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# Deep-ocean foraging northern elephant seals bioaccumulate persistent organic pollutants<sup>\*</sup>



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#### HIGHLIGHTS

#### GRAPHICAL ABSTRACT

- All elephant seals had detectable concentrations of DDTs, PCBs, CHLs, and PBDEs.
- We quantified changes in the blubber burdens of POPs, within individual seals.
- Despite mass dilution while foraging, blubber burdens showed POP ingestion.
- Bioaccumulation of some POP compounds in seals varied across the North Pacific.
- Ratio of ΣDDTs:ΣPCBs corroborated latitudinal variation seen in other species.

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#### ABSTRACT

As top predators in the northeast Pacific Ocean, northern elephant seals (*Mirounga angustirostris*) are vulnerable to bioaccumulation of persistent organic pollutants (POPs). Our study examined a suite of POPs in blubber (inner and outer) and blood (serum) of free-ranging northern elephant seals. For adult females (N = 24), we satellite tracked and sampled the same seals before and after their approximately seven month long foraging trip. For males, we sampled different adults and sub-adults before (N = 14) and after (N = 15) the same foraging trip. For females, we calculated blubber burdens for all compounds. The highest POP concentrations in males and females were found for  $\sum$  DDTs and  $\sum$  PCBs. In blubber and serum, males had significantly greater concentrations than females for almost all compounds. For males,  $\sum$  DDT and  $\sum$  PBDEs were highly correlated in blubber and serum. While  $\sum$  PCBs were highly correlated with  $\sum$  DDTs and  $\sum$  PBDEs in blubber and serum for males,  $\sum$  PCBs showed weaker correlations with both compounds in females. As females gained mass while foraging, concentrations of nearly all POPs in inner and outer blubber significantly decreased; however, the absolute burden in blubber significantly increased, indicating ingestion of contaminants while foraging. Additionally, we identified three clusters of seal foraging behavior, based on geography, diving behavior, and stable carbon and nitrogen isotopes, which corresponded with differences in  $\sum$  DDTs,  $\sum$  PBDEs, MeO-BDE 47, as well as the ratio of  $\sum$  DDTs to  $\sum$  PCBs, indicating the

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potential for behavior to heighten or mitigate contaminant exposure. The greatest concentrations of  $\sum$  DDTs and  $\sum$  PBDEs were observed in the cluster that foraged closer to the coast and had blood samples more enriched in <sup>13</sup>C. Bioaccumulation of POPs by elephant seals supports mesopelagic food webs as a sink for POPs and highlights elephant seals as a potential sentinel of contamination in deep ocean food webs.

#### 1. Introduction

Persistent organic pollutants (POPs) are a continued threat to wildlife because they are widely dispersed, bioaccumulate in top predators, and can disrupt physiological pathways, thus leading to adverse health effects (Tartu et al., 2015; Jenssen, 2006). Despite international regulation (2009 Stockholm Convention) and bans of some POPs by individual countries, the presence of polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT) and its metabolites, polybrominated diphenyl ethers (PBDEs), chlordanes (CHLs), and other POPs in marine and terrestrial ecosystems remains pervasive (Braune et al., 2005; Farrington and Takada, 2014). While POPs have multiple sources and mechanisms of transport, the deep ocean can serve as a sink (Farrington and Takada, 2014), where POPs can enter marine food webs and can magnify with trophic level (Weijs et al., 2009). POPs are resistant to biological degradation and associate mainly with lipids in biological organisms (Muir et al., 1999). The legacy of POPs is important for marine mammals because persistent exposure to even low levels of POPs can influence mammalian endocrine systems (Tanabe, 2002), neural function (Haijima et al., 2010; Winneke, 2011) and immune systems (Ross et al., 1996; Schwacke et al., 2012). Bioaccumulation of POPs in marine mammals may have population-level consequences through the combination of weakened immune function and infectious disease (Hall et al., 2006). In addition, POPs in adult female marine mammals are concerning because POPs can transfer from mother to offspring via placental transfer and lactation (Greig et al., 2007; Vanden Berghe et al., 2012; Wolkers et al., 2004), meaning that young animals are exposed to a suite of contaminants during critical periods of development.

