Environmental Pollution 218 (2016) 651–663

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

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

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ARTICLE INFO

Article history:
Received 22 March 2016
Received in revised form
22 July 2016
Accepted 22 July 2016
Available online 5 August 2016

Keywords:
Pinniped
Marine mammal
Tissue-correlation
Predictive-equations
Fasting
Partitioning coefficients

ABSTRACT

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

1. Introduction

Persistent organic pollutants (POPs) are lipophilic environmental contaminants that are pervasive in marine food webs and bioaccumulate in organisms, which presents particular concern for long-lived, upper trophic level marine mammals. Although close proximity to POP sources can result in higher POP concentrations (Frouin et al., 2011), even foraging strategies that place animals far from contaminant sources do not insulate them from POP exposure (Peterson et al., 2015). Elevated concentrations of POPs are associated with endocrine, immune, and reproductive effects in marine and terrestrial wildlife (Tanabe, 2002; Debier et al., 2005; Desforges et al., 2016). Among marine mammals, pinnipeds and odontocetes

☆ This paper has been recommended for acceptance by Maria Cristina Fossi.
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http://dx.doi.org/10.1016/j.envpol.2016.07.052
0269-7491/

are vulnerable to biomagnification of POPs due to their high trophic position, and therefore are often the target species for POP biomonitoring efforts (Weijs et al., 2010; Yordy et al., 2010b; Lopez et al., 2012).

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

Northern elephant seals (*Mirounga angustirostris*) are upper trophic level predators that bioaccumulate POPs as they forage in the northeastern Pacific Ocean (Peterson et al., 2015). The life history of northern elephant seals includes two foraging migrations per year interspersed with fasting periods for breeding and molting on land, which makes them relatively accessible for study among marine mammals (Robinson et al., 2012). Additionally, foraging and fasting life-history phases create dramatically different seasonal body conditions. For example, during the molting fast females spend roughly 4–6 weeks fasting and lose approximately 25% of their mass, of which 41% comes from fat stores (Worthy et al., 1992). Females then go to sea and forage for approximately 7 months, during which time they undergo a 95% mean mass gain (Robinson et al., 2012). At the end of their foraging trip, females return to land and undergo a breeding fast, lasting approximately 4–6 weeks, which occurs as they give birth to a pup and lactate, and results in loss of approximately 40% of their body mass and 57% of their fat stores (Costa et al., 1986; Crocker et al., 2001). After breeding, females once again return to sea for a shorter 2–3 month foraging trip, which ends when they return to land for molting (Robinson et al., 2012). Males undergo a similar proportional loss of body mass (34–41%) and fat stores (54–59%) while on shore for fasting (Deutsch et al., 1990; Crocker et al., 2012). The male breeding fast is longer than females, lasting approximately 2–3 months, while the male molting fast is approximately 4–6 weeks in duration (Le Boeuf et al., 2000). The two male foraging trips at sea each year are similar to each other in duration, lasting approximately 4 months each (Le Boeuf et al., 2000). Because body condition is an important determinant of POP concentrations in blood and blubber (Peterson et al., 2014), effective toxicological risk assessment relies upon understanding POP concentrations at extremes in body condition.

The naturally occurring extremes of body condition of northern elephant seals present an opportunity to examine how body condition impacts the relationships of contaminant concentrations among tissues. We examined commonly studied tissues with higher (serum, inner blubber) and lower (outer blubber) metabolic activity to describe relationships and determine predictive equations among tissues for POPs in northern elephant seals. Our primary focus was to investigate relationships for polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT) and metabolites of DDT, chlordanes (CHLs), and polybrominated diphenyl ethers (PBDEs), although we also included hexachlorobenzene (HCB), hexachlorocyclohexane (α-HCH and β-HCH), and the naturally produced 6-MeO-BDE 47. Specifically, our objectives were to 1) assess serum POP concentrations as a predictor of inner and outer blubber POP concentrations, and inner blubber POP concentrations as a predictor of outer blubber POP concentrations at two body condition extremes, 2) compare serum to blubber relationships between males and females, and 3) use partitioning coefficients to compare blubber/serum POPs and inner blubber/outer blubber POPs relationships between females and males at two naturally occurring extremes of elephant seal body condition.

2. Methods

2.1. Animal sampling

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

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

2.2. Laboratory analysis

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

Extraction, clean-up, and concentration measurement methods for blubber and serum followed protocols described in Vanden Berghe et al. (2012) and Peterson et al. (2015). In brief, for

blubber analyses, the skin layer and hair (Schwarz et al., 2015), was removed from the outer portion of the biopsy core, and the remaining blubber layer was cut into inner and outer segments of approximately equal mass. Inner and outer blubber segments were analyzed separately due to differences in metabolic activity and stratification in fatty acid profiles among layers (Strandberg et al., 2008; Fowler et al., 2014). Serum samples were split for separate determination of target contaminants and lipids. Four lipid classes in serum (total cholesterol, phospholipids, triacylglycerides, and non-esterified fatty acids) were determined with enzyme kits from Diasys Diagnostic Systems (Holzheim, Germany) and Wako Chemicals (Neuss, Germany), with the concentrations of each lipid class calculated on the basis of standard equivalents. Total lipid concentrations were calculated as the sum of the four lipid classes (Debier et al., 2006; Vanden Berghe et al., 2012). All POP concentrations in serum were lipid-normalized before statistical analyses.

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

2.3. Quality control

For quality control (QC), we randomly analyzed procedural blanks, solvent blanks, and standards throughout the extraction process. Recoveries for individual PCB and PBDE congeners ranged between 75 and 104% (RSD < 12%). For each analyte, the mean procedural blank value was used for subtraction to determine final analyte concentrations. After blank subtraction, the limit of quantification (LOQ) was set at 3 × SD of the procedural blank. For analytes that were not detected in procedural blanks, LOQs were calculated for a ratio S/N (signal to noise) equal to 10. A standard reference material SRM 1945 (PCBs, OCPs, and PBDEs in whale blubber) was used to test the accuracy of the method. Measured values did not deviate more than 15% from the certified values.

2.4. Statistical analysis

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

In addition, we calculated partitioning coefficients for outer blubber/serum, inner blubber/serum, and outer blubber/inner blubber for each contaminant in female and male elephant seals. Partitioning coefficients were calculated for each individual as the ratio between concentrations in the two tissues. We used mixed effects models to compare partitioning coefficients by sampling period (late molting, early breeding), sex, and a sex × sampling period interaction, with individual as a random effect. If the interaction was not significant, we removed it and reran the model. We conducted post-hoc pairwise comparisons on least-squares means with a Tukey adjustment on p-values. For statistical significance, α was set at $p = 0.05$.

3. Results

Serum, inner blubber, and outer blubber samples from all elephant seals had detectable concentrations of ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs. The concentrations of POPs used in this study for northern elephant seal serum, inner blubber, and outer blubber have been reported previously (Peterson et al., 2015), although a summary of medians and ranges of POP concentrations can be found in Table 1. The specific equations to predict POP concentrations in one tissue from POPs concentrations in another tissue for all compounds are found in Table 2 and specific statistical results are found in Table 3.

3.1. Serum and inner blubber

Late molting seals did not have a significant interaction between sex × serum concentrations of ΣDDTs, ΣPCBs, ΣCHLs, or ΣPBDEs on the concentrations of these POP classes in inner blubber ($F \leq 1.80$, $p \geq 0.18$). Similarly, early breeding seals did not have a significant interaction between serum POP concentrations × sex on the POP concentrations in inner blubber, except for PBDEs ($F_{1,35} = 6.45$, $p = 0.015$). Therefore, we removed the interaction from all models except for early breeding PBDEs, where we analyzed males and females separately.

