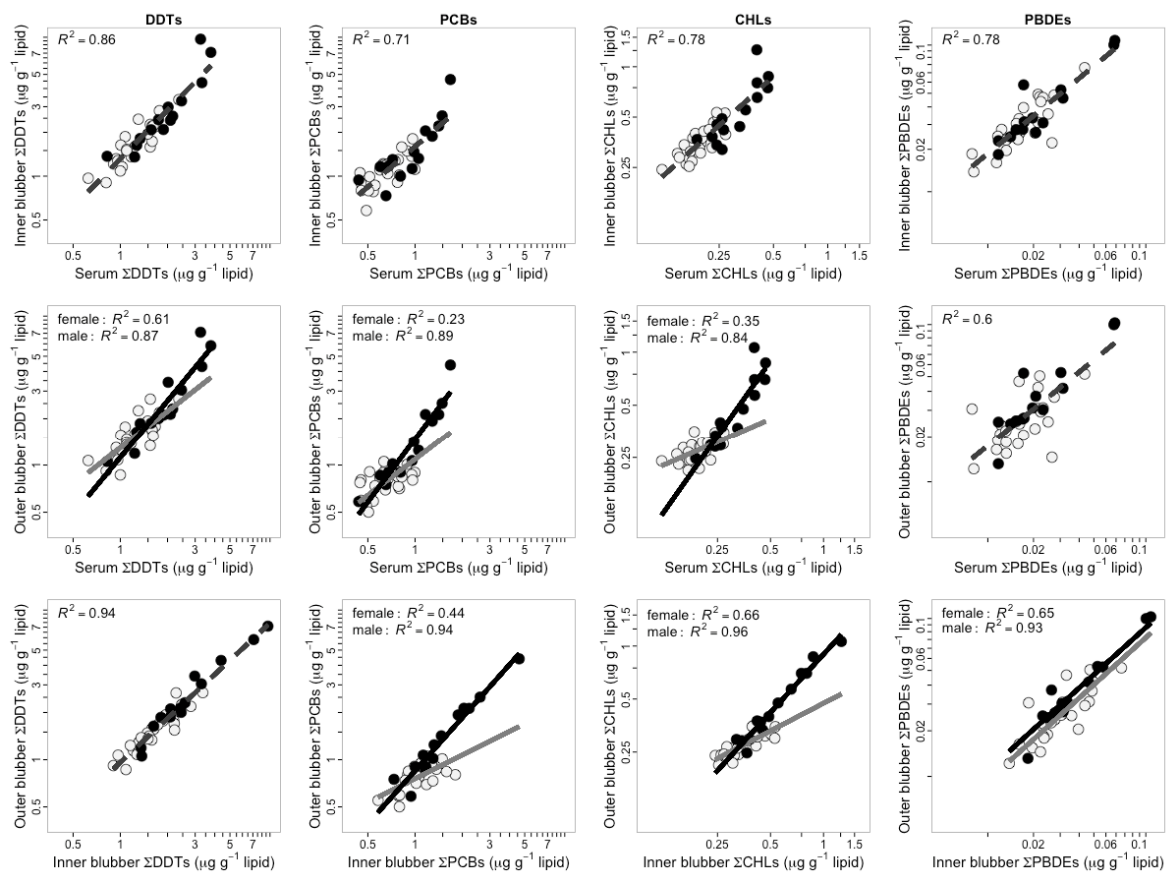
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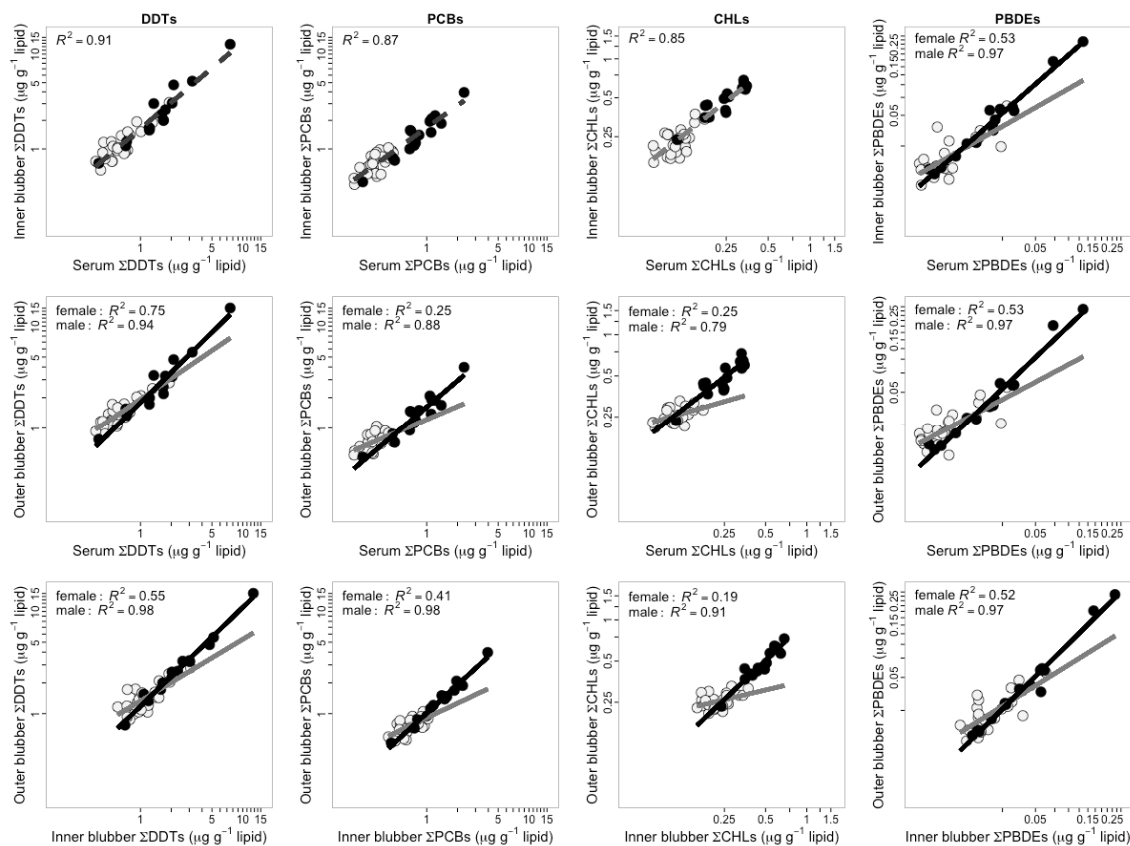
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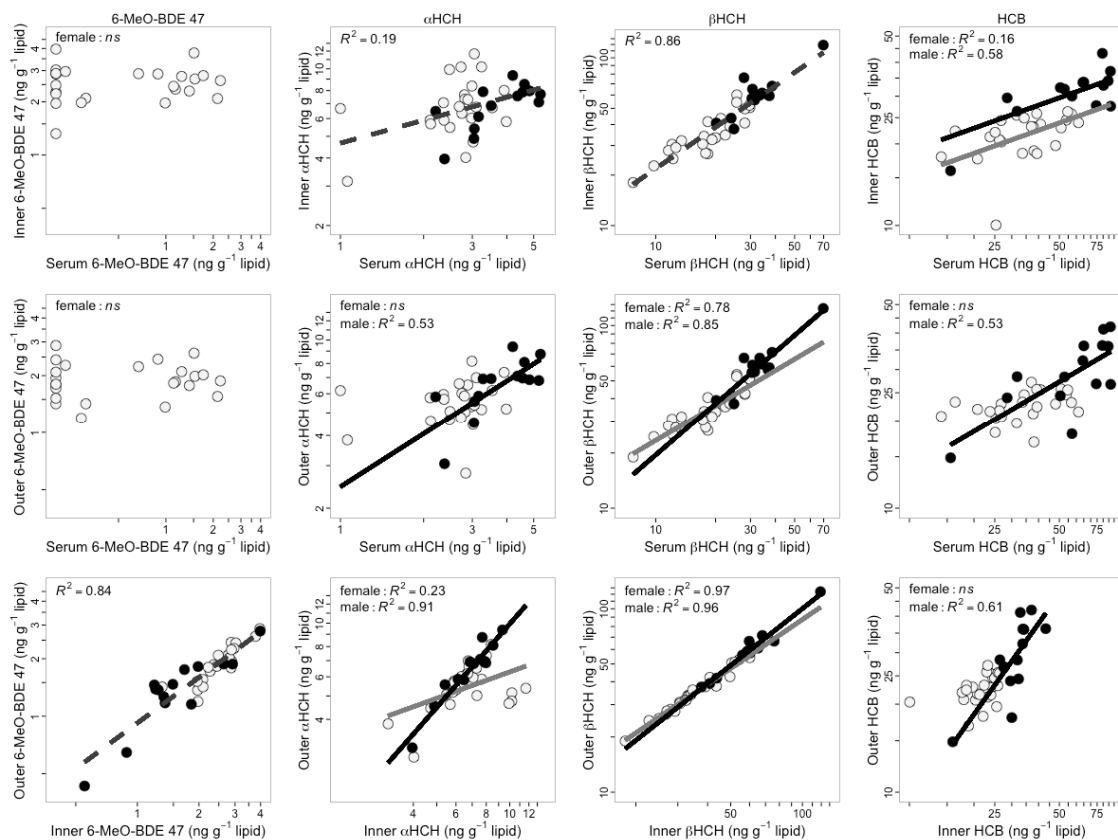
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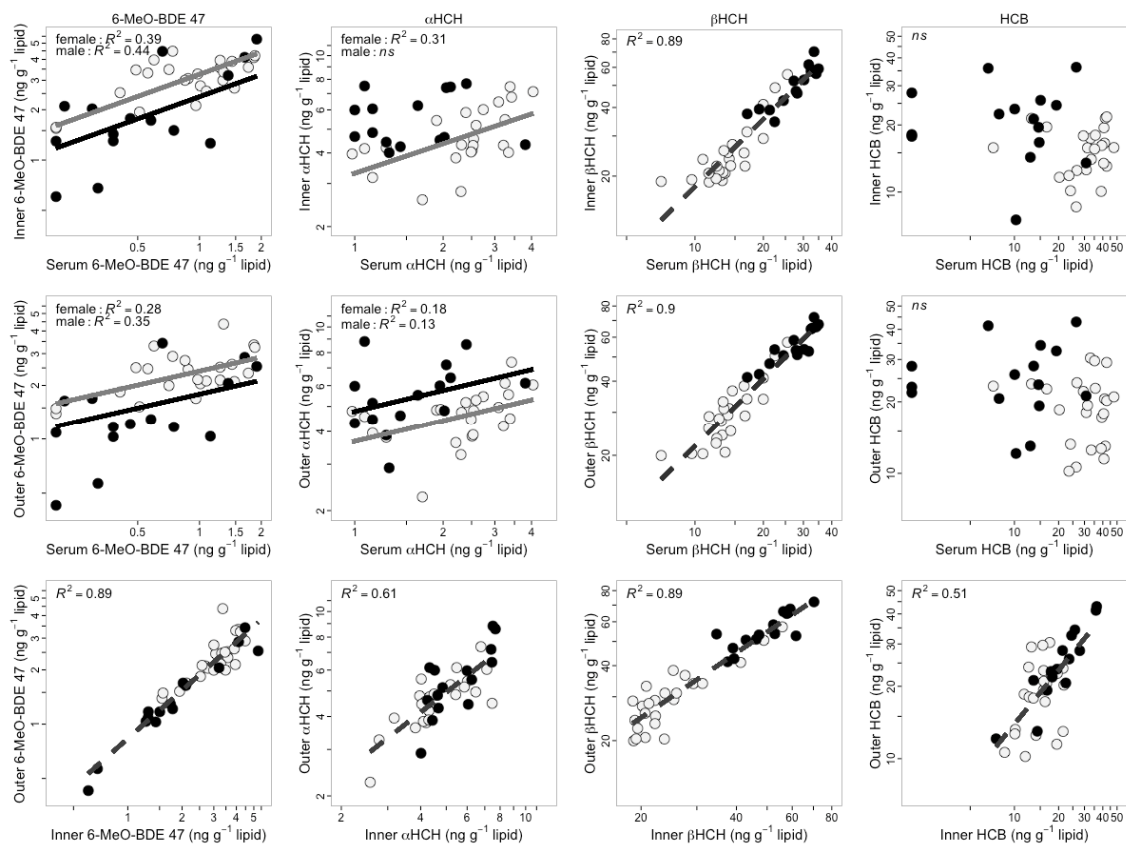
684
 685 Figure 1. Relationships between concentrations of Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs in inner blubber and
 686 serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels)
 687 of adult female and male northern elephant seals late in the molting fast (pre-foraging). Regression lines indicate
 688 significant relationships ($p < 0.05$) between the two tissues; if there was a significant difference in the slope or y-
 689 intercept between females and males, the relationship and R^2 value for both sexes are shown. Males are shown using
 690 solid circles and black lines, whereas females are open symbols and solid gray lines. Dashed lines represent the
 691 relationship for both males and females when they were not significantly different.