The main source of POPs to marine mammals is through food (Muir et al., 1999), therefore specific foraging behaviors, including location or diet, may exacerbate or mitigate POP exposure. For example, proximity to industrialization and point sources of contaminant entry into the environment can result in higher POP bioaccumulation in some marine mammals than others (Frouin et al., 2011; Lopez et al., 2014; Ross et al., 2004; Schwacke et al., 2012). Differences in diet, within and among species, can also influence bioaccumulation of POPs (Bentzen et al., 2008; Ross et al., 2000). Concentrations of POPs are often higher in males than females because females can transfer contaminants to offspring while males are unable to offload any of their accumulated contaminant burden (Barron et al., 2003; Storelli et al., 2012; Wang et al., 2007). Marine mammals are vulnerable to bioaccumulation of POPs because they often have relatively long life spans over which to accumulate POPs, and many species are more susceptible to biomagnification due to their high trophic position.

Studies of contaminant bioaccumulation in mammals are challenging because factors such as animal age and body condition can confound analyses. Contaminants are primarily reported in concentrations, either by wet weight or lipid weight; however, concentrations of POPs are significantly influenced by body condition and physiological state in both stranded and free-ranging animals (Debier et al., 2012; Hall et al., 2008; Myers and Atkinson, 2012; Peterson et al., 2014). Varying contaminant concentrations have been observed for a significant number of marine mammal species, although many studies have focused on quantification of contaminants in stranded animals and less so in freeranging animals. Contaminant concentrations in stranded and deceased animals may not represent healthy animals due to the complications of disease, dehydration, or starvation; therefore, sampling free-ranging animals provides a more complete quantification of the range of bioaccumulation experienced by a population as a whole. Additionally, guantification of the blubber burden of contaminants (mg contaminant) and the subsequent change in burden over time makes it possible to track changes in contaminants contained within the blubber layer, regardless of physiological changes. Indeed, variations of blubber POP concentrations, which are directly related to physiological state and blubber mass, do not necessarily reflect variations of POP burden. For example, increased POP concentrations in blubber of northern elephant seals at the end of the lactation-associated fast, compared with the beginning of the fast, may actually correspond to a decreased body burden as a result of POP excretion through milk (Debier et al., 2012). In contrast, lower POP concentrations in elephant seal blubber after a foraging trip (Peterson et al., 2014) may correspond to a greater body burden as a result of foraging. Furthermore, the inner and outer portions of blubber differ in their metabolic activity during fasting and foraging periods (Fowler et al., 2014; Strandberg et al., 2008), which may differentially influence contaminant concentrations across the blubber layer. The challenges associated with repeatedly sampling the same free-ranging animals have limited the number of studies that directly link individual foraging behavior with contaminant bioaccumulation (inner and outer blubber) or changes in contaminant burden.

As relatively long-lived, high trophic level predators foraging in the mesopelagic (200–1000 m) northeast Pacific Ocean, northern elephant seals (*Mirounga angustirostris*) can serve as biomonitors of remote ocean habitats that are difficult to sample. Elephant seals undergo biannual foraging trips, ranging upwards of 5000 or 10000 km depending on the season, within several open-ocean and near-coastal hydrographic ecoregions, including the Subarctic Gyre, the North Pacific Polar Front, and the California Current (Le Boeuf et al., 2000; Robinson et al., 2012; Springer et al., 1999). The northern elephant seal is the only pinniped species in the North Pacific that forages almost entirely on fish and squid in the mesopelagic zone (Antonelis et al., 1987; Le Boeuf et al., 2000; Naito et al., 2013). Biannual foraging trips (Fig. 1) are interspersed with extensive fasting periods on land, at which time individuals lose up to 40% of their body mass (Costa et al., 1986; Worthy et al., 1992).

