

1 **Title:**

2 Investigating decadal changes in persistent organic pollutants in Scottish grey seal pups

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21

22 **Abstract**

- 23 1. Persistent organic pollutants (POPs) remain a risk to marine ecosystem health.
24 POPs accumulate in fat tissue and biomagnify up food webs, generating high
25 concentrations in apex predators, including marine mammals. Seals are thus
26 often cited as sentinels of marine environment POP levels. Measuring changes
27 across decadal timescales in these animals is key to understanding the
28 effectiveness of regulations controlling POPs, predicting health, population, and
29 ecosystem level impacts and informing conservation and management strategies.
30 However, information on recent changes in legacy POPs in seals is relatively
31 sparse and datasets are not always continuous in the absence of dedicated POP
32 monitoring programmes.
- 33 2. Here, POP concentrations in blubber of weaned grey seal pups from the Isle of
34 May, Scotland were compared between studies investigating POP impacts on
35 survival and energy balance in 2002, and in 2015-2017. By 2017 Σ dioxin-like
36 (DL) and Σ non dioxin-like polychlorinated biphenyls (NDL-CBs) had decreased
37 to ~ 75% of 2002 levels.
- 38 3. The organochlorine pesticides (OCPs), dichlorodiphenyltrichloroethane (Σ DDT),
39 dichlorodiphenyldichloroethylene (DDE) and dichlorodiphenyldichloroethane
40 (DDD), and some CB congeners, did not fall over the 15 year period. However,
41 the power to detect small changes at low concentrations was limited.
- 42 4. High DDE and lack of change in DDD likely reflect low excretion of DDT
43 metabolites rather than recent exposure.
- 44 5. The limited change in many POPs over 15 years suggest that risks remain for
45 grey seal pups' energy balance, endocrine status and immune function, with
46 contingent effects on conservation and management objectives for this species.

47 6. These data highlight the need for long term datasets and parity in sampling and
48 analytical methods to evaluate ongoing impacts of POPs in grey seals and on
49 marine ecosystems more widely.

50

51 **Keywords:** coastal, contaminants, marine mammals, ocean, organochlorine pesticide,
52 polybrominated diethyl ether, polychlorinated biphenyl

53

54 **1. Introduction**

55 Persistent organic pollutants (POPs) are lipid soluble, highly stable, toxic
56 chemicals that have far-reaching negative impacts on aquatic ecosystems (Manzetti,
57 Roos van der Spoel., & van der Spoel, 2014; Sonne et al., 2017). Manufacture and
58 release of legacy POPs into the environment has been tightly regulated or banned,
59 starting with the polychlorinated biphenyls (CBs) in the late 1970s, and, more recently,
60 the polybrominated diethyl ethers (BDEs) in the early 2000s (UNEP, 2001), with more
61 added to the Stockholm Convention list as their effects are realized. Despite these
62 global regulations to control and reduce POP levels, their resistance to chemical,
63 biological and photolytic breakdown and potential for long distance transfer has resulted
64 in high POP concentrations in biota, even in some remote regions of the world,
65 including isolated marine environments (Jamieson, Malkocs, Piertney, Fujii, & Zhang,
66 2017). POPs can either be directly released into marine environments from poorly
67 regulated or unidentified sources (Gómez-Gutiérrez, Garnacho, Bayona, & Albaigés,
68 2007) or transported in the atmosphere and deposited into marine waters from distant
69 regions of the world (Dachs et al., 2002, Jurado et al., 2004). POPs may also be released
70 into marine environments or transferred to organisms via microplastic pollution
71 (reviewed in Cole, Lindeque, Halsband, & Galloway, 2011). Oceans thus act as key
72 sinks for these chemicals and can be slow to respond to regulatory measures that have
73 more immediate impacts in terrestrial environments. Therefore, despite bans on POP
74 production and use, there are still potential routes for these chemicals to enter the
75 marine environment, where they persist for decades. Monitoring POP concentrations is
76 essential to understand their occurrence and fate in ecosystems, evaluate mitigation
77 strategies, identify hotspots of contamination and document negative impacts on marine
78 wildlife (AMAP, 2018; Rigét et al., 2016; 2019; Tanabe, 2002).

79 POPs in natural environments readily bioaccumulate up food chains to generate
80 high concentrations in top predators (Johnson-Restrepo, Kannan, Rapaport, & Rodan,
81 2005), such as marine mammals (Helle, Olsson, & Jensen, 1976a; Ross, Ellis,
82 Ikonomou, Barrett-Lennard, & Addison, 2000). They have the potential to negatively
83 impact marine wildlife, and remain a conservation concern for marine mammal species
84 worldwide (AMAP, 2018; Desforges et al., 2018; Jepson & Law, 2016; Law et al.,
85 2012). POPs such as CBs, BDEs and organochlorine pesticides (OCPs), including
86 dichlorodiphenyltrichloroethane (DDT), have been linked to endocrine disruption (Noël
87 et al., 2017; Routti et al., 2010; Tanabe, 2002; Villanger et al., 2011; Villanger et al.,
88 2013) and negative impacts on reproductive (Helle, Olsson, & Jensen, 1976b; Hoydal et
89 al., 2017; Murphy et al., 2010; Reijnders, 1986) and immune function (Fisk et al., 2005,
90 Hall et al., 2006, Hammond, Hall, & Dyrnyda, 2005, reviewed in Desforges et al., 2016;
91 Penin et al., 2018) in marine mammals. POPs have also recently been connected with
92 lipid disruption in a range of phyla (Lee et al., 2016; Speranza et al., 2016; Yadetie et
93 al., 2017) including marine mammals (Castelli, Rusten, Goksøyr, & Routti., 2014;
94 Robinson et al., 2018; Tartu et al., 2017), because they alter pathways that enable
95 individuals to accumulate or mobilise fat reserves appropriately. Viable immune and
96 endocrine function, successful reproductive efforts and the ability to use energy stores
97 are all crucial to fitness and survival of individuals in the wild. It has been proposed that
98 marine wildlife populations with high POP burdens may be unable to reproduce or
99 survive due to chronic POP effects (Helle et al., 1976b; Ross, 2006), potentially
100 preventing population recovery in some declining species (AMAP, 2018; Desforges et
101 al., 2018, De Guise, Martineau, Béland, & Fournier, 1995; Jepson et al., 2016; Noël,
102 Barrett-Lennard, Guinet, Dangerfield, & Ross, 2009). POPs may negatively impact on
103 juvenile survivorship (Hall, Thomas, & McConnell, 2009) even in populations that are

104 increasing or stable in size, such as UK grey seals, *Halichoerus grypus* (SMRU, 2017).
105 Documenting the local and global trends of POP prevalence in the marine environment
106 is therefore vital for giving context to the population dynamics of wildlife species that
107 are at greatest risk from POP exposure.

108 Marine mammals are particularly vulnerable to the impacts of lipophilic POPs due
109 to their reliance on substantial subcutaneous blubber for fuel (Ofstedal, 1993; Reilly,
110 1991) and insulation (Rosen & Renouf, 1997). Data on trends in marine mammal POP
111 concentrations in the decades following their restriction are well represented in the
112 literature (e.g. Roos, Bäcklin, Helander, Rigét, & Eriksson, 2012). However, data on
113 temporal changes in POP levels in marine mammals since the 2010s are relatively
114 scarce (AMAP, 2018; Houde et al., 2017; Law 2014; Rigét et al., 2016; 2019). In this
115 study, blubber POP levels measured in weaned grey seal pups from 15 years ago (2002)
116 were compared to those measured in recent years (2015-2017) in the same UK North
117 Sea population. The recent changes in POP levels described are intended to facilitate
118 evaluation of mitigation strategies, understand conservation and management
119 implications for this population, and provide POP profiles for comparison with future
120 monitoring. These data will help to identify POP classes that remain a particular risk to
121 grey seals and more widely of ecosystem concern (Bossart, 2011).

122

123 **2. Methods**

124 There is no long-term monitoring programme for POPs in UK North Sea grey
125 seals. However, two separate studies were performed ~15 years apart to investigate the
126 impact of POPs on survivability (in 2002: Hall et al., 2009) and energy balance (2015-
127 2017: Robinson et al., 2018) in grey seal pups. Since these two studies have
128 considerable overlap in methodology and measurements, the data were used to

129 investigate whether changes have occurred in blubber POP concentrations in grey seal
130 pups in this region.