692
 693 Figure 2. Relationships between concentrations of Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs in inner blubber and
 694 serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber
 695 of adult female and male northern elephant seals early in the breeding fast (post-foraging). Regression lines indicate
 696 significant relationships ($p < 0.05$) between the two tissues; if there was a significant difference in the slope or y-
 697 intercept between females and males, the relationship and R^2 value for both sexes are shown. Males are shown using
 698 solid circles and black lines, whereas females are open symbols and solid gray lines. Dashed lines represent the
 699 relationship for both males and females when they were not significantly different.



700
701 Figure 3. Relationships between concentrations of 6-MeO-BDE 47, α -HCH, β -HCH, and HCB in inner blubber and
702 serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber
703 of adult female and male northern elephant seals late in the molting fast (pre-foraging). Regression lines indicate
704 significant relationships ($p < 0.05$) between the two tissues; if there was a significant difference in the slope or y-
705 intercept between females and males, the relationship and R^2 value for both sexes are shown. Males are shown using
706 solid circles and black lines, whereas females are open symbols and solid gray lines. Dashed lines represent the
707 relationship for both males and females when they were not significantly different.



708
709 Figure 4. Relationships between concentrations of 6-MeO-BDE 47, α-HCH, β-HCH, and HCB in inner blubber and
710 serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels)
711 of adult female and male northern elephant seals early in the breeding fast (post-foraging). Regression lines indicate
712 significant relationships ($p < 0.05$) between the two tissues; if there was a significant difference in the slope or y-
713 intercept between females and males, the relationship and R^2 value for both sexes are shown. Males are shown using
714 solid circles and black lines, whereas females are open symbols and solid gray lines. Dashed lines represent the
715 relationship for both males and females when they were not significantly different.

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Table 1. Lipid-normalized POP concentrations (ng g^{-1}) are reported as median (min-max) POP concentration for each compound in serum, inner blubber, and outer blubber of male and female northern elephant seals. The same females ($N = 24$) were sampled late in the molting fast and early in the breeding fast, before and after an extensive foraging migration. Unpaired males were sampled late in the molting fast ($N=14$) and early in the breeding fast ($N=15$). The full summary of these data are presented in Peterson et al. (2015).