Our study is one of few to repeatedly sample individuals at the start and end of a long foraging trip and to calculate variations in the contaminant mass in blubber. For this study, our objectives were to use freeranging northern elephant seals to: 1) Measure a suite of POP compounds and compare the concentrations between adult females and males before and after the foraging trip, 2) Determine how well correlated different POP compounds are in females and males upon arrival to the colony at the end of the foraging trip, 3) Quantify changes in concentrations (females – paired seals, males – unpaired seals) and blubber burdens (females) from the start to the end of the foraging trip, and 4) Determine if contaminant concentrations and blubber burdens in females vary with clusters of foraging behavior.

#### 2. Methods

#### 2.1. Animal sampling

We collected paired blubber and blood samples from adult northern elephant seals at the Año Nuevo State Reserve  $(37.11^{\circ} \text{ N}, 122.33^{\circ} \text{ W})$  in 2012 and 2013. 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) the approximately seven month foraging trip (Fig. 1). Blubber cores and blood samples were also 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 at the start of the early breeding fast (Fig. 1). Due to the challenges associated with repeatedly sampling males, we were unable to



Fig. 1. One year in the life of a northern elephant seal. A year in the life of an adult female (left) or adult male (right) northern elephant seal (*Mirounga angustirostris*). Individual females are on land for approximately four–five weeks during the breeding fast and five–six weeks during the molting fast. Males are on land for longer during the breeding fast and a similar length of time during the molting fast. Note that individual animals are onshore for less time than the full periods shown above because seals do not all arrive to the colony at the same time.

compare paired male samples across the foraging trip. However, we were able to collect blubber and serum samples from four paired males early and late in the breeding fast (48–53 days between samples), to examine if changes in POP concentrations were associated with changes in body condition.

To attach satellite tags, obtain tissue samples, and to collect morphometric measurements, we followed standard procedures for chemical immobilization of elephant seals (Le Boeuf et al., 2000; Robinson et al., 2012). We collected blood samples from the extradural vein, stored samples on ice for return to the lab on the same day, centrifuged blood, and stored serum samples in glass vials. A full-thickness blubber core was collected from the lateral pelvic area of each seal using a sterile 6 mm biopsy punch (Miltex, Inc., York, Pennsylvania, USA) and stored in aluminum foil. Red blood cells were collected for stable carbon and nitrogen analysis and were analyzed following previously published methods (Hückstädt et al., 2011; Peterson et al., 2015). Blubber cores and blood samples were stored at -20 °C until analysis.

We collected tissue samples and morphometric measurements each time an animal was handled. Measurements included girth and length taken at eight locations along the seal, including six locations where blubber thickness was measured dorsally, laterally, and ventrally, using either a handheld ultrasound backfat meter (Scanoprobe, Ithaca, New York, USA) or a Signos handheld portable ultrasound (Signostics Ltd, Clovelly Park, Australia) (Robinson et al., 2012; Schwarz et al., 2015). Body mass (females only) was determined by rolling each seal into a canvas sling and using a hand-winch to suspend them from a 1000  $\pm$  1.0 kg Dyna-Link digital scale attached to a metal tripod (Robinson et al., 2012).

#### 2.2. Laboratory analysis

In all samples, 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 ( $\alpha$ -,  $\beta$ -, and  $\gamma$ -HCH), and two naturally-produced methoxylated PBDEs (2'-MeO-BDE 68 and 6-MeO-BDE 47) were targeted. Extraction, clean-up, and concentration measurement methods followed protocols described in Vanden Berghe et al. (2012).

In brief, the skin layer and fur (1.3 cm deep on average, Schwarz et al., 2015), was removed from the outer portion of the biopsy core, and the remaining blubber was cut into inner and outer segments of approximately equal mass. Each blubber segment was dried with Na<sub>2</sub>SO<sub>4</sub>, placed in a thimble, and spiked with internal standards (BDE 77, BDE 128, ε-HCH, and CB 143). Lipids and target contaminants were extracted using hot Soxhlet extraction with hexane/acetone (3:1, v:v) for 2 h. An aliquot of the extract (typically 1/8th) was used for gravimetric determination of lipids (105 °C, 1 h). The rest of the extract was cleaned on approximately 8 g acid silica (44% H<sub>2</sub>SO<sub>4</sub>, w/w) and eluted with 20 ml hexane, followed by 15 ml dichloromethane. The cleaned extract was evaporated to dryness and reconstituted in 150 µl iso-octane.