131

132 *2.1 Study site*

133 Field work was conducted on the Isle of May, Scotland (56°11'N, 02°33'W) under
134 permit from Scottish Natural Heritage (SNH). Blubber samples were collected from
135 weaned pups in November to December in 2002 (n = 60; 31 female and 29 male; Hall et
136 al., 2009), and again in 2015 (n = 30; 18 female and 12 male), 2016 (n = 23; 7 female
137 and 16 male: Robinson et al., 2018) and 2017 (n = 29; 14 female and 15 male). All
138 sample collection was performed by personal licence holders/ designated competent
139 personnel under UK Home Office licence 80/1552 in 2002 and 70/7806 in 2015-2017.
140 This work received ethical approval from Abertay University and the University of St
141 Andrews Animal Welfare and Ethics Committee (AWEC) and was performed in
142 compliance with Animal (Scientific Procedures) Act (ASPA) 1986 and the EU directive
143 on the protection of animals used for scientific purposes (2010/63/EU).

144

145 *2.2 Study animals*

146 All study animals were wild grey seals of approximately 3-4 weeks old born on the Isle
147 of May in the year of sampling. All had successfully weaned at a normal weight from
148 their mothers and were in the 1-4 week post-weaning land-based fast (Hall, McConnell,
149 & Barker, 2001; Hall et al., 2009; Reilly, 1991). Pups were observed daily prior to and
150 after weaning. Weaning dates were obtained for all pups except two sampled in 2002.
151 The sex and mass of the individual were recorded at capture.

152

153 *2.3 Sample collection and storage*

154 Biopsy sampling and storage were performed as described previously (Hall et al.,
155 2009; Robinson et al., 2018). Briefly, prior to sampling, pups were given a mass-
156 specific dose of intravenous Zoletil™ and subcutaneous injections of Lignol™ at biopsy
157 sites. A full depth, 6 mm biopsy core was then taken from the dorso-lateral pelvic
158 region (Bennett et al., 2015). The 6 mm biopsy was immediately wrapped in foil and
159 frozen at -20 °C for transport back to the laboratory for POP concentration analysis.

160

161 *2.4 Sample analysis*

162 Samples collected in 2002 were analysed by the Centre for Chemicals
163 Management, Lancaster Environment Centre, Lancaster University, and samples
164 collected in 2015-2017 were analysed at the Centre for Analytical Research and
165 Technology (CART) at the University of Liège, Belgium. CBs and BDEs were analysed
166 at CART in the Department of Chemistry, while OCPs were analysed at CART in the
167 Laboratory of Animal Ecology and Ecotoxicology (LEAE). All POP concentrations are
168 reported as ng g⁻¹ lipid.

169 Samples collected in 2016 and 2017 were extracted and concentrations of the
170 following POPs were measured: six Non Dioxin-Like CBs (NDL-CBs) (CB28, 58, 101,
171 138, 153, 180), eight Dioxin-Like CBs (DL-CBs) (CB105, 114, 118, 123, 156, 157,
172 167, 189), nine BDEs (BDE28, 47, 66, 85, 99, 100, 153, 154, 183) and four OCPs
173 (DDT (summed op'DDT and pp'DDT), and its metabolites,
174 dichlorodiphenyldichloroethane (pp'DDD), dichlorodiphenyldichloroethylene
175 (pp'DDE), and hexachlorobenzene (HCB)). Samples collected in 2015 were only
176 analysed for the six NDL-CBs, nine BDEs and four OCPs. Samples from 2002 were
177 analysed for a larger range of congeners than the 2015 – 2017 samples (see Hall et al.,
178 2009). Therefore, for the current study the number of congeners from the 2002 data

179 included in the statistical analysis was restricted to match the congeners analysed in the
180 2016 – 2017 and data from 2015 was excluded from analysis of DL-CBs.

181 Methodological details of the extraction and detection methods used to analyse the
182 2002 samples are given in Hall et al. (2009). Briefly, samples were extracted with
183 dichloromethane using the Soxhlet extraction method. An aliquot was taken for
184 gravimetric lipid determination. Samples were spiked with seven ¹³C-labelled CBs and
185 ¹³C-BDE 209 before extraction. Samples were cleaned using silica gel treated with
186 sulphuric acid and secondarily with gel permeation chromatography. Samples were
187 analysed using a gas chromatography – mass spectroscopy (GC-MS) system in selected
188 ion monitoring (SIM) mode using an extracted ion chromatogram (EIC) source with
189 two ion masses monitored for each chemical. OCPs were analysed in electron ionization
190 (EI) mode and BDEs were analysed using GC-quadrupole MS in negative chemical
191 ionization (NCI) mode.

192 For the 2015 – 2017 samples, the analytical protocol was common at its early
193 stages for all the POPs. Briefly, approximately 200 mg wet weight of blubber
194 underwent Accelerated Solvent Extraction (ASE, Dionex 200, Thermo, US) using a
195 mixture of hexane: dichloromethane (90:10; V:V) at 125°C and 1.304 10⁷ Pa. The fat
196 content was determined gravimetrically after solvent evaporation with a TurboVap LV
197 concentration Evaporator workstation (Zymark TurboVap®LV, Charlotte, USA). The
198 fat was then re-dissolved in 40 ml of hexane and split into two equal fractions; one for
199 CB and BDE determination, and the other one for OCPs. For the CBs and BDE
200 fraction, known amounts of ¹³C labelled homologues in a standard solution containing
201 all the targeted CBs and BDEs was added to each sample as an internal standard. The
202 extract was then concentrated in 1 mL before being loaded for clean-up on a multilayer
203 basic alumina and acid silica column. This system removes major matrix interferences

204 from the extracts and separates the OCPs and mono-ortho fraction from the dioxin-like
205 compound fraction. The column was eluted with hexane, followed by a solvent
206 exchange to nonane in 100 μL as final volume. Recovery standards ($^{13}\text{C}_{12}$ BDE 77,
207 $^{13}\text{C}_{12}$ BDE138 and $^{13}\text{C}_{12}$ CB 80) were added into the vial prior to GC – high resolution
208 MS (GC-HRMS) analysis. The analysis was performed with an Autospec Ultima High
209 Res Mass Spectrometer (Waters, US) coupled to an Agilent 6890 GC (Agilent, US).
210 The injection was carried out in splitless mode and the mass spectrometry via electron
211 ionization EI (@40 eV) using SIM mode. For quantification of the OCPs, 50 μL of a
212 hexanic solution (100 $\text{pg } \mu\text{L}^{-1}$) of CB 112 (Dr. Ehrenstorfer®, Augsburg, Germany)
213 were added to the extract as a surrogate internal standard to evaluate the efficiency of
214 recovery. The clean-up of these extracts was conducted using H_2SO_4 98% and then
215 Florisil solid phase enrichment (Supelco, Envi-Florisil, Bellefonte, PA), as in Damseaux
216 et al., (2017). Five μL of nonane were added to the purified extract as a keeper. Each
217 extract was evaporated under a gentle stream of nitrogen until only the keeper remained
218 in the vial. The final extract was reconstituted with 45 μL of n-hexane and 500 μL of
219 Mirex (100 $\text{pg } \mu\text{L}^{-1}$ in hexane) as injection volume internal standard (Dr. Erhenstorfer®
220 GmbH, Augsburg, Germany). Neither Mirex nor CB112 were detected in the samples
221 during pre-test analysis, allowing their use as standards. Trace CB 209 was found in
222 samples, precluding its use as a standard. Finally, the extracts were analysed by high-
223 resolution GC (Thermo Quest Trace, 2000; Thermo Quest, Milan, Italy) equipped with
224 a ^{63}Ni electron capture detector (ECD) and on column injector to prevent pp' DDT from
225 being degraded in split/ splitless injectors (Muir & Sverko, 2006). OCPs were analysed
226 on a 60 m x 0.25 mm (0.25 mm film) DB5 ms capillary column (J &W Scientific,
227 USA). Other analytical parameters are described in Debier et al. (2003a). Quantification
228 was performed using internal standards. A calibration curve (1.5 - 250 $\text{pg } \mu\text{L}^{-1}$) was

229 established for each compound of interest. Confirmation of the identity and
230 concentrations of the compounds of interest were periodically performed by GC-HRMS
231 (Trace GC Ultra and ITQ 1100 from ThermoQuest). The transfer line temperature was
232 kept at 290 °C and the ion trap temperature was set at 250 °C. The electron ionization
233 (EI) was performed at 70 eV and the ion trap was operating in MS/MS mode. The
234 quality control (QC) was pork fat, free of the compounds of interest. The pork fat was
235 spiked with 5 ng g⁻¹ lipid weight (lw) nominal concentrations of a mixture of the OCPs
236 of interest, forming the QC. The OCP concentrations in each sample and in the QC were
237 corrected for initial fat weight, and the recovery percentage of the surrogate CB 112.
238 Recovery rates in QCs ranged between 96% ± 10.3 % and 113 % ± 12.5 % according to
239 the OCP and the recovery rates of the CB 112 surrogate internal standard was always
240 between 70 and 110 %, in good agreement with requirements of SANCO (SANCO,
241 2014). The limit of detection (LOD) was 0.02 ng g⁻¹ lw. The measured limit of
242 quantification (LOQ) determined with CB spiked pork fat was 0.07ng g⁻¹ lw.