Serum POP concentrations were positively related to inner

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)

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

Sex	Equation: ln (tissue Y) = slope × ln (tissue X) + intercept	R^2	95% CI slope	95% CI intercept
Late molting (N = 14 males, 24 females)				
Inner blubber:serum				
F + M	$\Sigma DDT_{In} = 1.107 \times \Sigma DDT_{Ser} - 0.467$	*0.86*	(0.957, 1.258)	(−1.561, 0.627)
F + M	$\Sigma PCB_{In} = 0.921 \times \Sigma PCB_{Ser} + 1.014$	*0.71*	(0.721, 1.121)	(−0.312, 2.341)
F + M	$\Sigma CHL_{In} = 0.971 \times \Sigma CHL_{Ser} + 0.739$	*0.78*	(0.795, 1.147)	(−0.221, 1.699)
F + M	$\Sigma PBDE_{In} = 0.888 \times \Sigma PBDE_{Ser} + 0.883$	*0.78*	(0.730, 1.045)	(0.416, 1.349)
F	MeOBDE: *Not significant*	–	–	–
M	MeOBDE: *Low detectability*	–	–	–
F + M	$\alpha\text{-}HCH_{In} = 0.340 \times \alpha\text{-}HCH_{Ser} + 1.539$	*0.19*	(0.105, 0.574)	(1.271, 1.807)
F + M	$\beta\text{-}HCH_{In} = 0.820 \times \beta\text{-}HCH_{Ser} + 1.197$	*0.86*	(0.707, 0.932)	(0.845, 1.549)
F	$HCB_{In} = 0.282 \times HCB_{Ser} + 2.071$	*0.16*	(0.145, 0.419)	(1.586, 2.557)
M	$HCB_{In} = 0.282 \times HCB_{Ser} + 2.284$	*0.58*	(0.145, 0.419)	(1.727, 2.841)
Outer blubber:serum				
F	$\Sigma DDT_{Out} = 0.782 \times \Sigma DDT_{Ser} + 1.761$	*0.61*	(0.504, 1.061)	(−0.219, 3.742)
M	$\Sigma DDT_{Out} = 1.212 \times \Sigma DDT_{Ser} - 1.349$	*0.87*	(0.924, 1.501)	(−3.531, 0.833)
F	$\Sigma PCB_{Out} = 0.370 \times \Sigma PCB_{Ser} + 4.269$	*0.23*	(0.075, 0.665)	(2.353, 6.184)
M	$\Sigma PCB_{Out} = 1.342 \times \Sigma PCB_{Ser} - 1.973$	*0.89*	(1.046, 1.637)	(−3.995, 0.050)
F	$\Sigma CHL_{Out} = 0.436 \times \Sigma CHL_{Ser} + 3.321$	*0.35*	(0.172, 0.701)	(1.929, 4.713)
M	$\Sigma CHL_{Out} = 1.423 \times \Sigma CHL_{Ser} - 2.055$	*0.84*	(1.031, 1.816)	(−4.308, 0.197)
F + M	$\Sigma PBDE_{Out} = 0.814 \times \Sigma PBDE_{Ser} + 0.980$	*0.60*	(0.587, 1.040)	(0.310, 1.650)
F	MeOBDE: *Not significant*	–	–	–
M	MeOBDE: *Low detectability*	–	–	–
F	α-HCH: *Not significant*	–	–	–
M	$\alpha\text{-}HCH_{Out} = 0.732 \times \alpha\text{-}HCH_{Ser} + 0.897$	*0.53*	(0.302, 1.163)	(0.321, 1.473)
F	$\beta\text{-}HCH_{Out} = 0.640 \times \beta\text{-}HCH_{Ser} + 1.682$	*0.78*	(0.490, 0.789)	(1.245, 2.119)
M	$\beta\text{-}HCH_{Out} = 0.939 \times \beta\text{-}HCH_{Ser} + 0.810$	*0.85*	(0.687, 1.190)	(−0.067, 1.687)
F	HCB: *Not significant*	–	–	–
M	$HCB_{Out} = 0.431 \times HCB_{Ser} + 1.618$	*0.53*	(0.178, 0.684)	(0.594, 2.642)
Outer blubber:inner blubber				
F + M	$\Sigma DDT_{Out} = 0.932 \times \Sigma DDT_{In} + 0.430$	*0.94*	(0.853, 1.011)	(−0.174, 1.034)
F	$\Sigma PCB_{Out} = 0.499 \times \Sigma PCB_{In} + 3.184$	*0.44*	(0.251, 0.747)	(1.450, 4.918)
M	$\Sigma PCB_{Out} = 1.124 \times \Sigma PCB_{In} - 1.019$	*0.94*	(0.942, 1.306)	(−2.351, 0.312)
F	$\Sigma CHL_{Out} = 0.521 \times \Sigma CHL_{In} + 2.563$	*0.66*	(0.355, 0.687)	(1.589, 3.537)
M	$\Sigma CHL_{Out} = 1.098 \times \Sigma CHL_{In} - 0.782$	*0.96*	(0.957, 1.238)	(−1.665, 0.101)
F	$\Sigma PBDE_{Out} = 0.906 \times \Sigma PBDE_{In} + 0.150$	*0.65*	(0.753, 1.060)	(−0.377, 0.677)
M	$\Sigma PBDE_{Out} = 0.906 \times \Sigma PBDE_{In} + 0.303$	*0.93*	(0.753, 1.060)	(−0.266, 0.872)
F + M	$MeOBDE_{Out} = 0.793 \times MeOBDE_{In} - 0.083$	*0.84*	(0.675, 0.912)	(−0.183, 0.018)
F	$\alpha\text{-}HCH_{Out} = 0.362 \times \alpha\text{-}HCH_{In} + 1.005$	*0.23*	(0.070, 0.653)	(0.444, 1.566)
M	$\alpha\text{-}HCH_{Out} = 1.144 \times \alpha\text{-}HCH_{In} - 0.340$	*0.91*	(0.912, 1.376)	(−0.788, 0.108)
F	$\beta\text{-}HCH_{Out} = 0.885 \times \beta\text{-}HCH_{In} + 0.393$	*0.97*	(0.821, 0.950)	(0.163, 0.624)
M	$\beta\text{-}HCH_{Out} = 1.028 \times \beta\text{-}HCH_{In} - 0.127$	*0.96*	(0.900, 1.156)	(−0.651, 0.396)
F	HCB: *Not significant*	–	–	–
M	$HCB_{Out} = 1.034 \times \Sigma HCB_{In} - 0.181$	*0.61*	(0.516, 1.552)	(−1.955, 1.593)
Early breeding (N = 15 males, 24 females)				
Inner blubber:serum				
F + M	$\Sigma DDT_{In} = 0.911 \times \Sigma DDT_{Ser} + 1.110$	*0.91*	(0.814, 1.008)	(0.448, 1.772)
F + M	$\Sigma PCB_{In} = 0.774 \times \Sigma PCB_{Ser} + 2.082$	*0.87*	(0.674, 0.873)	(1.471, 2.692)
F + M	$\Sigma CHL_{In} = 0.851 \times \Sigma CHL_{Ser} + 1.451$	*0.85*	(0.732, 0.970)	(0.854, 2.048)
F	$\Sigma PBDE_{In} = 0.615 \times \Sigma PBDE_{Ser} + 1.684$	*0.53*	(0.357, 0.872)	(1.138, 2.229)
M	$\Sigma PBDE_{In} = 0.947 \times \Sigma PBDE_{Ser} + 0.913$	*0.97*	(0.848, 1.047)	(0.608, 1.218)
F	$MeOBDE_{In} = 0.440 \times MeOBDE_{Ser} + 1.181$	*0.39*	(0.269, 0.611)	(1.035, 1.327)
M	$MeOBDE_{In} = 0.440 \times MeOBDE_{Ser} + 0.873$	*0.44*	(0.269, 0.611)	(0.661, 1.084)
F	$\alpha\text{-}HCH_{In} = 0.403 \times \alpha\text{-}HCH_{Ser} + 1.195$	*0.32*	(0.141, 0.664)	(0.958, 1.432)
M	α-HCH: *Not significant*	–	–	–
F + M	$\beta\text{-}HCH_{In} = 0.984 \times \beta\text{-}HCH_{Ser} + 0.623$	*0.89*	(0.868, 1.100)	(0.285, 0.962)
F + M	HCB: *Not significant*			
Outer blubber:serum				
F	$\Sigma DDT_{Out} = 0.683 \times \Sigma DDT_{Ser} + 2.847$	*0.75*	(0.509, 0.857)	(1.725, 3.969)
M	$\Sigma DDT_{Out} = 0.986 \times \Sigma DDT_{Ser} + 0.677$	*0.94*	(0.831, 1.141)	(−0.460, 1.814)
F	$\Sigma PCB_{Out} = 0.389 \times \Sigma PCB_{Ser} + 4.410$	*0.25*	(0.093, 0.685)	(2.709, 6.111)
M	$\Sigma PCB_{Out} = 0.857 \times \Sigma PCB_{Ser} + 1.467$	*0.88*	(0.669, 1.045)	(0.205, 2.730)
F	$\Sigma CHL_{Out} = 0.287 \times \Sigma CHL_{Ser} + 4.194$	*0.25*	(0.066, 0.509)	(3.151, 5.237)
M	$\Sigma CHL_{Out} = 0.777 \times \Sigma CHL_{Ser} + 1.904$	*0.79*	(0.540, 1.014)	(0.600, 3.207)
F	$\Sigma PBDE_{Out} = 0.532 \times \Sigma PBDE_{Ser} + 2.044$	*0.53*	(0.310, 0.754)	(1.574, 2.513)
M	$\Sigma PBDE_{Out} = 0.946 \times \Sigma PBDE_{Ser} + 0.941$	*0.97*	(0.840, 1.053)	(0.615, 1.268)
F	$MeOBDE_{Out} = 0.350 \times MeOBDE_{Ser} + 0.891$	*0.28*	(0.183, 0.518)	(0.748, 1.034)
M	$MeOBDE_{Out} = 0.350 \times MeOBDE_{Ser} + 0.518$	*0.35*	(0.183, 0.518)	(0.311, 0.724)
F	$\alpha\text{-}HCH_{Out} = 0.270 \times \alpha\text{-}HCH_{Ser} + 1.295$	*0.18*	(0.059, 0.480)	(1.094, 1.496)
M	$\alpha\text{-}HCH_{Out} = 0.270 \times \alpha\text{-}HCH_{Ser} + 1.569$	*0.13*	(0.059, 0.480)	(1.410, 1.728)
F + M	$\beta\text{-}HCH_{Out} = 0.905 \times \beta\text{-}HCH_{Ser} + 0.998$	*0.90*	(0.803, 1.008)	(0.699, 1.298)
F + M	HCB: *Not significant*	–	–	–