Compound	Sex	Late molting fast			Early breeding fast		
		Serum	Inner	Outer	Serum	Inner	Outer
ΣDDTs	Female	1136 (623-2360)	1589 (902-3354)	1513 (865-2677)	593 (365-1912)	994 (598-3086)	1408 (875-2803)
	Male	1950 (823-3778)	2512 (1054-7107)	2222 (1355-8730)	1604 (390-7532)	2030 (705-12689)	2556 (770-15061)
ΣPCBs	Female	651 (445-997)	1097 (580-1806)	821 (501-1053)	311 (194-470)	713 (422-1029)	787 (539-1046)
	Male	971 (437-1679)	1346 (584-4396)	1333 (733-4610)	778 (236-2317)	1495 (448-3942)	1456 (517-3975)
ΣCHLs	Female	196 (120-271)	345 (242-532)	278 (211-352)	109 (74-175)	229 (158-373)	507 (207-327)
	Male	335 (187-468)	486 (247-1061)	436 (320-1262)	249 (111-350)	492 (237-684)	445 (233-729)
ΣPBDEs	Female	16 (7-43)	27 (14-76)	24 (12-52)	7 (4-27)	16 (10-61)	21 (13-55)
	Male	19 (11-68)	32 (13-112)	31 (18-118)	21 (5-135)	38 (13-264)	38 (14-284)

721 Table 2. Predictive equations, including only significant terms, for concentrations of Σ DDTs, Σ PCBs, Σ CHLs,
 722 Σ PBDEs, 6-MeO-BDE 47 (MeOBDE), α -HCH, β -HCH, and HCB in inner blubber:serum, outer blubber:serum, and
 723 outer:inner blubber for two times of year (late molting and early breeding). The R^2 refers to goodness of fit of the
 724 data to the predictive equation. Subscripts indicate the tissue type: In (inner), Out (outer), and Ser (serum). The 95%
 725 confidence interval (CI) is provided for the slope and intercept of each equation. Equations are developed using the
 726 natural log of tissue concentrations.

Sex	Equation: $\ln(\text{tissue Y}) = \text{slope} \times \ln(\text{tissue X}) + \text{intercept}$	R^2	95% CI for slope	95% CI for intercept
Late molting (N=14 males, 24 females)				
<i>Inner blubber:serum</i>				
F+M	$\Sigma\text{DDT}_{\text{In}} = 1.107 \times \Sigma\text{DDT}_{\text{Ser}} - 0.467$	0.86	(0.957, 1.258)	(-1.561, 0.627)
F+M	$\Sigma\text{PCB}_{\text{In}} = 0.921 \times \Sigma\text{PCB}_{\text{Ser}} + 1.014$	0.71	(0.721, 1.121)	(-0.312, 2.341)
F+M	$\Sigma\text{CHL}_{\text{In}} = 0.971 \times \Sigma\text{CHL}_{\text{Ser}} + 0.739$	0.78	(0.795, 1.147)	(-0.221, 1.699)
F+M	$\Sigma\text{PBDE}_{\text{In}} = 0.888 \times \Sigma\text{PBDE}_{\text{Ser}} + 0.883$	0.78	(0.730, 1.045)	(0.416, 1.349)
F	MeOBDE: <i>Not significant</i>	--	--	--
M	MeOBDE: <i>Low detectability</i>	--	--	--
F+M	$\alpha\text{-HCH}_{\text{In}} = 0.340 \times \alpha\text{-HCH}_{\text{Ser}} + 1.539$	0.19	(0.105, 0.574)	(1.271, 1.807)
F+M	$\beta\text{-HCH}_{\text{In}} = 0.820 \times \beta\text{-HCH}_{\text{Ser}} + 1.197$	0.86	(0.707, 0.932)	(0.845, 1.549)
F	$\text{HCB}_{\text{In}} = 0.282 \times \text{HCB}_{\text{Ser}} + 2.071$	0.16	(0.145, 0.419)	(1.586, 2.557)
M	$\text{HCB}_{\text{In}} = 0.282 \times \text{HCB}_{\text{Ser}} + 2.28$	0.58	(0.145, 0.419)	(1.727, 2.841)
<i>Outer blubber:serum</i>				
F	$\Sigma\text{DDT}_{\text{Out}} = 0.782 \times \Sigma\text{DDT}_{\text{Ser}} + 1.761$	0.61	(0.504, 1.061)	(-0.219, 3.742)
M	$\Sigma\text{DDT}_{\text{Out}} = 1.212 \times \Sigma\text{DDT}_{\text{Ser}} - 1.349$	0.87	(0.924, 1.501)	(-3.531, 0.833)
F	$\Sigma\text{PCB}_{\text{Out}} = 0.370 \times \Sigma\text{PCB}_{\text{Ser}} + 4.269$	0.23	(0.075, 0.665)	(2.353, 6.184)
M	$\Sigma\text{PCB}_{\text{Out}} = 1.342 \times \Sigma\text{PCB}_{\text{Ser}} - 1.973$	0.89	(1.046, 1.637)	(-3.995, 0.050)
F	$\Sigma\text{CHL}_{\text{Out}} = 0.436 \times \Sigma\text{CHL}_{\text{Ser}} + 3.321$	0.35	(0.172, 0.701)	(1.929, 4.713)
M	$\Sigma\text{CHL}_{\text{Out}} = 1.423 \times \Sigma\text{CHL}_{\text{Ser}} - 2.055$	0.84	(1.031, 1.816)	(-4.308, 0.197)
F+M	$\Sigma\text{PBDE}_{\text{Out}} = 0.814 \times \Sigma\text{PBDE}_{\text{Ser}} + 0.980$	0.60	(0.587, 1.040)	(0.310, 1.650)
F	MeOBDE: <i>Not significant</i>	--	--	--
M	MeOBDE: <i>Low detectability</i>	--	--	--
F	$\alpha\text{-HCH}$: <i>Not significant</i>	--	--	--
M	$\alpha\text{-HCH}_{\text{Out}} = 0.732 \times \alpha\text{-HCH}_{\text{Ser}} + 0.897$	0.53	(0.302, 1.163)	(0.321, 1.473)
F	$\beta\text{-HCH}_{\text{Out}} = 0.640 \times \beta\text{-HCH}_{\text{Ser}} + 1.682$	0.78	(0.490, 0.789)	(1.245, 2.119)
M	$\beta\text{-HCH}_{\text{Out}} = 0.939 \times \beta\text{-HCH}_{\text{Ser}} + 0.810$	0.85	(0.687, 1.190)	(-0.067, 1.687)
F	HCB: <i>Not significant</i>	--	--	--
M	$\text{HCB}_{\text{Out}} = 0.431 \times \text{HCB}_{\text{Ser}} + 1.618$	0.53	(0.178, 0.684)	(0.594, 2.642)
<i>Outer blubber:inner blubber</i>				
F+M	$\Sigma\text{DDT}_{\text{Out}} = 0.932 \times \Sigma\text{DDT}_{\text{In}} + 0.430$	0.94	(0.853, 1.011)	(-0.174, 1.034)
F	$\Sigma\text{PCB}_{\text{Out}} = 0.499 \times \Sigma\text{PCB}_{\text{In}} + 3.184$	0.44	(0.251, 0.747)	(1.450, 4.918)
M	$\Sigma\text{PCB}_{\text{Out}} = 1.124 \times \Sigma\text{PCB}_{\text{In}} - 1.019$	0.94	(0.942, 1.306)	(-2.351, 0.312)
F	$\Sigma\text{CHL}_{\text{Out}} = 0.521 \times \Sigma\text{CHL}_{\text{In}} + 2.563$	0.66	(0.355, 0.687)	(1.589, 3.537)
M	$\Sigma\text{CHL}_{\text{Out}} = 1.098 \times \Sigma\text{CHL}_{\text{In}} - 0.782$	0.96	(0.957, 1.238)	(-1.665, 0.101)
F	$\Sigma\text{PBDE}_{\text{Out}} = 0.906 \times \Sigma\text{PBDE}_{\text{In}} + 0.150$	0.65	(0.753, 1.060)	(-0.377, 0.677)