243

244 *2.5 Statistical analysis*

245 All statistical analyses were performed using R 3.4.1 (R Development Core Team,
246 2012). Marine mammal species typically show high individual variability in POP
247 concentrations (Bjurlid, Dam, Hoydal, & Hagberg, 2018; Pomeroy et al., 1996). The
248 smaller sample sizes for the three recent sampling years (2015: n = 30, 2016: n = 23,
249 2017: n = 29) compared to 2002 (n = 60) may thus be more strongly affected by the
250 presence of a few individuals with very low or high POP concentrations in their blubber
251 tissue. All data from 2015, 2016 and 2017 were therefore combined to generate a
252 sample size more comparable with that collected in 2002 for individual congeners (n =
253 82 for NDL-CBs and OCPs 2015-2017; n = 52 for DL-CBs 2016-2017). In addition to

254 individual congener analysis, POPs were also summed into Σ NDL-CBs, Σ DL-CBs,
255 Σ BDEs and Σ DDXs (DDT, DDD, DDE). CB and DDX concentrations were analysed
256 for differences between 2002, and 2015 to 2017. Methodological differences in the
257 standards used for spiking between the Lancaster University and the University of Liège
258 laboratories for detection of BDEs precluded statistical comparison between the 2002
259 and 2015-2017 datasets for this class of POPs.

260 As the distribution of POP concentrations was positively skewed, a generalised
261 linear model (GLM) with a Gamma distribution and a log link function was used to
262 investigate (a) the relationship between POP concentrations and sex, mass and days
263 postweaning (Debier et al., 2006; Louis et al., 2014; 2016) as potential confounders and
264 (b) the relationship between POP concentrations and time of sampling (year group),
265 taking into account the effect of the confounders if necessary. Model fits to the data
266 were assessed using residual analysis and outliers were identified from QQ plots and
267 using Cook's distance.

268 A power analysis was performed using G*Power software (Faul, Erdfelder, Lang,
269 & Buchner 2007) to determine the probability of detecting a change between 2002 and
270 2015-2017 (or 2016-2017 for DL-CBs), on the basis of the sample sizes used, and
271 observed means and standard deviations for all summed POP classes.

272

273 **3. Results**

274 *3.1 Summary of detected POP concentrations in weaned grey seal pups*

275 The median concentrations and ranges of the summed and individual POPs
276 detected in grey seal pup blubber tissue are given in Table 1. The CB profiles detected
277 in pups from the Isle of May were consistent between year groups for both the NDL-
278 CBs and DL-CBs measured in both studies, with the following abundance pattern for

279 the different congeners; NDL-CB 153 > 138 > 180 > 101 > 52 > 28; DL-CB 118 >105
280 = 156 > 114 = 157 > 189, 167 > 123.

281 For the 8 BDEs measured in all years, the profiles have remained consistent over
282 time despite different methods used to measure them. BDE 47 was consistently the most
283 abundant congener, and BDEs 66 and 183 were the lowest in abundance. BDE 99 was
284 consistently one of the more abundant congeners. BDEs 153 and 154 have been of
285 consistently low concentration. The profiles of OCP abundances followed the same
286 pattern between year groups: DDE > DDT > HCB > DDD.

287

288 *3.2 Analysis of potentially confounding variables; pup mass and days post-weaning*

289 There was no relationship between pup mass and the blubber concentration of any
290 of the POPs nor between the sex of the pup and blubber POP concentrations, either as
291 summed groups or on an individual congener basis (GLMs, all $p > 0.05$). The fit of the
292 models to the data was investigated by residual analysis. Six of the data points in both
293 model groups (sex and mass) were identified as outliers that were causing some
294 skewness in the residuals. Removal of these data points resulted in residuals with
295 normal distribution, but no change in any of the results. Therefore, there was no need to
296 include sex or mass as additional explanatory variables in the final model. There was
297 also no relationship between days postweaning and the concentration of any of the
298 POPs, again either as summed groups or by individual congener (again after the
299 removal of the outliers to ensure the results were robust, GLMs, all $p > 0.05$). This
300 allowed a simple model with time of sampling (i.e. year group) as the explanatory
301 variable and POP concentration as the dependent variable to be fitted to the data.

302

303 *3.3 Comparison of CBs between year groups*

304 Blubber from weaned grey seal pups from the Isle of May contained lower
305 concentrations of Σ NDL-CBs in 2015-2017 than in 2002 by ~25% (Figure 1; Table 2).
306 Typically, animals in 2002 had higher levels of all NDL-CB congeners than in 2015 –
307 2017 (Figure 2; Table 2). CB 28 was 9 fold lower in 2015-2017 than 2002. Smaller
308 declines were apparent in the more abundant congeners: CB 138 was lower by 35%,
309 and CB 153 and CB 180 by ~20%. CB 101 did not differ between years, and CB 52,
310 one of the least abundant congeners, was ~30% higher in the more recent samples
311 (Figure 2; Table 2).

312 Concentrations of Σ DL-CBs in blubber from the weaned grey seal pups were
313 ~25% higher in 2002 than 2016-2017 (Figure 3; Table 2). CBs 114, 123, 156, 157, 167
314 and 189 were higher in 2002 compared to 2016 and 2017 (Figure 4; Table 2). The
315 degree of reduction differed between congeners. In 2016-2017, CB189 was ~50%
316 lower, CBs 114 and 156 were ~35% lower, and CB 157 was ~20% lower than in 2002.
317 The congeners with the lowest abundance, CB 123 and CB 167, were 98% and 75%
318 lower in 2016-2017, respectively. There was no significant decrease from 2002 to
319 2015-2017 in two major DL-CBs, CB 105 and CB 118 (Table 1 and 2).

320

321 *3.4 Comparison of OCPs between year groups*

322 There was no change in Σ DDX concentrations between 2002 and 2015-17 in blubber
323 from grey seal pups (Figure 5; Table 2). However, DDT was ~70% lower in 2015-2017
324 than 2002 (Figure 6; Table 2). In contrast, DDE increased by ~46%, and DDE as a
325 proportion of DDX was 0.67 in 2002 but 0.88 in 2015-2017. Neither DDD nor HCB
326 changed over the same period (Figure 6; Table 2).

327

328 *3.5. Power analysis*

329 The power to detect an effect size of 0.5 for Σ NDL-CBs with the sample sizes
330 here size was 0.876. Similarly, for the total Σ CBs (n = 60 from 2002 and n = 52 from
331 2016-2017) the power to detect a medium effect (Cohen, 1988) of 0.5 was
332 0.956. However, where the concentrations were much lower, the power to detect an
333 effect was reduced. For Σ DL-CBs, the power to detect the effect size of 0.26 seen here
334 was only 0.27. For Σ DDX the effect size here was 0.36, and the power to detect this size
335 of change was 0.474.