Table 2 (*continued*)

Sex	Equation: ln (tissue Y) = slope × ln (tissue X) + intercept	R^2	95% CI slope	95% CI intercept
Outer blubber:inner blubber				
F	$\Sigma DDT_{Out} = 0.603 \times \Sigma DDT_{In} + 3.027$	*0.55*	(0.360, 0.847)	(1.320, 4.735)
M	$\Sigma DDT_{Out} = 0.967 \times \Sigma DDT_{In} + 0.393$	*0.98*	(0.893, 1.042)	(−0.184, 0.970)
F	$\Sigma PCB_{Out} = 0.475 \times \Sigma PCB_{In} + 3.535$	*0.41*	(0.222, 0.727)	(1.877, 5.193)
M	$\Sigma PCB_{Out} = 0.920 \times \Sigma PCB_{In} + 0.565$	*0.98*	(0.835, 1.004)	(−0.047, 1.178)
F	$\Sigma CHL_{Out} = 0.235 \times \Sigma CHL_{In} + 4.265$	*0.19*	(0.017, 0.453)	(3.081, 5.450)
M	$\Sigma CHL_{Out} = 0.951 \times \Sigma CHL_{In} + 0.317$	*0.91*	(0.767, 1.135)	(−0.815, 1.449)
F	$\Sigma PBDE_{Out} = 0.624 \times \Sigma PBDE_{In} + 1.299$	*0.52*	(0.360, 0.888)	(0.512, 2.085)
M	$\Sigma PBDE_{Out} = 0.988 \times \Sigma PBDE_{In} + 0.070$	*0.97*	(0.892, 1.085)	(−0.296, 0.435)
F + M	$MeOBDE_{Out} = 0.889 \times MeOBDE_{In} - 0.183$	*0.89*	(0.787, 0.991)	(−0.289, −0.077)
F + M	$\alpha\text{-}HCH_{Out} = 0.782 \times \alpha\text{-}HCH_{In} + 0.340$	*0.61*	(0.575, 0.988)	(0.006, 0.673)
F + M	$\beta\text{-}HCH_{Out} = 0.865 \times \beta\text{-}HCH_{In} + 0.618$	*0.89*	(0.264, 0.972)	(0.264, 0.972)
F + M	$HCB_{Out} = 0.746 \times HCB_{In} + 0.916$	*0.51*	(0.500, 0.992)	(0.217, 1.615)