M	$\Sigma\text{PBDE}_{\text{Out}} = 0.906 \times \Sigma\text{PBDE}_{\text{In}} + 0.303$	0.93	(0.753, 1.060)	(-0.266, 0.872)
F+M	$\text{MeOBDE}_{\text{Out}} = 0.793 \times \text{MeOBDE}_{\text{In}} - 0.083$	0.84	(0.675, 0.912)	(-0.183, 0.018)
F	$\alpha\text{-HCH}_{\text{Out}} = 0.362 \times \alpha\text{-HCH}_{\text{In}} + 1.005$	0.23	(0.070, 0.653)	(0.444, 1.566)
M	$\alpha\text{-HCH}_{\text{Out}} = 1.144 \times \alpha\text{-HCH}_{\text{In}} - 0.340$	0.91	(0.912, 1.376)	(-0.788, 0.108)
F	$\beta\text{-HCH}_{\text{Out}} = 0.885 \times \beta\text{-HCH}_{\text{In}} + 0.393$	0.97	(0.821, 0.950)	(0.163, 0.624)
M	$\beta\text{-HCH}_{\text{Out}} = 1.028 \times \beta\text{-HCH}_{\text{In}} - 0.127$	0.96	(0.900, 1.156)	(-0.651, 0.396)
F	HCB: <i>Not significant</i>	--	--	--
M	$\text{HCB}_{\text{Out}} = 1.034 \times \Sigma\text{HCB}_{\text{In}} - 0.181$	0.61	(0.516, 1.552)	(-1.955, 1.593)

Early breeding (N=15 males, 24 females)

Inner blubber:serum

F+M	$\Sigma\text{DDT}_{\text{In}} = 0.911 \times \Sigma\text{DDT}_{\text{Ser}} + 1.110$	0.91	(0.814, 1.008)	(0.448, 1.772)
F+M	$\Sigma\text{PCB}_{\text{In}} = 0.774 \times \Sigma\text{PCB}_{\text{Ser}} + 2.082$	0.87	(0.674, 0.873)	(1.471, 2.692)
F+M	$\Sigma\text{CHL}_{\text{In}} = 0.851 \times \Sigma\text{CHL}_{\text{Ser}} + 1.451$	0.85	(0.732, 0.970)	(0.854, 2.048)
F	$\Sigma\text{PBDE}_{\text{In}} = 0.615 \times \Sigma\text{PBDE}_{\text{Ser}} + 1.684$	0.53	(0.357, 0.872)	(1.138, 2.229)
M	$\Sigma\text{PBDE}_{\text{In}} = 0.947 \times \Sigma\text{PBDE}_{\text{Ser}} + 0.913$	0.97	(0.848, 1.047)	(0.608, 1.218)
F	$\text{MeOBDE}_{\text{In}} = 0.440 \times \text{MeOBDE}_{\text{Ser}} + 1.181$	0.39	(0.269, 0.611)	(1.035, 1.327)
M	$\text{MeOBDE}_{\text{In}} = 0.440 \times \text{MeOBDE}_{\text{Ser}} + 0.873$	0.44	(0.269, 0.611)	(0.661, 1.084)
F	$\alpha\text{-HCH}_{\text{In}} = 0.403 \times \alpha\text{-HCH}_{\text{Ser}} + 1.195$	0.32	(0.141, 0.664)	(0.958, 1.432)
M	$\alpha\text{-HCH}$: <i>Not significant</i>	--	--	--
F+M	$\beta\text{-HCH}_{\text{In}} = 0.984 \times \beta\text{-HCH}_{\text{Ser}} + 0.623$	0.89	(0.868, 1.100)	(0.285, 0.962)
F+M	HCB: <i>Not significant</i>	--	--	--