336

337 **4. Discussion**

338 *CB profiles and concentrations*

339 The abundance profile of CBs in blubber from postweaned Isle of May grey seal
340 pups did not change substantially between year groups. CB 153 was the most abundant
341 congener, followed by CB 138, which is directly comparable to the most abundant
342 congeners detected in previous studies on North Sea grey seals, both in blubber
343 (Vanden Berghe et al., 2012) and serum (Debier et al., 2003b; Vanden Berghe et al.,
344 2012) and in phocid seals from other regions (Ross et al., 2013; Shaw, Brenner,
345 Bourakovsky, Mahaffey, & Perkins, 2005; Weijs, Das, et al., 2009). PCB concentrations
346 here were approximately 10 fold lower than those found in grey seal pups during the
347 late 1990s – early 2000s in heavily polluted regions, such as the Baltic Sea (Sørmo,
348 Skaare, Jüssi, Jüssi, & Jenssen., 2003) and 20-150 fold lower than found in juvenile and
349 adult harbour seals from the North Sea in 2006-2008 (Weijs, Dirtu, et al., 2009). The
350 median concentrations and ranges of NDL-CBs detected in this study in 2002 were
351 comparable to those in grey seal pup in the Norwegian Sea and Gulf of St Lawrence
352 (Sørmo et al., 2003) and in harbour seal pups during the late 1990s – early 2000s from
353 the North Eastern Pacific (Ross et al., 2013). Data specifically documenting trends in

354 DL-CBs in wild marine mammal species are limited (Law, 2014). However, one study
355 reporting individual congener concentrations for four of the DL-CBs detected in this
356 study (DL-CB 105, 118, 156, 157) exists for grey seal pups in the late 1990s for the
357 Baltic Sea, East and West Atlantic (Sørmo et al., 2003) and concentrations were more
358 than double those detected here. The summed CB concentrations from 2015 -2017 here
359 were comparable to those in juvenile and subadult ringed seals from Greenland and
360 coastal USA and Canada sampled in 2009-2011 (Brown et al., 2014; Rigét et al., 2016).
361 Concentrations in adult ringed seals from 2009-2011 from the Labrador coast were two
362 to four-fold higher (Brown et al., 2014). Ringed seals from the Baltic in 2015 had
363 concentrations that were an order of magnitude higher for both NDL and DL-CBs than
364 concentrations reported here (Bjurlid, Roos, Ericsson-Jogsten, & Hagberg, 2018).

365

366 *Differences in CB concentrations between year groups in Isle of May pups*

367 There was a modest, ~25% reduction in both NDL- and DL-CBs in the blubber
368 of North Sea grey seal pups from 2002 to 2015-2017. Despite a large gap in the timing
369 of the data collection, the power to detect this change in total CBs and NDL-CBs was
370 high. However, except for CB 52 and 101, NDL-CB congeners analysed here showed at
371 least a marginal decrease over the 15 year time period, which is consistent with several
372 studies reporting that the declines in the environment in this group of chemicals have
373 slowed down after an initial rapid drop during the 1980s – early 1990s (Aguilar, Borrell,
374 & Reijnders, 2002; Bjurlid, Roos, et al., 2018; Jepson et al., 2016; Law et al., 2012;
375 Law, 2014; Rigét et al., 2006; Rigét et al., 2016; Rigét et al., 2019; Ross et al., 2013;
376 Vorkamp, Rigét, Bossi, & Dietz, 2011).

377 The most abundant DL congener (CB 118) did not significantly change between
378 2002 and 2015-2017. However, for Σ NDL-CBs there was limited power to detect the

379 effect size of 0.26 seen here, and the ability to detect changes in individual congeners is
380 lower still, such that modest changes in CB 105 and 118 (~9% and ~7%, respectively)
381 are not large enough to be reliable with the sample size in this study. The limited power
382 to detect the observed change in DL-CBs results from their lower abundance, slow rate
383 of change and high degree of variation. This limited power is consistent with other
384 studies showing that long-term monitoring over decades is required to identify time
385 trends (AMAP 2018; Rigét et al., 2016; 2019). Indeed, only 12% of long-term studies
386 have the ability to detect a 5% annual change in most POPs, and require more than 20
387 years of data to detect such a change (Rigét et al., 2019).

388

389 *BDE profiles and concentrations*

390 The BDE profiles from 2015-2017 are similar to the same population of grey
391 seals, from the late 1990s and in 2008 (Kalantzi, Hall, Thomas, & Jones, 2005; Vanden
392 Berghe et al., 2012), blubber from grey seals elsewhere (Ikonomidou & Addison, 2008),
393 and recent previous studies on other marine mammals (Bjurlid, Dam, et al., 2018; Muir
394 et al., 2006; Ochiai et al., 2017; Ramu, Kajiwara, Tanabe, Lam, & Jefferson, 2005; Rigét
395 et al., 2006; Ross et al., 2013; Weijs, Dirtu et al., 2009). BDE 47 was always the most
396 abundant congener, and accounted for 12.5 – 88.1% (median: 80.9%) of BDEs detected.

397 BDE levels reported here from weaned pups in 2002 were 2-7 fold lower than
398 the values reported recently in maternal grey seal blubber in the UK from 2008 (Vanden
399 Berghe et al., 2012). BDE values were comparable with maternal blubber from
400 populations in Canada in 1995 (Ikonomidou & Addison, 2008). Recent BDE values here
401 are 10-20 fold lower than levels in adult harbour seals from the North Sea from 1999-
402 2004 (Weis, Dirtu et al., 2009), and an order of magnitude lower than values reported in
403 UK grey seal pup blubber from 1998-2000 (Kalantzi et al., 2005) and in Salish Sea

404 harbour seal pup blubber from 1984-2009 (Ross et al., 2013). Findings from other
405 studies in the marine environment globally suggest that BDE levels peaked in the early
406 2000s and then fell, as a result of the ban on the penta- and tetra-mixes of BDEs
407 (Airaksinen et al., 2014; Bjurlid, Dam, et al., 2018; Bjurlid, Roos, et al., 2018; Houde
408 et al., 2017; Rigét et al., 2006; 2019; Ross et al., 2013; Sanganyado, Rashid, Rajput,
409 Liu, 2018; Shunthirasingham et al., 2018) and may have stabilized recently in some
410 regions (Brown et al., 2018; Fernandes et al., 2018; Houde et al., 2017; Rigét et
411 al., 2016; 2019). In combination with technical disparities discussed below, the BDE
412 data here do not provide a sufficient recent time-series to determine whether the bans
413 have been effective at reducing BDEs in grey seal blubber, but provide a starting point
414 for future studies or comparisons to investigate the efficacy of the ban and its impact on
415 levels in top predators.

416

417 *OCP profiles and concentrations*

418 The profiles of OCP abundances DDE > DDT > DDD matches previous results
419 from phocid seals (Sørmo et al., 2003) and small cetaceans (Méndez-Fernandez et al.,
420 2018). Concentrations of DDE were ~20%, and DDT were 20-60%, of those found in
421 pups and juveniles from the same population in 1998-2000 (Kalantzi et al., 2005). They
422 were 2.7- 3.5 fold lower than the concentrations found in inner and outer blubber of Isle
423 of May adult females during lactation in 2008 (Vanden Berghe et al., 2012).
424 Concentrations were comparable to those detected in grey seal pups from the Gulf of St
425 Lawrence and the Norwegian Sea in 1995, but approximately 16 fold lower than levels
426 in pups from the Baltic (Sørmo et al., 2003). ΣDDX in all years of the current study
427 were comparable to concentrations detected in ringed seals from Greenland across 1999
428 – 2004 (Vorkamp et al., 2008). HCB concentrations remained low across all years.

429

430 *Differences in OCP concentrations between year groups in Isle of May pups*

431 The concentrations of DDT more than halved from 2002 to recent years of
432 analysis. This difference was expected, because DDT is mainly converted to DDE or
433 DDD over time, which are both more stable in organisms than the original compound
434 (Ricking & Schwarzbauer, 2012). Provided that no new sources of DDT enter an
435 environment, the ratio of DDE: DDX in organisms should shift to greater than 0.6
436 (Aguilar, 1984). Ratios calculated here indicate that there have been no new sources of
437 DDT in the North Sea since 2002. For our data, DDE: DDX ratios have shifted from a
438 mean of ~0.6 in 2002 to means of ~0.9 in all recent years of analysis. Of the 60 seal
439 pups sampled in 2002, 17 had ratios of < 0.6, indicating that in 2002 there may have
440 been a source of DDT where their mothers foraged.