Serum samples were split for separate determination of target contaminants and lipids. For contaminant determination, serum samples were spiked with internal standards (CB 143, E-HCH, and BDE 77) diluted with Milli-Q water, mixed with formic acid for protein denaturation, and sonicated for 20 min. Samples were extracted on solid phase extraction cartridges (Oasis HLB, 60 mg/3 ml). Extracts were eluted with dichloromethane (7.5 ml), concentrated, and further reconstituted with hexane (0.5 ml). Extracts were cleaned on 1 g acid silica (44% H<sub>2</sub>SO<sub>4</sub>, w/w) topped with 100 mg Na<sub>2</sub>SO<sub>4</sub>, eluted with 10 ml hexane:dichloromethane (1:1, v:v), evaporated to near-dryness, and resolubilized in 100 µl isooctane. 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).

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  $\times$  0.25 mm  $\times$  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  $\times$  0.22 mm  $\times$  0.25 µm HT-8 column (SGE, Zulte, Belgium) by monitoring 2 ions for each homologue group.

We scaled lipid-normalized POP concentrations up to a blubber burden for each seal using a multiple step process. We used the relative contribution of the inner and outer blubber layers to the full thickness blubber core, based on mass, to obtain an overall lipid-normalized POP

#### Table 1

The same adult female elephant seals (N = 24) were sampled during late molting and early breeding for POPs in inner blubber, outer blubber and serum. Unpaired males were sampled during late molting (N = 14) and early breeding (N = 15). POP concentrations were quantified in all three tissue types for every animal. Blubber percent lipid is reported as percent lipid (w/w) and serum is reported as g lipid dl<sup>-1</sup> serum (mean  $\pm$  SD). Lipid-normalized concentrations (ng g<sup>-1</sup> lipid) are reported as mean  $\pm$  SD (*number of samples above the detection limit*<sup>a</sup>: range of concentrations). Italics indicate what percent of the total (i.e.,  $\Sigma$ PCBs or  $\Sigma$ PBDEs) can be attributed to a specific group of congeners (i.e.,  $\Sigma$ tri-CBs) or a specific congener (i.e., BDE 47).