Table 3
Results from linear models examining the relationships between POP concentrations in pairs of tissues (inner blubber:serum, outer blubber:serum, and 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-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: $F_{1,35} = 156.92$, $p < 0.001$	ΣDDTs: $F_{1,36} = 227.55$, $p < 0.001$
	Sex: $F_{1,35} < 0.01$, $p = 0.95$	Sex: $F_{1,36} = 0.75$, $p = 0.39$
ΣPCBs	ΣPCBs: $F_{1,35} = 63.83$, $p < 0.001$	ΣPCBs: $F_{1,36} = 133.32$, $p < 0.001$
	Sex: $F_{1,35} < 0.01$, $p = 0.96$	Sex: $F_{1,36} = 3.35$, $p = 0.08$
ΣCHLs	ΣCHLs: $F_{1,35} = 76.84$, $p < 0.001$	ΣCHLs: $F_{1,36} = 49.38$, $p < 0.001$
	Sex: $F_{1,35} = 1.27$, $p = 0.27$	Sex: $F_{1,36} = 2.13$, $p = 0.15$
ΣPBDEs	ΣPBDEs: $F_{1,35} = 116.72$, $p < 0.001$	ΣPBDEs (female): $F_{1,22} = 24.55$, $p < 0.001$
	Sex: $F_{1,35} = 0.01$, $p = 0.91$	ΣPBDEs (male): $F_{1,13} = 422.54$, $p < 0.001$
αHCH	αHCH: $F_{1,35} = 10.96$, $p = 0.002$	αHCH (female): $F_{1,22} = 10.2$, $p = 0.004$
	Sex: $F_{1,35} = 2.09$, $p = 0.16$	αHCH (male): $F_{1,13} = 0.04$, $p = 0.84$
βHCH	βHCH: $F_{1,35} = 111.79$, $p < 0.001$	βHCH: $F_{1,36} = 153.3$, $p < 0.001$
	Sex: $F_{1,35} = 2.81$, $p = 0.10$	Sex: $F_{1,36} = 6.54$, $p = 0.015$
HCB	HCB: $F_{1,35} = 17.57$, $p < 0.001$	HCB: $F_{1,36} = 0.02$, $p = 0.89$
	Sex: $F_{1,35} = 9.76$, $p = 0.003$	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: $F_{1,36} = 6.54$, $p = 0.015$
		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 (female): $F_{1,22} = 66.43$, $p < 0.001$
	ΣDDTs (male): $F_{1,12} = 83.77$, $p < 0.001$	ΣDDTs (male): $F_{1,13} = 188.90$, $p < 0.001$
ΣPCBs	ΣPCBs (female): $F_{1,22} = 6.77$, $p = 0.016$	ΣPCBs (female): $F_{1,22} = 7.44$, $p = 0.012$
	ΣPCBs (male): $F_{1,12} = 97.86$, $p < 0.001$	ΣPCBs (male): $F_{1,13} = 97.18$, $p < 0.001$
ΣCHLs	ΣCHLs (female): $F_{1,22} = 11.71$, $p = 0.002$	ΣCHLs (female): $F_{1,22} = 7.22$, $p = 0.013$
	ΣCHLs (male): $F_{1,12} = 62.53$, $p < 0.001$	ΣCHLs (male): $F_{1,13} = 50.00$, $p < 0.001$
ΣPBDEs	ΣPBDEs: $F_{1,35} = 45.13$, $p < 0.001$	ΣPBDEs (female): $F_{1,22} = 24.80$, $p < 0.001$
	Sex: $F_{1,35} = 2.37$, $p = 0.13$	ΣPBDEs (male): $F_{1,13} = 368.10$, $p < 0.001$
αHCH	αHCH (female): $F_{1,22} = 1.13$, $p = 0.30$	αHCH: $F_{1,36} = 5.78$, $p = 0.007$
	αHCH (male): $F_{1,12} = 13.74$, $p = 0.003$	Sex: $F_{1,36} = 9.49$, $p = 0.004$
βHCH	βHCH (female): $F_{1,22} = 78.55$, $p < 0.001$	βHCH: $F_{1,36} = 166.13$, $p < 0.001$
	βHCH (male): $F_{1,12} = 65.98$, $p < 0.001$	Sex: $F_{1,36} = 2.09$, $p = 0.16$
HCB	HCB (female): $F_{1,22} = 3.34$, $p = 0.08$	HCB: $F_{1,34} = 3.05$, $p = 0.06$
	HCB (male): $F_{1,12} = 13.79$, $p = 0.003$	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: $F_{1,36} = 20.61$, $p \leq 0.001$
		Sex: $F_{1,36} = 10.04$, $p = 0.003$
Outer blubber	Inner blubber	Inner blubber
ΣDDTs	ΣDDTs: $F_{1,35} = 409.70$, $p < 0.001$	ΣDDTs (female): $F_{1,22} = 26.36$, $p < 0.001$
	Sex: $F_{1,35} = 0.64$, $p = 0.43$	ΣDDTs (male): $F_{1,13} = 791.90$, $p < 0.001$
ΣPCBs	ΣPCBs (female): $F_{1,22} = 17.40$, $p < 0.001$	ΣPCBs (female): $F_{1,22} = 15.15$, $p < 0.001$
	ΣPCBs (male): $F_{1,12} = 181.40$, $p < 0.001$	ΣPCBs (male): $F_{1,13} = 552.00$, $p < 0.001$
ΣCHLs	ΣCHLs (female): $F_{1,22} = 42.37$, $p < 0.001$	ΣCHLs (female): $F_{1,22} = 5.02$, $p = 0.036$
	ΣCHLs (male): $F_{1,12} = 290.40$, $p < 0.001$	ΣCHLs (male): $F_{1,13} = 124.82$, $p < 0.001$
ΣPBDEs	ΣPBDEs: $F_{1,35} = 143.80$, $p < 0.001$	ΣPBDEs (female): $F_{1,22} = 24.05$, $p < 0.001$
	Sex: $F_{1,35} = 4.26$, $p < 0.047$	ΣPBDEs (male): $F_{1,13} = 489.40$, $p < 0.001$
αHCH	αHCH (female): $F_{1,22} = 6.63$, $p = 0.017$	αHCH: $F_{1,36} = 49.34$, $p < 0.001$
	αHCH (male): $F_{1,12} = 115.60$, $p < 0.001$	Sex: $F_{1,36} = 0.60$, $p = 0.45$
βHCH	βHCH (female): $F_{1,22} = 807.20$, $p < 0.001$	βHCH: $F_{1,36} = 92.61$, $p < 0.001$
	βHCH (male): $F_{1,12} = 307.00$, $p < 0.001$	Sex: $F_{1,36} = 2.10$, $p = 0.16$
HCB	HCB (female): $F_{1,22} = 4.16$, $p = 0.054$	HCB: $F_{1,36} = 27.18$, $p < 0.001$
	HCB (male): $F_{1,12} = 18.90$, $p < 0.001$	Sex: $F_{1,36} = 0.52$, $p = 0.47$
6-MeO-BDE 47	6-MeO-BDE 47: $F_{1,35} = 140.92$, $p < 0.001$	6-MeO-BDE 47: $F_{1,36} = 224.36$, $p < 0.001$
	Sex: $F_{1,35} = 1.04$, $p = 0.31$	Sex: $F_{1,36} = 3.30$, $p = 0.08$

[a] Model 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 if males and females were analyzed separately.

blubber POP concentrations for all major POP classes (ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs) both late in the molting fast (Fig. 1) and early in the breeding fast (Fig. 2), while accounting for any potential effect of sex. Early in the breeding fast, concentrations of ΣPBDEs in serum were positively related to concentrations of ΣPBDEs in inner blubber for females and males. For ΣDDTs, ΣPCBs, and ΣCHLs at both sampling periods, and ΣPBDEs at late molting, all relationships between serum and inner blubber had R^2 values > 0.7, and relationships for all POP classes were stronger at early breeding than at late molting (Table 2). In addition, the highest R^2 values at each life history phase were found for ΣDDTs.

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

In early breeding, we did not detect a significant serum × sex interaction for 6-MeO-BDE 47 ($F_{1,35} = 2.20$, $p = 0.15$) or β-HCH ($F_{1,35} = 0.11$, $p = 0.75$). For α-HCH, we detected a marginally significant interaction ($F_{1,35} = 3.05$, $p = 0.09$) and analyzed female and male seals separately. Serum concentrations of 6-MeO-BDE 47 and β-HCH were positively related to inner blubber concentrations, while accounting for any potential effect of sex. For α-HCH we detected a significant relationship between serum and inner blubber concentrations for females, but not males. While the relationships between serum concentrations and inner blubber concentrations of β-HCH at late molting and early breeding were strong ($R^2 \geq 0.58$), we observed weaker relationships for α-HCH ($R^2 \leq 0.38$). In addition, there was no significant relationship between serum and inner blubber for HCB, while accounting for sex.

3.2. Serum and outer blubber

We detected a significant interaction between POP concentrations in serum × sex on POP concentrations in outer blubber for ΣDDTs, ΣPCBs, and ΣCHLs during late molting ($F_{1,34} \geq 5.27$, $p \leq 0.001$) and early breeding ($F_{1,35} \geq 7.48$, $p \leq 0.010$). In addition, we detected a serum × sex interaction in early breeding ΣPBDEs