Outer blubber:serum

F	$\Sigma\text{DDT}_{\text{Out}} = 0.683 \times \Sigma\text{DDT}_{\text{Ser}} + 2.847$	0.75	(0.509, 0.857)	(1.725, 3.969)
M	$\Sigma\text{DDT}_{\text{Out}} = 0.986 \times \Sigma\text{DDT}_{\text{Ser}} + 0.677$	0.94	(0.831, 1.141)	(-0.460, 1.814)
F	$\Sigma\text{PCB}_{\text{Out}} = 0.389 \times \Sigma\text{PCB}_{\text{Ser}} + 4.410$	0.25	(0.093, 0.685)	(2.709, 6.111)
M	$\Sigma\text{PCB}_{\text{Out}} = 0.857 \times \Sigma\text{PCB}_{\text{Ser}} + 1.467$	0.88	(0.669, 1.045)	(0.205, 2.730)
F	$\Sigma\text{CHL}_{\text{Out}} = 0.287 \times \Sigma\text{CHL}_{\text{Ser}} + 4.194$	0.25	(0.066, 0.509)	(3.151, 5.237)
M	$\Sigma\text{CHL}_{\text{Out}} = 0.777 \times \Sigma\text{CHL}_{\text{Ser}} + 1.904$	0.79	(0.540, 1.014)	(0.600, 3.207)
F	$\Sigma\text{PBDE}_{\text{Out}} = 0.532 \times \Sigma\text{PBDE}_{\text{Ser}} + 2.044$	0.53	(0.310, 0.754)	(1.574, 2.513)
M	$\Sigma\text{PBDE}_{\text{Out}} = 0.946 \times \Sigma\text{PBDE}_{\text{Ser}} + 0.941$	0.97	(0.840, 1.053)	(0.615, 1.268)
F	$\text{MeOBDE}_{\text{Out}} = 0.350 \times \text{MeOBDE}_{\text{Ser}} + 0.891$	0.28	(0.183, 0.518)	(0.748, 1.034)
M	$\text{MeOBDE}_{\text{Out}} = 0.350 \times \text{MeOBDE}_{\text{Ser}} + 0.518$	0.35	(0.183, 0.518)	(0.311, 0.724)
F	$\alpha\text{-HCH}_{\text{Out}} = 0.270 \times \alpha\text{-HCH}_{\text{Ser}} + 1.295$	0.18	(0.059, 0.480)	(1.094, 1.496)
M	$\alpha\text{-HCH}_{\text{Out}} = 0.270 \times \alpha\text{-HCH}_{\text{Ser}} + 1.569$	0.13	(0.059, 0.480)	(1.410, 1.728)
F+M	$\beta\text{-HCH}_{\text{Out}} = 0.905 \times \beta\text{-HCH}_{\text{Ser}} + 0.998$	0.90	(0.803, 1.008)	(0.699, 1.298)
F+M	HCB: <i>Not significant</i>	--	--	--

Outer blubber:inner blubber

F	$\Sigma\text{DDT}_{\text{Out}} = 0.603 \times \Sigma\text{DDT}_{\text{In}} + 3.027$	0.55	(0.360, 0.847)	(1.320, 4.735)
M	$\Sigma\text{DDT}_{\text{Out}} = 0.967 \times \Sigma\text{DDT}_{\text{In}} + 0.393$	0.98	(0.893, 1.042)	(-0.184, 0.970)
F	$\Sigma\text{PCB}_{\text{Out}} = 0.475 \times \Sigma\text{PCB}_{\text{In}} + 3.535$	0.41	(0.222, 0.727)	(1.877, 5.193)

M	$\Sigma\text{PCB}_{\text{Out}} = 0.920 \times \Sigma\text{PCB}_{\text{In}} + 0.565$	0.98	(0.835, 1.004)	(-0.047, 1.178)
F	$\Sigma\text{CHL}_{\text{Out}} = 0.235 \times \Sigma\text{CHL}_{\text{In}} + 4.265$	0.19	(0.017, 0.453)	(3.081, 5.450)
M	$\Sigma\text{CHL}_{\text{Out}} = 0.951 \times \Sigma\text{CHL}_{\text{In}} + 0.317$	0.91	(0.767, 1.135)	(-0.815, 1.449)
F	$\Sigma\text{PBDE}_{\text{Out}} = 0.624 \times \Sigma\text{PBDE}_{\text{In}} + 1.299$	0.52	(0.360, 0.888)	(0.512, 2.085)
M	$\Sigma\text{PBDE}_{\text{Out}} = 0.988 \times \Sigma\text{PBDE}_{\text{In}} + 0.070$	0.97	(0.892, 1.085)	(-0.296, 0.435)
F+M	$\text{MeOBDE}_{\text{Out}} = 0.889 \times \text{MeOBDE}_{\text{In}} - 0.183$	0.89	(0.787, 0.991)	(-0.289, -0.077)
F+M	$\alpha\text{-HCH}_{\text{Out}} = 0.782 \times \alpha\text{-HCH}_{\text{In}} + 0.340$	0.61	(0.575, 0.988)	(0.006, 0.673)
F+M	$\beta\text{-HCH}_{\text{Out}} = 0.865 \times \beta\text{-HCH}_{\text{In}} + 0.618$	0.89	(0.264, 0.972)	(0.264, 0.972)
F+M	$\text{HCB}_{\text{Out}} = 0.746 \times \text{HCB}_{\text{In}} + 0.916$	0.51	(0.500, 0.992)	(0.217, 1.615)