441 Similar to the patterns of CB decline in biota over the last decades, OCP
442 concentrations initially fell steeply in the 1970s – 1990s after their use was banned, but
443 reductions have slowed or plateaued in a variety of marine top predator species since the
444 2000s (Roos et al., 2012). There are many studies reporting downward trends, or a lack
445 of them, for DDX values in marine mammals before around 2010 (Brown et al., 2018;
446 Rigét et al., 2016). Significant declines of ~8% annually in DDX have been reported for
447 ringed seals (Greenland 1986-2013, Rigét et al., 2016; Greenland 1994-2006, Vorkamp
448 et al., 2008; Canadian Arctic 1998- 2010; Brown et al., 2018); polar bears (Greenland
449 1984-2013, Rigét et al., 2016; Canadian Arctic 1989- 2008; Brown et al., 2018);
450 harbour porpoise (UK 1992 – 2006, Law et al., 2012); and spotted dolphin (four regions
451 in the Atlantic 2005 -2015, Méndez-Fernandez et al., 2018), while studies showing no
452 declining DDX trends exist for South American sea lions (Argentina 1991-2005;
453 Borrell, Garcia-Solà, Aguilar, García, & Crespo, 2010), striped dolphins (Japan 1978-

454 2003; Isobe et al., 2009) and harbour seals (North-western Atlantic 1991-2001; Shaw et
455 al., 2005). The current results also show a drop in DDT, an increase in DDE and no
456 change in DDD and HCB between 2002 and 2015- 2017. This is typical of time trends
457 in these chemicals in a range of biota from the Arctic, which generally show falling
458 DDT, but no change in DDD and DDE in marine mammals (Rigét et al., 2019). The
459 limited power to detect differences in Σ DDX here is in line with recent work showing
460 similar difficulties even in long term monitoring programmes (Rigét et al., 2019).

461

462 *Methodological and technical considerations*

463 The dependence of variability expressed as relative standard deviation on
464 concentration is typical of the Horwitz trend that is a well-recognized phenomenon in
465 POP analysis, and variability in POP measurements between labs is also typical from
466 samples of high fat content (De Boer & Wells, 2006), such as blubber. Since samples
467 were not available from 2002 to reanalyse, and the ability to replicate the 2002 method
468 using recent samples was not an option, an interlaboratory comparison was not possible
469 to conduct. However, extraction, clean-up and detection methods used in each case are
470 accepted and equivalent approaches for PCB and OCP analysis (Covaci, Voorspoels,
471 Ramos, Neels, & Blust, 2007; Muir & Sverko, 2006; Van Leeuwen, van Bavel, & de
472 Boer 2013; Wang et al., 2010).

473 The standards used to measure BDEs in 2002 and more recent years differed.
474 This is probably the most significant parameter that could explain the discrepancies
475 observed for BDEs and is the reason we avoided a direct statistical comparison. While
476 the apparent 4-fold reduction in BDEs since 2002 seen here is consistent with recent
477 downward trends from the early 2000s, a direct comparison cannot be made with the
478 more recent data because the samples from 2002 were analysed using only ^{13}C BDE

479 209 as the internal standard (Hall et al., 2009), rather than using standards for each
480 congener. Since BDE 209 can degrade during high temperature separation in the GC
481 column to lower brominated congeners (Stapleton, 2006), the older method that used
482 ¹³C BDE 209 as a standard for detection may contain some inaccuracies in the levels
483 measured, which could overestimate between year differences. In conjunction with the
484 high degree of variability in the measurement of low abundance congeners in recent
485 samples, although the data are strongly suggestive of a decline, it is difficult to conclude
486 that a real reduction in BDEs has occurred in this population.

487 Using the same laboratory methods decades apart can be challenging in the face
488 of necessary ongoing efforts to refine the analytical methods and improve
489 reproducibility between labs (Abalos et al., 2013; De Boer & Wells, 2006; Covaci, et
490 al., 2007). The discrepancies in sampling and analysis that prevent direct comparison
491 here highlight one difficulty in comparing historical with contemporary data and in
492 obtaining comparable results for long-term data analysis. This argues for the need for
493 standardisation in sampling protocols where possible, and banking of tissue samples
494 using appropriate storage to allow historical and contemporary samples to be analysed
495 at the same time (Rigét et al., 2016; 2019).

496

497 *Conservation and management implications*

498 UK grey seal numbers are currently stable or increasing throughout their
499 monitored range (SMRU, 2017), suggesting their population status is not under threat.
500 However, POPs are important influencers of first year survival in grey seal pups, which
501 is a key component of the species' population dynamics (Hall et al., 2009). The
502 mechanism linking POPs with survival is unknown and may be multifaceted, including
503 through endocrine disruption and reduced immunocompetence. Although POP

504 concentrations in North Sea grey seal pups in this study are all well below the estimated
505 toxic threshold values (Kannan, Blankenship, Jones, & Giesy, 2000), summed
506 concentrations of DL-CB and DDX have been linked to altered glucose uptake, lactate
507 production and lipolytic rates in the blubber tissue of the same pups that provided the
508 2016 data for this study (Robinson et al., 2018). Even apparently low concentrations of
509 these chemicals may thus have significant physiological impacts with potential health
510 and energetic consequences early in life. In addition, POP effects are not additive and
511 the cocktail present in the animal may produce effects at levels below accepted
512 thresholds for individual congeners or POP classes. While concentrations of CB 118 in
513 the blubber of grey seals showing immunosuppression was higher (1998, n = 17,
514 median: 98 ng g⁻¹ lipid, range: 52 – 211 ng g⁻¹ lipid, Sørmo et al., 2009) than levels
515 detected here (combined data 2016 – 2017, n = 52, median: 10.2 ng g⁻¹ lipid, range: 3.7
516 – 92.9 ng g⁻¹ lipid), the two ranges do overlap. The lack of significant changes in the
517 most abundant DL-CB, CB 118, and in DDX in the blubber of grey seal pups thus have
518 potential conservation implications. Continued reductions in POPs would thus likely
519 have beneficial effects on first year survival probability for grey seal pups, with
520 potential contingent effects on population structure and dynamics.

521

522 *Conclusions*

523 Despite decades of regulations restricting the use of POPs globally, these persistent,
524 toxic chemicals are still present in marine ecosystems and organisms. After substantial
525 initial drops in the 1970s – 1990s, declines have slowed or stalled in many regions. The
526 findings from grey seal pups in the North Sea here concur with this global pattern, and
527 provide some of the most recent data for POP concentrations in these top marine
528 predators. In the absence of long term monitoring, we have shown that comparisons can

529 be made between POP measurements from discrete studies many years apart,
530 particularly for CBs, but attention should also be paid to methodological differences,
531 which may make spatial and temporal comparisons difficult. The current study
532 highlights that a lack of time series data can make interpretation of spot measures
533 challenging, and emphasises the need for continued, standardised monitoring of POP
534 concentrations. Larger sample sizes and longer time series are needed to reliably detect
535 changes in low abundance summed POPs and individual congeners. Such work will
536 allow better prediction of population impacts, particularly when animals face multiple
537 threats, which will improve conservation efforts directed at grey seals and other species.

538 While the POP concentrations detected in this study are below values that cause
539 severe toxic effects, even these ‘low’ concentrations are enough to cause endocrine
540 disruption and alter physiological pathways, with unknown consequences for individual
541 health and survival. The fact that low levels of POPs decrease the probability of survival
542 and that levels are slow to decline should be taken into consideration in the
543 development of conservation and management plans, because these compounds may
544 continue to affect population structure as well as dynamics into the future.

545

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562

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900 **Tables**

901 Table 1. Median (range) blubber concentrations of individual and summed POP
 902 congeners, % blubber lipid and body mass at capture of weaned grey seal pups from the
 903 Isle of May in 2002, 2015, 2016 and 2017. All concentrations are in ng g⁻¹ lipid.