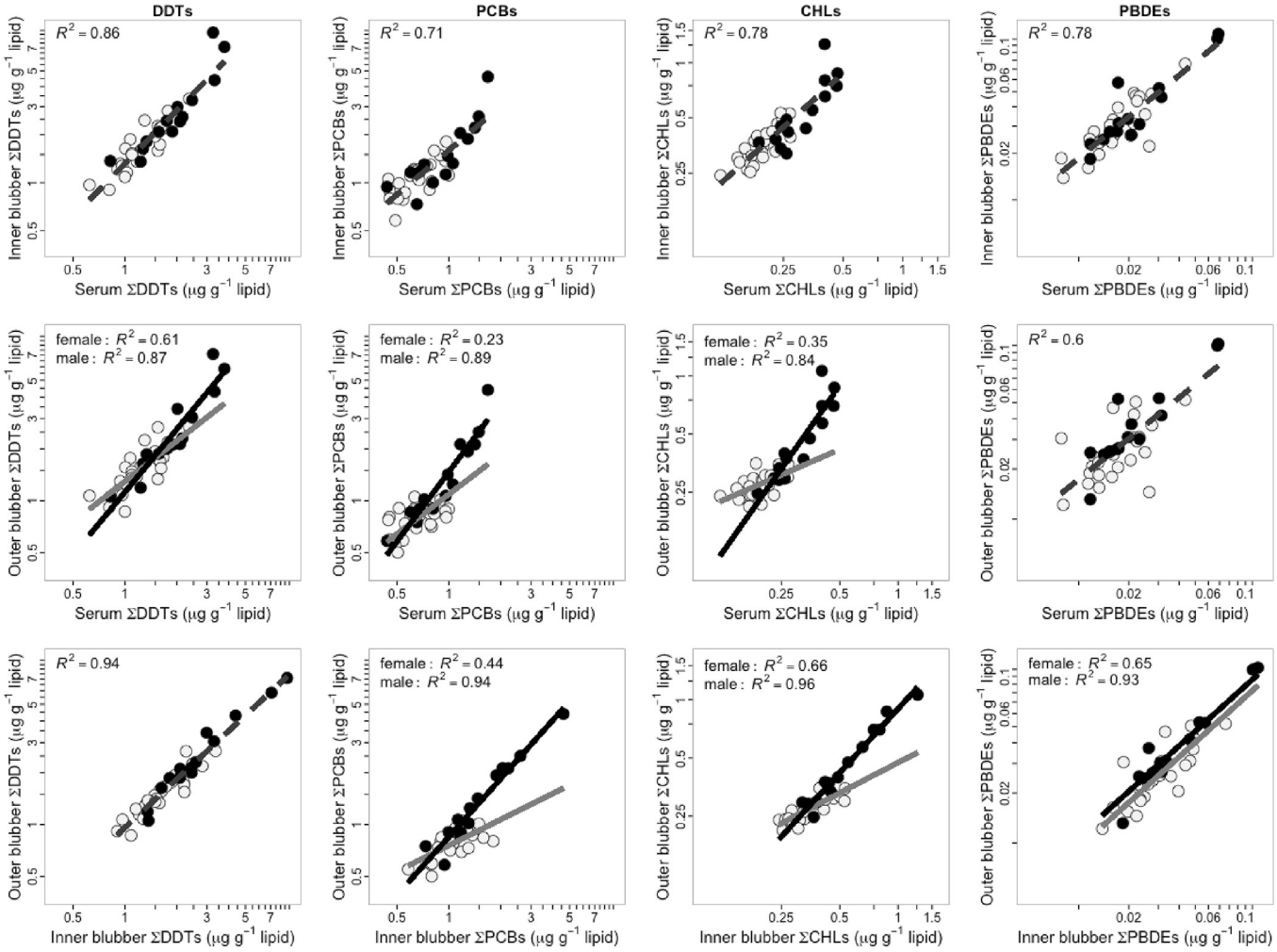

Fig. 1. Relationships between concentrations of ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs in inner blubber and serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels) of adult female and male northern elephant seals late in the molting fast (pre-foraging).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

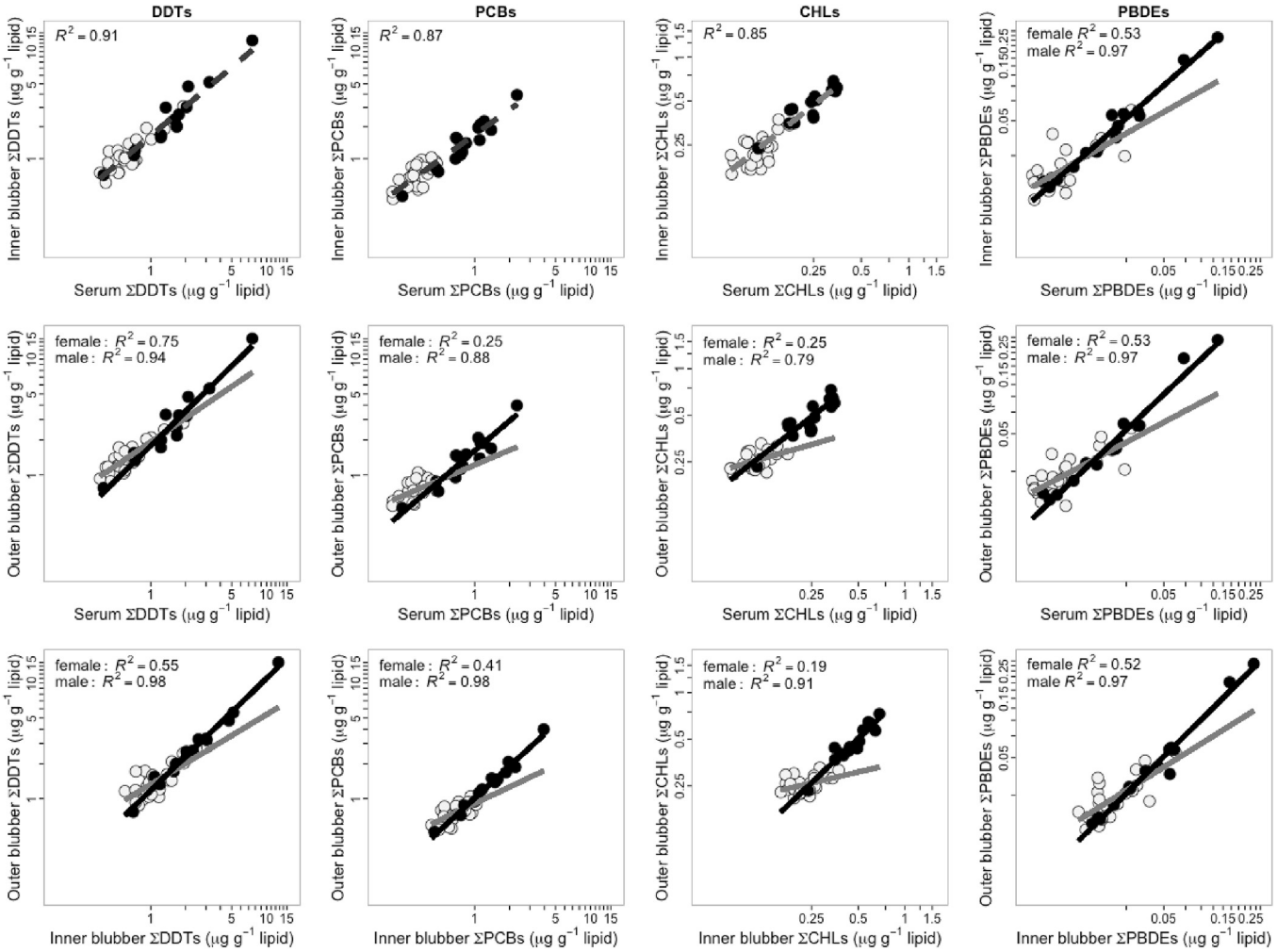

Fig. 2. Relationships between concentrations of ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs in inner blubber and serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels) of adult female and male northern elephant seals early in the breeding fast (post-foraging).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

($F_{1,35} = 12.58$, $p < 0.001$), but not late molting ΣPBDEs ($F_{1,34} = 3.57$, $p = 0.067$). Therefore, we removed the interaction from the model for late molting ΣPBDEs, but for all other POP classes we conducted separate statistical analyses on female and male elephant seals.

For combined male and female seals late in the molting fast, concentrations of ΣPBDEs in serum were positively related to concentrations of ΣPBDEs in outer blubber, when accounting for sex. There was not a significant sex effect for ΣPBDEs, indicating that similar ΣPBDE concentrations in serum corresponded to similar ΣPBDE concentrations in outer blubber for males and females (Fig. 1). Early in breeding, serum ΣPBDE concentrations were positively related to outer blubber concentrations for males and females. In addition, serum ΣDDT, ΣPCB, and ΣCHL concentrations were positively related to outer blubber concentrations during both sampling periods for females and males (Figs. 1–2). Outer blubber:serum relationships were stronger for males ($0.94 \geq R^2 \geq 0.79$) than females ($0.75 \geq R^2 \geq 0.23$) for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs at both sampling periods, but the differences between the sexes were more pronounced for ΣPCBs and ΣCHLs at both periods than for ΣDDTs and ΣPBDEs (Figs. 1–2). For ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs, the slope of the relationship for males was steeper than females, indicating that at higher serum concentrations individual males had proportionately higher outer blubber contaminant concentrations than females (Table 2).