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729 Table 3. Results from linear models examining the relationships between POP concentrations in pairs of tissues (inner blubber:serum, outer blubber:serum, and
 730 outer blubber:inner blubber) and the influence of sex for northern elephant seals late in the molting fast (pre-foraging trip) and early in the breeding fast (post-
 731 foraging trip).

Predicted tissue contaminant	Late molting model (predictor tissue and sex ^a)	Early breeding model (predictor tissue and sex ^a)
Inner blubber	Serum	Serum
ΣDDTs	ΣDDTs: $F_{1,35}=156.92, p<0.001$ Sex: $F_{1,35}<0.01, p=0.95$	ΣDDTs: $F_{1,36}=227.55, p<0.001$ Sex: $F_{1,36}=0.75, p=0.39$
ΣPCBs	ΣPCBs: $F_{1,35}=63.83, p<0.001$ Sex: $F_{1,35}<0.01, p=0.96$	ΣPCBs: $F_{1,36}=133.32$ Sex: $F_{1,36}=3.35, p=0.08$
ΣCHLs	ΣCHLs: $F_{1,35}=76.84, p<0.001$ Sex: $F_{1,35}=1.27, p=0.27$	ΣCHLs: $F_{1,36}=49.38, p<0.001$ Sex: $F_{1,36}=2.13, p=0.15$
ΣPBDEs	ΣPBDEs: $F_{1,35}=116.72, p<0.001$ Sex: $F_{1,35}=0.01, p=0.91$	ΣPBDEs (female): $F_{1,22}=24.55, p<0.001$ ΣPBDEs (male): $F_{1,13}=422.54, p<0.001$
αHCH	αHCH: $F_{1,35}=10.96, p=0.002$ Sex: $F_{1,35}=2.09, p=0.16$	αHCH (female): $F_{1,22}=10.2, p=0.004$ αHCH (male): $F_{1,13}=0.04, p=0.84$
βHCH	βHCH: $F_{1,35}=111.79, p<0.001$ Sex: $F_{1,35}=2.81, p=0.10$	βHCH: $F_{2,36}=153.3, p<0.001$ Sex: $F_{1,36}=6.54, p=0.015$
HCB	HCB: $F_{1,35}=17.57, p<0.001$ Sex: $F_{1,35}=9.76, p=0.003$	HCB: $F_{1,36}=0.02, p=0.89$ Sex: $F_{1,36}=4.2, p=0.048$
6-MeO-BDE 47	6-MeO-BDE 47: $F_{1,22}=0.347, p=0.56$ (females only)	6-MeO-BDE 47: $F_{2,36}=23.65, p<0.001$ Sex: $F_{1,35}=1.95, p=0.17$
Outer blubber	Serum	Serum
ΣDDTs	ΣDDTs (female): $F_{1,22}=33.95, p<0.001$ ΣDDTs (male): $F_{1,12}=83.77, p<0.001$	ΣDDTs (female): $F_{1,22}=66.43, p<0.001$ ΣDDTs (male): $F_{1,13}=188.90, p<0.001$
ΣPCBs	ΣPCBs (Female): $F_{1,22}=6.77, p=0.016$ ΣPCBs (male): $F_{1,12}=97.86, p<0.001$	ΣPCBs (female): $F_{1,22}=7.44, p=0.012$ ΣPCBs (male): $F_{1,13}=97.18, p<0.001$
ΣCHLs	ΣCHLs (female): $F_{1,22}=11.71, p=0.002$ ΣCHLs (male): $F_{1,12}=62.53, p<0.001$	ΣCHLs (female): $F_{1,22}=7.22, p=0.013$ ΣCHLs (male): $F_{1,13}=50.00, p<0.001$
ΣPBDEs	ΣPBDEs: $F_{1,35}=45.13, p<0.001$ Sex: $F_{1,35}=2.37, p=0.13$	ΣPBDEs (female): $F_{1,22}=24.80, p<0.001$ ΣPBDEs (male): $F_{1,13}=368.10, p<0.001$
αHCH	αHCH (female): $F_{1,22}=1.13, p=0.30$ αHCH (male): $F_{1,12}=13.74, p=0.003$	αHCH: $F_{1,36}=5.78, p=0.007$ Sex: $F_{1,36}=9.49, p=0.004$
βHCH	βHCH (female): $F_{1,22}=78.55, p<0.001$ βHCH (male): $F_{1,12}=65.98, p<0.001$	βHCH: $F_{1,36}=166.13, p<0.001$ Sex: $F_{1,36}=2.09, p=0.16$
HCB	HCB (female): $F_{1,22}=3.34, p=0.08$ HCB (male): $F_{1,12}=13.79, p=0.003$	HCB: $F_{1,34}=3.05, p=0.06$ Sex: $F_{1,36}=3.34, p=0.08$
6-MeO-BDE 47	6-MeO-BDE 47: $F_{1,22}=0.11, p=0.74$ (females only)	6-MeO-BDE 47: $F_{1,36}=20.61, p<0.001$ Sex: $F_{1,36}=10.04, p=0.003$