	2002	2015	2016	2017
NDL PCBs				
28	13.0 (8.0 – 37.4)	6.2 (0.8 – 33.3)	1.4 (0.4 – 62.6)	0.4 (0.4 – 32.3)
52	12.3 (6.0 – 63.0)	24.7 (7.4 – 127.5)	16.3 (4.6 – 211.6)	8.4 (0.8 – 146.7)
101	25.6 (17.0 – 199.0)	28.7 (4.1 – 76.8)	36.4 (20.1 – 193.8)	19.1 (10.6 – 150.4)
138	216.2 (104.0 – 1842.0)	142.9 (50.7 – 319.8)	155.4 (91.8 – 309.4)	128.1 (66.8 – 526.9)
153	318.5 (129.0 – 2613.0)	302.1 (107.6 – 661.2)	260.3 (151.9 – 547.7)	226.3 (110.7 – 1108.9)
180	59.1 (17.0 – 734.0)	53.6 (21.1 – 210.1)	43.9 (14.3 – 122.6)	35.1 (16.5 – 479.9)
∑NDL-PCB	635.1 (316.0 – 5165.8)	545.5 (211.3 - 1275.50)	541.7 (211.3 - 1275.5)	412.9 (219.9 – 2192.9)
DL PCBs				
105	4.0 (1.6 – 21.5)		3.8 (0.9 – 38.3)	3.4 (1.7 – 21.9)
114	1.4 (0.8 – 5.2)		1.2 (0.7 – 3.5)	0.4 (0.1 – 2.8)
118	11.1 (5.9 – 67.6)		10.7 (3.7 – 92.9)	9.8 (4.1 – 63.1)
123	0.9 (0.6 – 7.9)		0.02 (0.0 – 1.5)	0.02 (0.02 – 0.9)
156	5.2 (2.1 – 47.6)	NA	3.2 (1.7 – 9.4)	3.9 (1.9 – 20.9)
157	1.0 (2.1 – 47.6)		0.8 (0.5 – 2.2)	1.1 (0.5 – 5.5)
167	0.6 (1.4 – 1.6)		0.2 (0.1 – 2.3)	0.1 (0.01 – 0.9)
189	0.9 (0.01 – 0.9)		0.6 (0.2 – 1.9)	0.5 (0.2 – 4.0)
∑DL-PCBs	26.2 (14.1 – 153.8)		19.8 (9.6 – 151.2)	19.3 (9.0 – 100.8)
PBDE				
28	9.4 (6.0 – 23.3)	0.2 (0.1 – 0.5)	0.2 (0.1 – 1.2)	0.1 (0.1 – 0.4)
47	67.1 (14.0 – 288.0)	18.4 (8.8 – 57.3)	16.4 (7.9 – 58.7)	13.5 (6.2 – 60.9)

66	1.6 (1.0 – 4.1)	0.0 (0.0 – 0.1)	0.0 (0.0 – 0.4)	0.005 (0.005 – 0.2)
	2002	2015	2016	2017
85	5.3 (3.4 – 13.5)	1.6 (0.7 – 3.9)	0.1 (0.0 – 0.3)	0.1 (0.005 – 0.4)
99	6.4 (4.0 – 28.0)	0.7 (0.3 – 4.4)	0.7 (0.3 – 5.6)	0.4 (0.1 – 4.1)
100	4.4 (2.2 – 19.7)	0.1 (0.0 – 0.3)	1.5 (0.7 – 3.3)	1.1 (0.5 – 3.2)
153	5.3 (4.0 – 14.0)	0.5 (0.2 – 2.0)	0.4 (0.2 – 1.8)	0.3 (0.1 – 2.2)
154	3.8 (2.0 – 28.0)	0.6 (0.2 – 8.9)	0.4 (0.2 – 1.0)	0.4 (0.2 – 1.5)
183	2.8 (1.8 – 7.2)	0.0 (0.0 – 43.4)	0.0 (0.0 – 0.2)	0.005 (0.005 – 0.1)
ΣPBDE	104.3 (69.6 – 344.0)	23.4 (10.4 – 67.7)	23.2 (10.2 – 70.1)	16.4 (7.6 – 69.2)
OCP				
DDT	72.5 (22.0 – 450.3)	21.1 (8.2 – 50.9)	21.9 (16.6 – 35.7)	16.6 (4.4 – 89.8)
DDD	8.1 (4.2 – 27.3)	20.3 (8.1 – 55.2)	6.5 (1.9 – 13.4)	3.6 (0 – 14.3)
DDE	140.0 (10.4 – 1046.5)	288.3 (146.7 – 573.6)	180.6 (108.4 – 301.5)	185.4 (111.5 – 477.9)
ΣDDX	219.3 (67 – 1524)	331.3 (170.7 – 666.6)	209.6 (128.4 – 345.7)	206.9 (126.3 – 582.1)
HCB	8.9 (1.7 – 45.4)	11.8 (4.1 – 68.7)	9.5 (0.0 – 45.1)	6.9 (2.3 – 58.2)
% lipid	Data unavailable	88.0 (39.6-93.0)	85.8 (56.3-88.8)	84.2 (77.9-89.2)
Body mass at capture (kg)	41.7 (28.7-56.4)	41.8 (31.8-50.0)	39.8 (32.8-50.4)	37.2 (23.2-48.8)

904

905

906 Table 2: Generalised Linear Model results with a Gamma distribution and a log link
 907 function (F values from comparison with null, intercept only model using ANOVA and
 908 model p values) comparing blubber POP concentrations from weaned grey seal pups in
 909 2002 (n = 60) and 2015-2017 (NDL-PCBs and OCPs; n = 82, Degrees of freedom =
 910 140) or 2016-2017 (DL-PCB ; n = 52, Degrees of freedom = 110). Bold indicates a
 911 significant (p < 0.05) difference.

	F	p
NDL PCBs		
28	22.8	<0.001
52	8.4	0.004
101	0.02	0.894
138	29.3	<0.001
153	11.9	<0.001
180	8.5	0.004
∑NDL-PCB	14.7	<0.001
DL PCBs		
105	0.50	0.482
114	24.9	<0.001
118	0.67	0.404
123	40.7	<0.001
156	13.4	<0.001
157	9.2	0.002
167	28.1	<0.001
189	14.8	<0.001
∑DL-PCBs	4.3	0.040
OCP		
DDT	163.9	<0.001
DDE	5.1	0.020
DDD	0.97	0.222
∑DDX	0.05	0.823
HCB	2.0	0.160

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914

915 **Figure legends**

916

917 Figure 1. Median concentrations of Σ NDL-CBs in blubber from weaned grey seal pups
918 from 2002 (n = 60) and 2015-2017 (n = 82) on the Isle of May, with upper and lower
919 quartiles, 1.5 x interquartile range and outliers Red * shows a significant reduction
920 (GLM, p < 0.05).

921

922 Figure 2. Median NDL-CB concentrations for each congener measured in blubber
923 from weaned pups in 2002 (n = 60) and 2015-2017 (n = 82) on the Isle of May, with
924 upper and lower quartiles, 1.5 x interquartile range and outliers. Red * shows when
925 levels were significantly lower (GLM, p < 0.05). Note differences in y axis ranges to
926 allow visualisation of data range for each congener.

927

928

929 Figure 3. Median concentrations of Σ DL -CBs in blubber from weaned grey seal pups
930 from 2002 (n = 60) and 2016-2017 (n = 52) on the Isle of May, with upper and lower
931 quartiles, 1.5 x interquartile range and outliers. Red * shows a significant reduction
932 (GLM, p < 0.05).

933

934 Figure 4. Median DL-CB concentrations for each congener measured in blubber from
935 weaned pups in 2002 (n = 60) and 2016-2017 (n = 52) on the Isle of May, with upper
936 and lower quartiles, 1.5 x interquartile range and outliers. Red * shows when levels
937 were significantly lower (GLM, p < 0.05). Note differences in y axis ranges to allow
938 visualisation of data range for each congener.