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

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

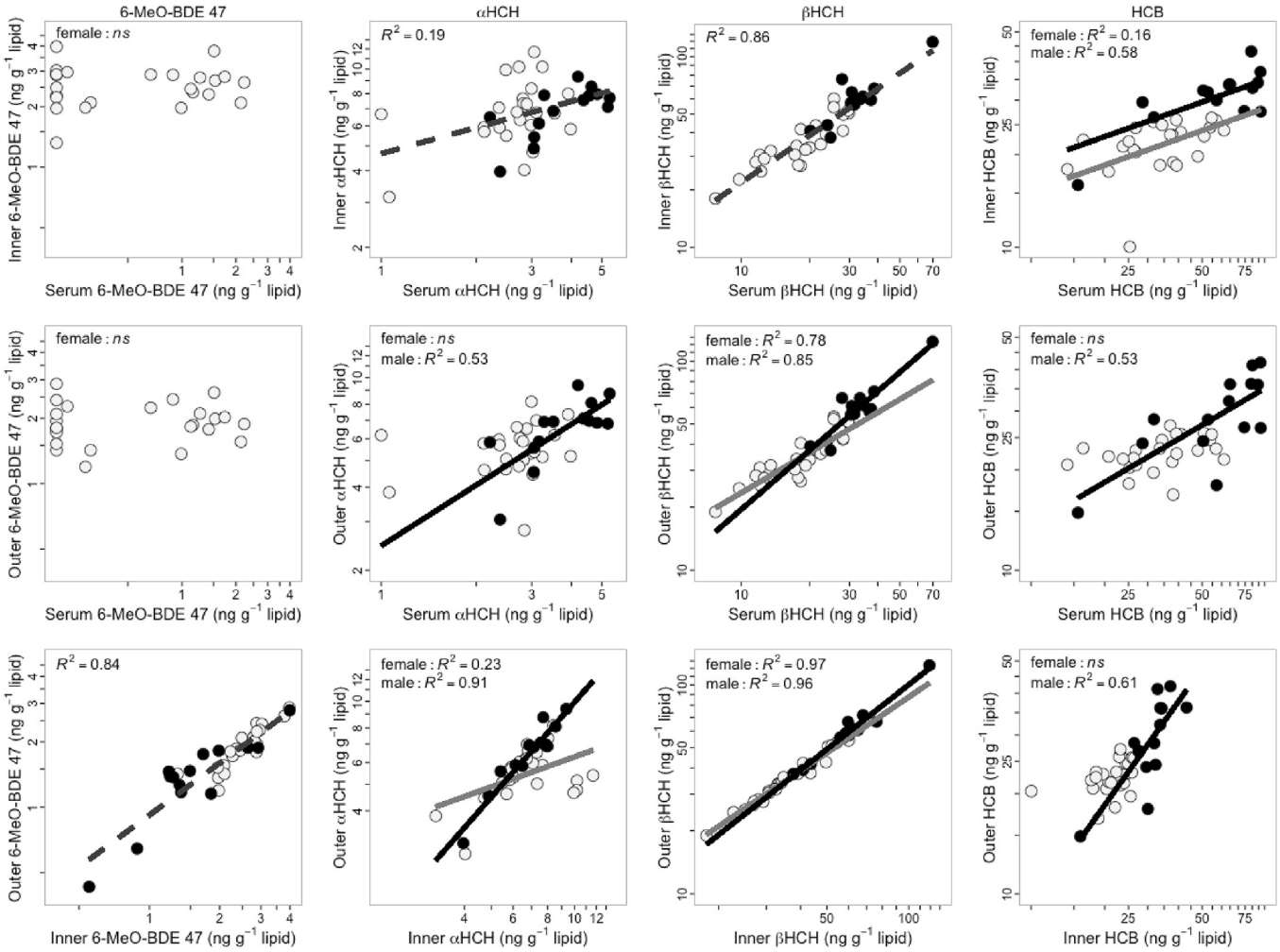

Fig. 3. Relationships between concentrations of 6-MeO-BDE 47, α-HCH, β-HCH, and HCB in inner blubber and serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels) of adult female and male northern elephant seals late in the molting fast (pre-foraging).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

3.3. Inner blubber and outer blubber

We observed a significant interaction between POP concentrations in inner blubber × sex on ΣPCB and ΣCHL concentrations in outer blubber during late molting ($F_{1,34} \geq 18.37$, $p \leq 0.001$) and ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs during early breeding ($F_{1,35} \geq 7.80$, $p \leq 0.008$). In addition, we found no detectable interaction for ΣDDTs and ΣPBDEs during late molting ($F_{1,34} \leq 2.32$, $p \geq 0.14$). Therefore, in the case of late molting, we removed the interaction from the models for ΣDDTs and ΣPBDEs, and for ΣPCBs and ΣCHLs we analyzed females and males separately.

Inner blubber POP concentrations were positively related to outer blubber POP concentrations for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs during both sampling periods, for both females and males (Fig. 2). For late molting seals, concentrations of ΣDDTs and ΣPBDEs in serum were significantly related to concentrations of ΣDDTs and ΣPBDEs in outer blubber, while accounting for sex (Fig. 1). The significant sex effect for ΣPBDEs in outer blubber showed that males and females had the same slope, but males had higher concentrations of ΣPBDEs in outer blubber than females for similar concentrations in inner blubber.

Additionally, the relationships between inner and outer blubber were stronger for males than females for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs at late molting (Fig. 1) and early breeding (Fig. 2). For female concentrations of ΣCHLs in inner blubber and outer blubber, the relationship was stronger in late molting ($R^2 = 0.66$) relative to early breeding ($R^2 = 0.19$), whereas the relationships between concentrations of ΣPCBs and ΣPBDEs in inner blubber and outer blubber were similar between the two life history stages (Figs. 1–2). For ΣDDTs, ΣPCBs, and ΣCHLs from both time periods, and early breeding ΣPBDEs, the slope of the relationship for males was steeper than females, indicating that males with higher inner blubber concentrations had proportionately higher concentrations than females in outer blubber when compared to males and females with lower inner blubber concentrations.

Additionally, during the late molt we detected a significant interaction between POP concentrations in inner blubber × sex on POP concentrations in outer blubber for α-HCH, β-HCH 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-BDE 47, inner blubber POP concentrations were positively related to outer blubber concentrations (Fig. 3). Inner blubber concentrations of α-HCH and β-HCH were positively related to outer blubber POP concentrations at late molting for females and males, separately (Fig. 3). Inner blubber concentrations