TypeYetJetJetaJetaJetaJetaJetaJetaJetaJetaJetaSeries158,97,97,97,97,97,97,97,97,97,97,97,97,97,			Late molting fast			Early breeding fast			
Prevent lipid         Featule         63.2 ± 6.0 $(24 \pm 32)$ $(0.84 + 2.4)$ $(0.73 + 4.5)$ $(0.87 + 1.08)$ SDDT <sup>b</sup> Featule $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.74 - 1.23)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$ $(0.84 + 3.5)$	Туре	Sex	Inner	Outer	Serum	Inner	Outer	Serum	
Pretent inpo         Penne         No.1 ± 0.01 (75-805)         No.1 ± 0.03 (75-805)	Demonst Hald	<b>F</b>	02.1 + 0.0	02.4 + 2.2	0.042 + 0.120	07.4 + 2.4	027 45	0.000 + 0.000	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	Percent lipid	Feinale	$82.1 \pm 0.0$	$82.4 \pm 3.2$	$0.942 \pm 0.120$	$8/.4 \pm 2.4$	$82.7 \pm 4.5$	$0.998 \pm 0.003$	
Instr.         Cont. 2017         Cont. 2017<		Mala	(05.7 - 91.7)	(75.9-67.2)	(0.710 - 1.252) 1 109 + 0 147	(01.9-91.1)	(73.6-94.5)	(0.675 - 1.095) 1.020 + 0.215	
SDDP <sup>1</sup> Penale         (1756 ± 4m7.4)         (1774 ± 9m7.4)         (106 ± 9m5.4)		Wale	(670.995)	(60.0, 92.1)	(0.740, 1.221)	$(72.0 \pm 3.3)$	(606, 825)	(0.706, 1.622)	
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	5'DDTs <sup>b</sup>	Female	(07.9-88.5) 1736 0 $\pm$ 603 2	(00.0-85.1) 1579 1 $\pm$ 487 6	(0.740 - 1.321) 1273 9 $\pm$ 387 2	(72.3-89.0) 11805 $\pm$ 5365	(00.0-85.5) 1473 4 $\pm$ 485 7	(0.790 - 1.033) 684.8 $\pm$ 339.0	
	20013	remaie	(902.9 - 3354.8)	(865.1 - 2677.2)	(6237-23606)	(598.1 - 3086.8)	(875.3 - 2803.7)	(365.3 - 1912.0)	
		Male	(302.3 + 3034.0) 3161 3 + 2189 2	$(803.1 \pm 2077.2)$ 2848 1 $\pm$ 1774 6	(025.7 + 2500.0) 2066 3 + 869 0	(330.1 + 2952.9)	$(075.5 \pm 2005.7)$ 3458 8 $\pm$ 3450 0	(305.5 + 1699.8)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		mare	(1355 3 - 8730 2)	(1054.6-7107.3)	(823 4-3778 1)	(7057 - 126895)	(770.8 - 15061.3)	(390.1 - 7532.6)	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$	% <i>p.p</i> ′-DDE	Female	$99.2 \pm 0.3$	$99.2 \pm 0.3$	$99.0 \pm 0.2$	$98.6 \pm 0.5$	$99.0 \pm 0.3$	$98.3 \pm 0.4$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(98.3–99.7)	(98.5-99.7)	(98.0-99.0)	(97.4–99.7)	(98.3-99.7)	(98.0-99.0)	
SAC05'         (980-989)         (980-989)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (980-969)         (		Male	$99.5 \pm 0.3$	$99.5 \pm 0.3$	$99.1 \pm 0.5$	$99.2 \pm 0.6$	$99.5 \pm 0.3$	$99.1 \pm 0.7$	
SPCBs'         Fenale         11114 ± 278.3         800.5 ± 142.1         07.3 ± 10.90         722.5 ± 10.90         781.1 ± 135.5         322.0 ± 73.7           Male         1084.2 ± 997.6         155.9 ± 10.3.2         994.4 ± 72.7         1557.0 ± 38.81         1558.1 ± 815.9         922.3 ± 494.6           % 201.0 B         Fenale         -0.1 ± -0.1         0.0 ± 0.0         -0.1 ± -0.1         (0.0 ± 0.0)         -0.1 ± -0.1         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.1 ± -0.1)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.0 ± 0.0)         (0.			(98.9–99.8)	(99.0-99.9)	(98.0-100.0)	(97.5-99.9)	(98.9-99.9)	(98.0-100.0)	