939

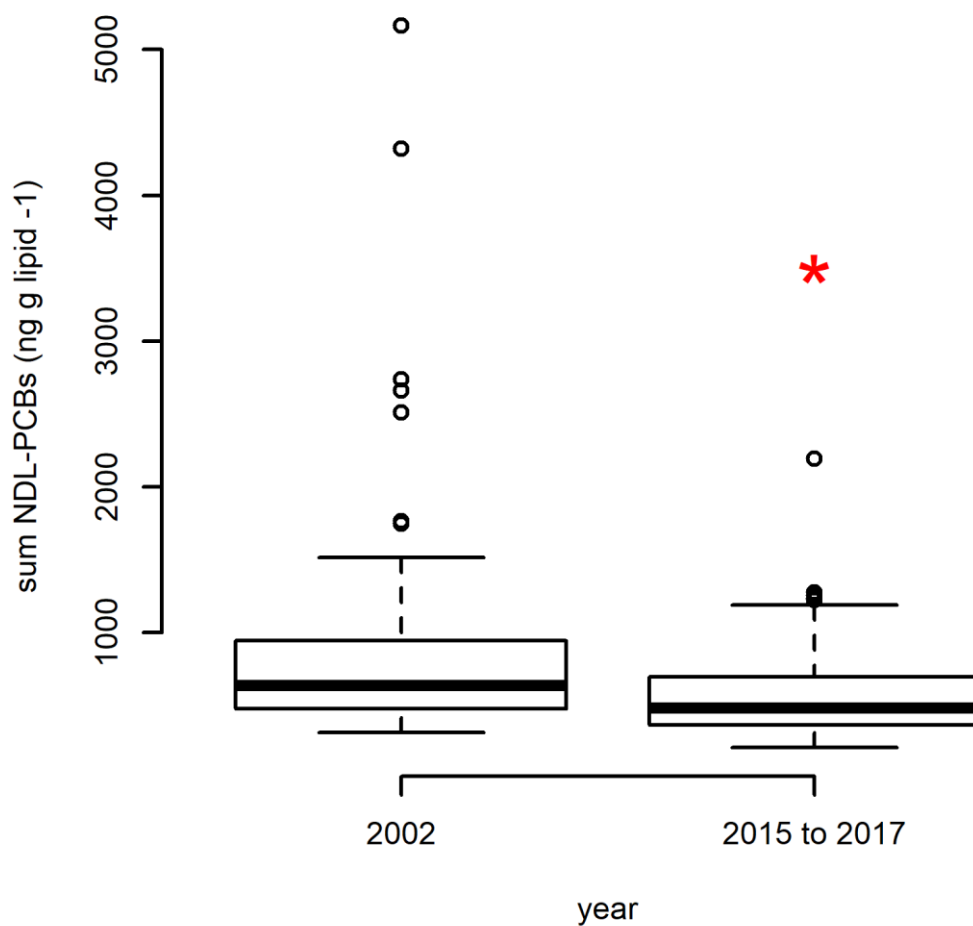
940 Figure 5. Median Σ DDX (DDT, DDD and DDE) concentrations in blubber from
941 weaned grey seal pups from 2002 (n = 60) and 2015-2017 (n = 82) on the Isle of May,
942 with upper and lower quartiles, 1.5 x interquartile range and outliers.

943

944 Figure 6. Median OCP concentrations in blubber from weaned grey seal pups from
945 2002 (n = 60) and 2015-2017 (n = 82) on the Isle of May, with upper and lower
946 quartiles, 1.5 x interquartile range and outliers. Red * shows when levels were
947 significantly lower (GLM, $p < 0.05$). Note differences in y axis ranges to allow
948 visualisation of data range for each OCP.