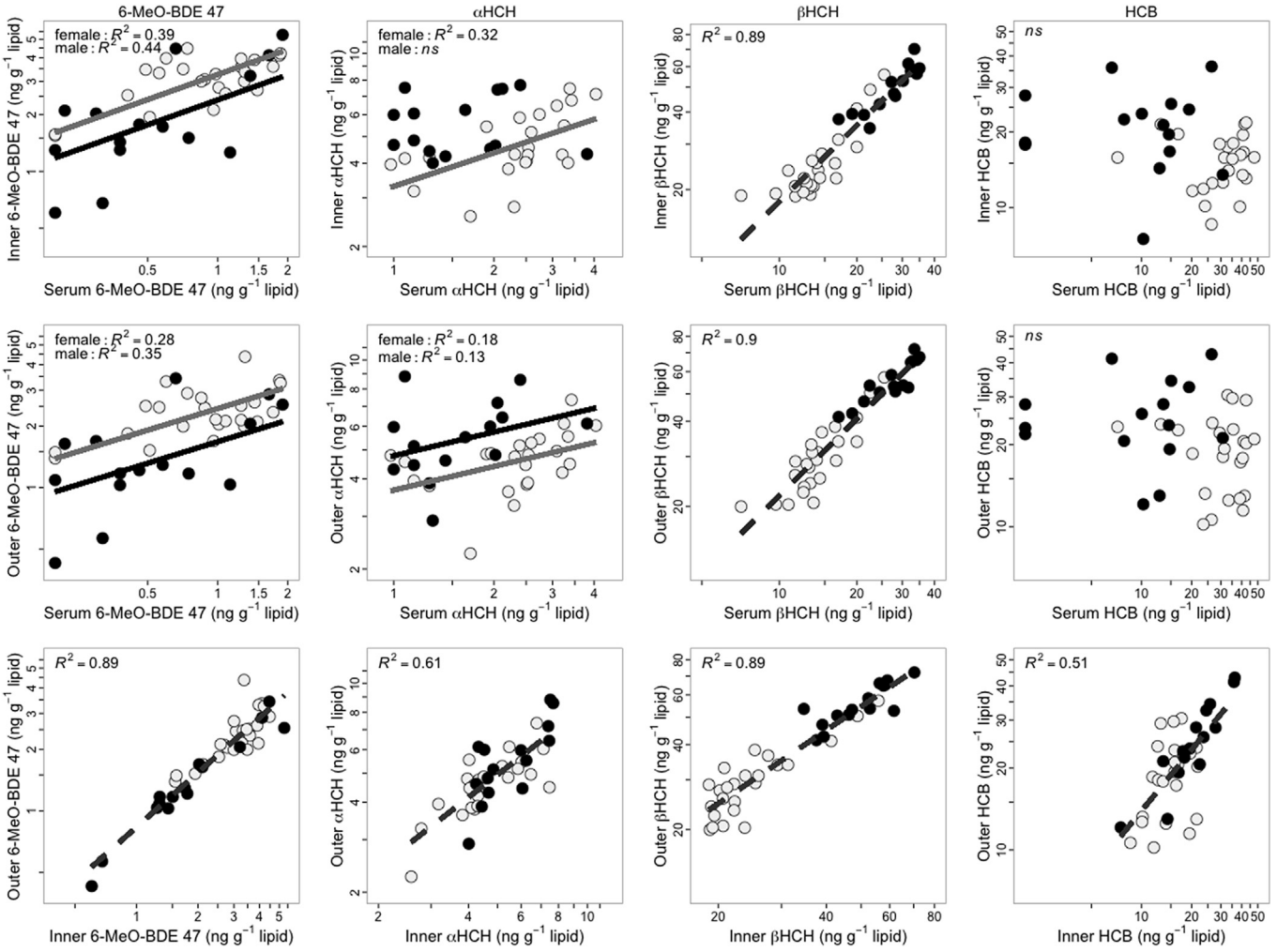

Fig. 4. Relationships between concentrations of 6-MeO-BDE 47, α-HCH, β-HCH, and HCB in inner blubber and serum (upper panels), outer blubber and serum (middle panels), and outer blubber and inner blubber (lower panels) of adult female and male northern elephant seals early in the breeding fast (post-foraging).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

of HCB were only significantly related for males and not females.

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

3.4. Partitioning coefficients

Inner blubber/serum partitioning coefficients for ΣDDTs, ΣCHLs, and ΣPBDEs were significantly lower at late molt than early breeding ($F_{1,45.9} = 15.64$, $p < 0.001$), but did not differ by sex ($F_{1,60.0} = 0.61$, $p = 0.44$; Table 4). For ΣPCBs, we detected a significant sex × time period interaction ($F_{1,59.6} = 10.25$, $p = 0.002$). Females and males did not differ in ΣPCB inner blubber/serum partitioning coefficients at late molting ($t = 1.15$, $p = 0.99$), but females had higher inner blubber/serum partitioning coefficients than males at early breeding ($t = 4.73$, $p < 0.001$), indicating that females had proportionally higher outer blubber concentrations relative to serum than males at early breeding. Female partitioning coefficients for ΣPCBs were lower at late molt relative to early breeding ($t = 5.75$, $p < 0.001$), while male coefficients for ΣPCBs did not differ ($t = 0.413$, $p = 0.98$).

For outer blubber/serum partitioning coefficients, we detected a significant interaction between sex and time period for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs ($F_{1,59.6} \geq 6.57$, $p \leq 0.01$; Table 4). Females and males did not differ at late molting for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs ($t \leq 1.85$, $p \geq 0.26$), but partitioning coefficients for females were significantly higher than males for each contaminant compound at early breeding ($t \geq 3.27$, $p \leq 0.01$). For males, partitioning coefficients were lower at late molting than early breeding for ΣDDTs and ΣCHLs ($t \geq 3.75$, $p \leq 0.002$), but not for ΣPCBs and ΣPBDEs ($t \leq 1.94$, $p \geq 0.22$; Table 4). For females, partitioning coefficients were lower during late molting than early breeding for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs ($t \geq 7.39$, $p \leq 0.001$; Table 4).

For outer blubber/inner blubber partitioning coefficients, we detected a significant sex × time period interaction for ΣPCBs ($F_{1,59.6} = 15.10$, $p = 0.003$), ΣCHLs ($F_{1,59.6} = 5.80$, $p = 0.02$), and ΣPBDEs ($F_{1,61.1} = 6.63$, $p = 0.01$), but not ΣDDTs ($F_{1,59.6} = 2.45$, $p = 0.12$). At late molting, outer blubber/inner blubber coefficients were lower for females relative to males for ΣPCBs ($t = 3.05$, $p = 0.02$; Table 4), but ΣCHLs and ΣPBDEs did not differ between

Table 4
Partitioning coefficient least-squares means (95% confidence interval) for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs in female and male elephant seals late in the molting fast and early in the breeding fast.

Type	Sex	Late molting	Early breeding
Inner blubber/serum			
ΣDDTs	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)
ΣPCBs	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)
ΣCHLs	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)
ΣPBDEs	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)
Outer blubber/serum			
ΣDDTs	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)
ΣPCBs	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)
ΣCHLs	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)
ΣPBDEs	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)
Outer blubber/inner blubber			
ΣDDTs	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)
ΣPCBs	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)
ΣCHLs	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)
ΣPBDEs	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)

males and females at this time period ($t \leq 1.13$, $p \geq 0.67$). Early in breeding, females and males did not differ for ΣPCBs, ΣCHLs, or ΣPBDEs (Table 4). For males, outer blubber/inner blubber partitioning coefficients were lower at late molting than early breeding for ΣCHLs ($t = 2.86$, $p = 0.028$), but the time periods did not differ for ΣPCBs or ΣPBDEs ($t \leq 1.48$, $p \geq 0.45$). Female outer blubber/inner blubber partitioning coefficients were lower at late molting than early breeding for ΣPCBs, ΣCHLs, and ΣPBDEs ($t \geq 5.05$, $p \leq 0.0001$). For ΣDDTs, outer blubber/inner blubber partitioning coefficients were significantly lower at late molt than early breeding ($F_{1,45.9} = 42.37$, $p < 0.001$), but did not differ by sex ($F_{1,60.0} = 2.96$, $p < 0.09$).

4. Discussion

4.1. Body condition fluctuations

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

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

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

4.2. Predictive equations

Serum generally performed well as a predictor of inner blubber POP concentrations for female and male elephant seals at two extremes in body condition. For example, we found relatively strong predictive relationships between serum and inner blubber for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs at both body condition extremes. Among tissue comparisons, serum and inner blubber were notable because both female and male seals could be included in the same equation for nearly all POP compounds during both time periods. Serum and inner blubber have higher turnover rates compared with outer blubber, which may be more important in these tissues than the differences in offloading mechanisms between males and females. The lack of detectable interactions between serum POP concentrations × sex, or an effect of sex, except for ΣPBDEs during early breeding, indicates that serum and inner blubber relate similarly in males and females despite concurrent gestation of a pup by females and the highly probable corresponding placental transfer of POPs, as well as transfer of POPs from mother to pup during the first 5 days of lactation.

Unlike inner blubber:serum relationships, for outer blubber:serum and outer blubber:inner blubber we detected a significant interaction between predictor tissue POP concentration and sex for all major POP compounds, except ΣPBDEs. When there was a significant interaction, and thus a difference in the slope of the relationships, male elephant seals were always characterized by a steeper slope compared with females, resulting in greater differences between male and female outer blubber concentrations at higher concentrations of serum or inner blubber than at lower concentrations. Although this may be attributed to differences in the impact of concurrent lactation on specific POP compounds during the breeding fast (Debier et al., 2012; Vanden Berghe et al., 2012), the fact that significant interactions also existed in late

molting, when lactation and gestation should not be having immediate effects, suggest that a lifetime of offloading opportunities for females may cause persistent differences in bioaccumulation among tissue compartments.