Outer blubber	Inner blubber	Inner blubber
ΣDDTs	ΣDDTs: $F_{1,35}=409.70, p<0.001$ Sex: $F_{1,35}=0.64, p=0.43$	ΣDDTs (female): $F_{1,22}=26.36, p<0.001$ ΣDDTs (male): $F_{1,13}=791.90, p<0.001$
ΣPCBs	ΣPCBs (female): $F_{1,22}=17.40, p<0.001$ ΣPCBs (male): $F_{1,12}=181.40, p<0.001$	ΣPCBs (female): $F_{1,22}=15.15, p<0.001$ ΣPCBs (male): $F_{1,13}=552.00, p<0.001$
ΣCHLs	ΣCHLs (female): $F_{1,22}=42.37, p<0.001$ ΣCHLs (male): $F_{1,12}=290.40, p<0.001$	ΣCHLs (female): $F_{1,22}=5.02, p=0.036$ ΣCHLs (male): $F_{1,13}=124.82, p<0.001$
ΣPBDEs	ΣPBDEs: $F_{1,35}=143.80, p<0.001$ Sex: $F_{1,35}=4.26, p<0.047$	ΣPBDEs (female): $F_{1,22}=24.05, p<0.001$ ΣPBDEs (male): $F_{1,13}=489.40, p<0.001$
αHCH	αHCH (female): $F_{1,22}=6.63, p=0.017$ αHCH (male): $F_{1,12}=115.6, p<0.001$	αHCH: $F_{1,36}=49.34, p<0.001$ Sex: $F_{1,36}=0.60, p=0.45$
βHCH	βHCH (female): $F_{1,22}=807.2, p<0.001$ βHCH (male): $F_{1,12}=307.0, p<0.001$	βHCH: $F_{1,36}=92.61, p<0.001$ Sex: $F_{1,36}=2.10, p=0.16$
HCB	HCB (female): $F_{1,22}=4.16, p=0.054$ HCB (male): $F_{1,12}=18.90, p<0.001$	HCB: $F_{1,36}=27.18, p<0.001$ Sex: $F_{1,36}=0.52, p=0.47$
6-MeO-BDE 47	6-MeO-BDE 47: $F_{1,34}=140.92, p<0.001$ Sex: $F_{1,34}=1.04, p=0.31$	6-MeO-BDE 47: $F_{1,36}=224.36, p<0.001$ Sex: $F_{1,36}=3.30, p=0.08$

732 ^aModel results are shown for sex unless males and females were run separately because of a significant predictor tissue × sex interaction. Results are shown by sex
733 if males and females were analyzed separately.

734 Table 4. Partitioning coefficient least-squares means (95% confidence interval)
 735 for Σ DDTs, Σ PCBs, Σ CHLs, and Σ PBDEs in female and male elephant seals
 736 late in the molting fast and early in the breeding fast.

Type	Sex	Late molting	Early breeding
<i>Inner blubber/serum</i>			
Σ DDTs	Female	1.36 (1.23-1.49)	1.77 (1.64-1.90)
	Male	1.45 (1.28-1.62)	1.57 (1.40-1.74)
Σ PCBs	Female	1.68 (1.53-1.82)	2.28 (2.13-2.42)
	Male	1.66 (1.46-1.85)	1.71 (1.53-1.90)
Σ CHLs	Female	1.84 (1.70-1.99)	2.13 (1.98-2.27)
	Male	1.76 (1.57-1.95)	1.96 (1.77-2.14)
Σ PBDEs	Female	1.80 (1.53-2.06)	2.59 (2.32-2.85)
	Male	1.76 (1.42-2.11)	2.16 (1.82-2.49)
<i>Outer blubber/serum</i>			
Σ DDTs	Female	1.26 (1.12-1.40)	2.27 (2.13-2.41)
	Male	1.31 (1.13-1.50)	1.80 (1.62-1.98)
Σ PCBs	Female	1.23 (1.08-1.39)	2.50 (2.34-2.66)
	Male	1.48 (1.27-1.68)	1.69 (1.49-1.89)
Σ CHLs	Female	1.44 (1.30-1.59)	2.37 (2.23-2.52)
	Male	1.49 (1.30-1.68)	2.00 (1.81-2.18)
Σ PBDEs	Female	1.59 (1.30-1.88)	3.10 (2.81-3.39)
	Male	1.70 (1.32-2.08)	2.22 (1.85-2.59)
<i>Outer blubber/inner blubber</i>			
Σ DDTs	Female	0.93 (0.84-1.02)	1.33 (1.23-1.42)
	Male	0.92 (0.80-1.04)	1.15 (1.04-1.27)
Σ PCBs	Female	0.74 (0.68-0.81)	1.11 (1.05-1.18)
	Male	0.90 (0.82-0.99)	0.99 (0.91-1.07)
Σ CHLs	Female	0.79 (0.72-0.85)	1.14 (1.08-1.21)
	Male	0.85 (0.76-0.93)	1.02 (0.94-1.10)
Σ PBDEs	Female	0.87 (0.76-0.99)	1.27 (1.15-1.38)
	Male	0.97 (0.83-1.12)	1.03 (0.89-1.18)

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