949

950 Figure 1

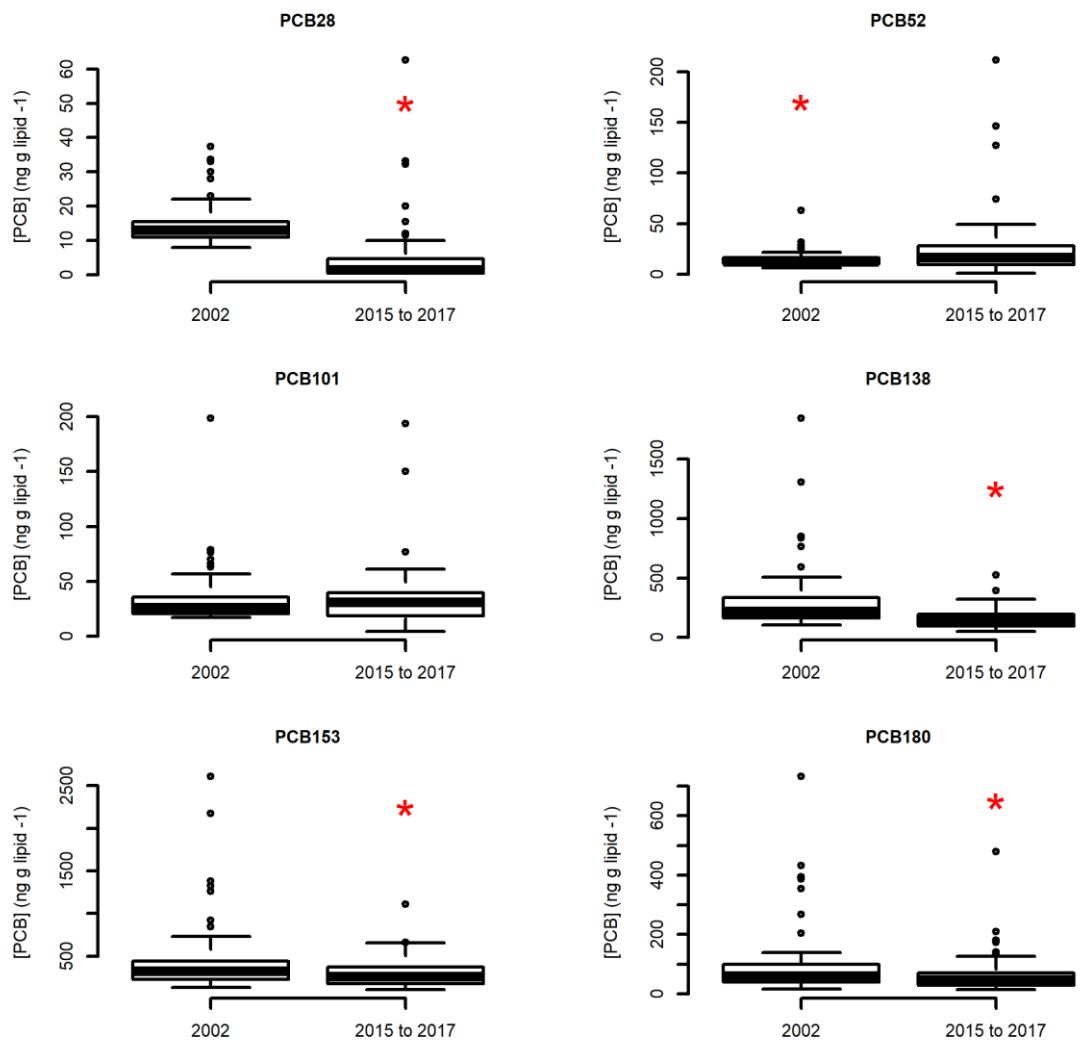


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954 Figure 2

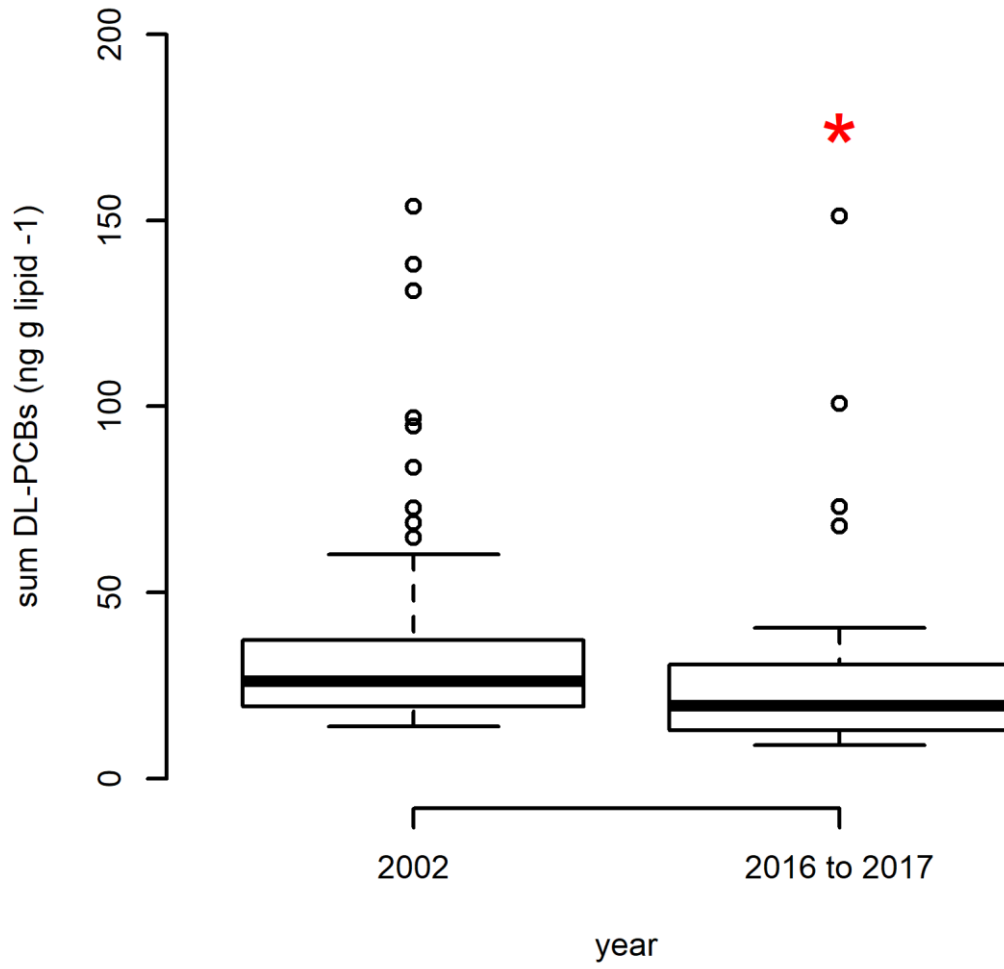


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958 Figure 3

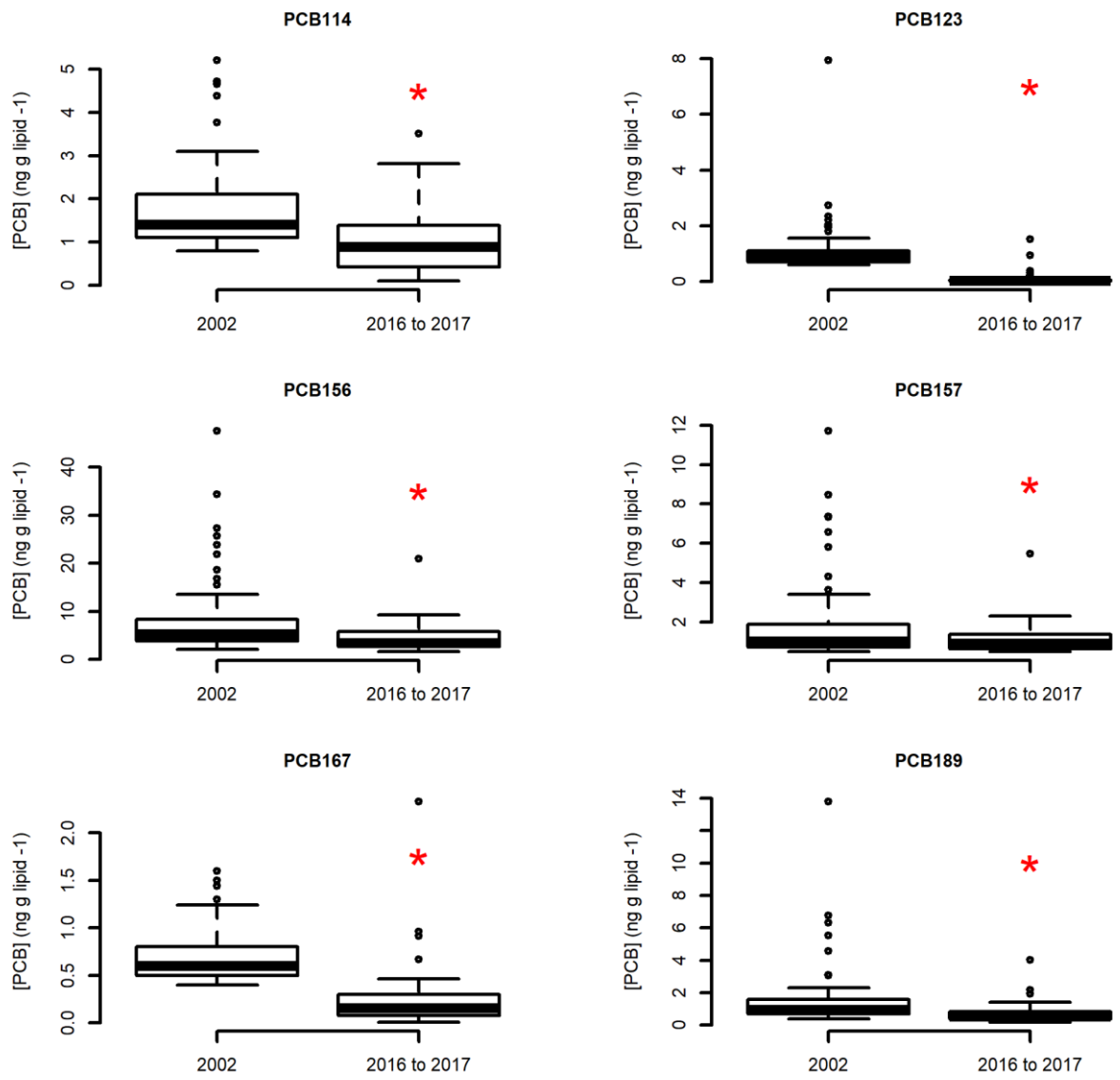


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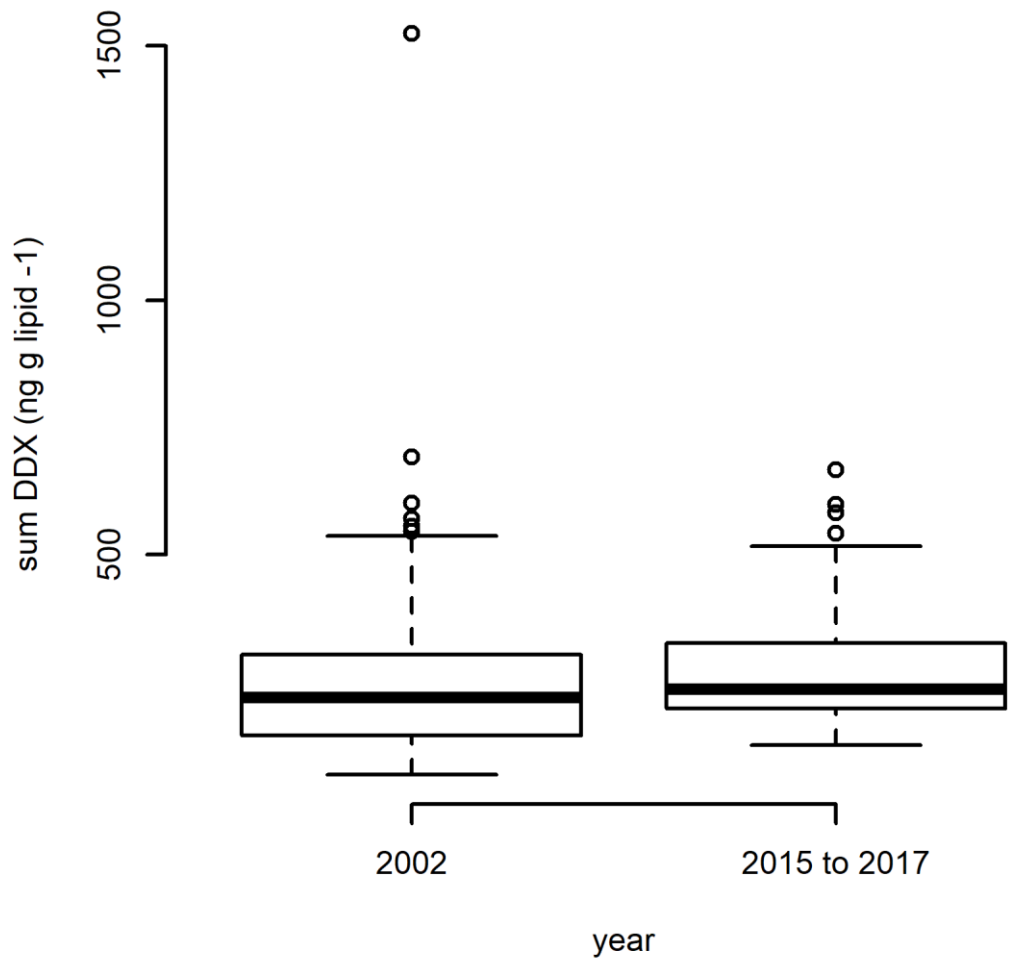
962 Figure 4



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