Furthermore, while predictive equations for males performed well among all tissues and nearly all POP compounds at both sampling periods, the strength of female tissue relationships was inconsistent across POP compounds. Female serum and inner blubber were strong predictors of outer blubber for ΣDDTs and ΣPBDEs, but weaker predictors for ΣPCBs and ΣCHLs. Female lactation physiology results in disproportionate mobilization and transfer to milk of some POP compounds over others (Debier et al., 2012; Vanden Berghe et al., 2013), which may explain the weaker performance of predictive equations for ΣPCBs and ΣCHLs. For example, ΣPCBs represents the sum of a suite of congeners, each with compound-specific log K_{ow} (octanol-water partitioning coefficient; Hawker and Connell, 1988). When females lactate during the breeding fast, lower lipophilic congeners are more readily mobilized from inner blubber to serum and from serum to milk during lactation (Debier et al., 2003, 2012a; Vanden Berghe et al., 2012), which may make serum POP concentrations an unreliable predictor of inner or outer blubber POP concentrations for some compounds in certain scenarios. In the current study, differences in lipophilicity of specific congeners may distort the ability to predict outer blubber concentrations from serum or inner blubber for ΣPCBs in both seasons ($R^2 \leq 0.44$) and for ΣCHLs in early breeding ($R^2 \leq 0.25$).

4.3. Interspecies comparisons

Significant positive relationships between blood and fat compartments have been observed and quantified for POPs in the tissues of polar bears (Bernhoft et al., 1997), turtles (Keller et al., 2004), dolphins (Yordy et al., 2010c), and Hawaiian monk seals (Lopez et al., 2012). Hawaiian monk seals and northern elephant seals are both pinnipeds, the taxonomic group that includes seals, sea lions, and walrus. Similar to the elephant seal inner blubber:-serum relationships in the present study, Hawaiian monk seals of various ages and both sexes had strong relationships between blubber and serum for ΣDDTs, ΣPCBs, and ΣCHLs (Lopez et al., 2012), while ΣPBDE relationships were not reported.

Our study of a pinniped species is similar in approach to a study on an odontocete, the taxonomic group that includes toothed cetaceans, which determined predictive equations in male and female bottlenose dolphins (*Tursiops truncatus*) from Sarasota Bay, Florida, USA (Yordy et al., 2010c). The equations were developed for ΣDDTs, ΣPCBs, ΣCHLs, and ΣPBDEs using plasma and full-thickness blubber, to encompass the variability of fatty acids and POP concentrations in the vertical blubber profile (Ellisor et al., 2013). Partitioning coefficients (blubber/plasma) were calculated for males, juveniles, and females (Yordy et al., 2010c). Northern elephant seals differ from bottlenose dolphins in several aspects, namely that they come ashore and undergo extreme fasting periods that mobilize POPs from blubber to serum (Debier et al., 2006). Additionally, the inter-offspring interval for dolphins is longer than for elephant seals, which provides less frequent opportunities for contaminant elimination. Finally, dolphins have a lipid-rich internal melon tissue that can accumulate POPs (Yordy et al., 2010a), while elephant seals do not. Although different in certain life history aspects, both northern elephant seals and bottlenose dolphins are relatively long-lived, top marine predators with relatively large fat compartments (blubber) that can bioaccumulate POPs from marine prey.

Both studies found positive relationships among POP concentrations in male blood and blubber. Predictive equations for bottlenose dolphins showed strong relationships for all POP classes ($R^2 \geq 0.91$), but Yordy et al. (2010) did not quantify predictive relationships for females. Bottlenose dolphin predictive equations for males and juvenile had slopes from 0.91 to 1.08, similar to male elephant seals at early breeding (0.78–0.99), when they had recently completed their foraging trip and thus were most directly comparable to continuously foraging bottlenose dolphins. Male elephant seal equations showed a greater range of slopes (0.89–1.42) for POP compounds late in the molting fast.

Additionally, blubber/plasma partitioning coefficients reported in Yordy et al. (2010) for male bottlenose dolphins were higher for ΣDDTs (mean = 1.95, 95% CI = 1.73–2.17) and ΣCHLs (2.52, 2.23–2.81), but lower for ΣPCBs (1.42, 1.24–1.60) and ΣPBDEs (1.24, 1.04–1.44), than either inner blubber/serum or outer blubber/serum partitioning coefficients for male elephant seals (Table 4). Further, female bottlenose dolphins (Yordy et al., 2010c) had lower partitioning coefficients for ΣDDTs (1.93, 1.66–2.20), ΣPCBs (1.77, 1.42–2.10), and ΣCHLs (2.35, 1.14–3.55) than female elephant seals early in the breeding fast, but higher partitioning coefficients than female elephant seals late in the molting fast. Early in breeding, female elephant seals have just returned from a long foraging trip, and have very recently given birth to a pup and begun lactation. Although recent lactation could eliminate some POPs from serum, higher partitioning coefficients in elephant seals early in the breeding fast are likely representative of greater dilution of POPs in serum compared with blubber, in response to recent foraging.

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

4.4. Implications for biomonitoring

Measurements of POP concentrations in marine mammals are common in oceans around the globe, but selection of tissues for sampling varies among studies. Therefore, the ability to estimate POP blubber concentrations from serum, or vice versa, has the potential to enhance toxicological assessment in marine mammals by providing a tool for expanding studies where only one tissue is collected. In cases where blubber samples are more attainable than blood (Elfes et al., 2010), the ability to estimate blood concentrations increases the capacity for toxicological risk assessment, because blood interacts with vital organs. In other cases, blood samples are more attainable or are banked from past collections and could be used to estimate blubber concentrations, which may enable comparisons across space or time.

Further, the distinction between inner and outer blubber is important. The use of biopsy darting to sample marine mammal blubber is increasingly used in the study of wild, free-ranging marine mammals, yet the relationship of that sample to inner blubber or serum is unclear. For example, outer blubber samples are commonly collected from large free-ranging mysticetes for studies of hormones and contaminants because outer blubber is often the only sample that can be obtained (*e.g.*, Hunt et al., 2015); therefore, the potential to link outer blubber to other tissues is particularly important. Because blubber is the largest reservoir for POPs, the relationship between blubber reserves and serum may enable estimations of peak serum concentrations relative to life history phases and variability in body condition. The seals in the current study were seemingly healthy animals that naturally

fluctuate in body condition, but may represent the processes that occur in other mammals undergoing rapid changes in body condition. Therefore, the relationship between contaminant concentrations in blood and blubber at such extremes may be an important factor to assessments of animal health and relative vulnerability to contaminant accumulation.

The detection of strong, positive relationships between concentrations of POP compounds in blood and blubber of two marine mammals, in the current study and in Yordy et al. (2010c), with clear behavior and life history differences raises the question: are predictive equations species-specific or can we determine generalities among species? If future research that involves direct handling of animals can produce additional blood-blubber relationships, then it may be possible to construct more generalized equations for groups of similar mammals. Generalized relationships, even for a few POPs, would enhance monitoring of contaminants in wildlife, particularly in situations where free-ranging animals are difficult to sample. Furthermore, comparisons among many species, including pinnipeds and odontocetes, could identify differences in contaminant metabolism among species, and thereby further inform models of potential health effects or risk assessment.

Acknowledgements

The Institutional Animal Care and Use Committee at the University of California, Santa Cruz approved all procedures and we captured animals under National Marine Fisheries Service permit 14636. We thank the many volunteers, technicians, and graduate students who were instrumental in the success of this project, especially P. Robinson, C. Goetsch, M. Tift, L. Hückstädt, L. McHuron, J. Sharick, D. Somo, D. Ensminger, and C. Louis. We thank the rangers and docents at Año Nuevo State Reserve, as well as P. Morris, and B. Hatfield for assistance in resighting seals, and R. Condit for development of the demographic database. Research and logistic support was provided by funds to S.H.P. from the Friends of Long Marine Lab, the Earl and Ethel Myers Oceanographic Trust, the University of California NRS Mildred Mathias Graduate Student Research Grant Program, the Rebecca and Steve Sooy Graduate Fellowship in Marine Mammals, the ARCS Foundation, and grants N00014-13-1-0134 and N00014-10-1-0356 to D.P.C. from the Office of Naval Research (ONR). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of this manuscript.

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