Male         (5807-1802.)         (510-1052.)         (482-4997.)         (482-4997.)         (482-4997.)         (580-1028.)         (151-397.1)         (230-217.2)           % 2ni-GB         Penale         (13-0)         (0.4-4.399.7)         (482-4997.2)         (0.4-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)         (0.1-0.1)	ΣPCBs <sup>c</sup>	Female	$1111.4 \pm 278.3$	$800.5 \pm 142.1$	$673.9 \pm 169.0$	$722.6 \pm 169.9$	$781.1 \pm 135.5$	$322.0\pm73.7$	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			(580.7-1806.2)	(501.3-1053.2)	(445.4-997.1)	(422.1-1029.8)	(539.6-1046.8)	(194.7-470.2)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $		Male	$1684.2 \pm 997.6$	$1559.6 \pm 1013.2$	$994.4 \pm 372.7$	$1557.0 \pm 838.1$	$1516.1 \pm 815.9$	$922.3\pm494.6$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(733.5-4610.6)	(584.4-4396.7)	(437.1-1679.7)	(448.8-3942.7)	(517.0-3975.1)	(236.6-2317.7)	
$ \begin{array}{c c} & (-a) - (-a) & (a) - (a) & (-a) - (-a) & (-a) - (-a) & (a) - (a) & (-a) & (-a) & (-a) \\ & (-a) - (-a) & (a) & (-a) & $	% Σtri-CBs	Female	$<0.1 \pm <0.1$	$0.0\pm0.0$	$<\!0.1 \pm <\!0.1$	$<0.1 \pm <0.1$	$0.0\pm0.0$	$0.1\pm0.4$	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			(<0.1-<0.1)	(0.0-0.0)	(<0.1-<0.1)	(<0.1-0.3)	(0.0-0.0)	(<0.1–1.9)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	$<0.1 \pm <0.1$	$0.0\pm0.0$	$<0.1 \pm <0.1$	$<0.1 \pm <0.1$	$0.0\pm0.0$	<0.1 ± <0.1	
$ \begin{array}{c} $ 2 \mbox{ Lettra-Lus} \\ $ $ Penhale $ 2s \pm 1/$, $ 2s \pm 1/$, $ 2s \pm 1/$, $ 2s \pm 1/$, $ 1/1 \pm 0.8$ $ 1$	0( 5) · · · · · · · · · · · · · · · · · ·		(<0.1-<0.1)	(0.0-0.0)	(<0.1-<0.1)	(<0.1-<0.1)	(0.0-0.0)	(<0.1-<0.1)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% Stetra-CBs	Female	$2.8 \pm 0.7$	$3.7 \pm 0.8$	$2.1 \pm 0.8$	$3.4 \pm 0.9$	$3.7 \pm 0.8$	$2.5 \pm 0.9$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Mala	(1.8-5.0)	(2.6-5.6)	(0.8-4.5)	(1.9-5.4)	(2.3-5.1)	(1.6-4.7)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	$3.3 \pm 0.8$	$3.6 \pm 1.1$	$3.3 \pm 0.8$	$3.0 \pm 1.4$	$3.5 \pm 1.5$	$2.3 \pm 1.1$	
$ \begin{array}{c} \text{s} 2, \text{prime-bs} & \text{reline} & 10, \pm 2.2, \\ (13, \pm 2.2), & (13, \pm 2.3), & (13, \pm 2.3), \\ (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), \\ (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), \\ (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), \\ (14, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), & (13, \pm 2.3), \\ (19, -244), & (19, -244), & (19, -244), & (12, \pm 2.4), & (12, -23), & (12, \pm 2.4), & (12, \pm 2.4), \\ (19, -244), & (19, -244), & (12, -425), & (22, \pm 1.4), & (23, -52, -56), & (24, -44, -48), & (23, \pm 2.4), & (23, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (24, -56), & (23, -57), & (23, -56), & (23, -56), & (23, -56), & (23, -56), & (23, -56), & (23, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, -56), & (23, -56), & (24, $	% Sponta CPc	Formalo	(1.8 - 4.0) 17.0 + 2.2	(1.4-0.0)	(1.0-3.1)	(1.4-0.5)	(1.3-0.3)	(0.8 - 5.0)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% 2pend-Cbs	rennale	(13.4-22.3)	$19.4 \pm 2.0$ (15.5-24.0)	$23.3 \pm 3.4$ (105-310)	(137-225)	$(15.0 \pm 2.0)$	$32.4 \pm 3.1$ (275-385)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	(13.4-22.5) 21.8 + 1.5	(13.3-24.0) 22.0 + 1.6	(19.3-51.5) $32.9 \pm 2.8$	(13.7 - 22.5) 20.2 + 2.6	(13.0-22.3) 21.4 + 2.9	(27.5-30.5) 29 2 + 4 3	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		where	(190-244)	(194-245)	(281-377)	(162-238)	(170-267)	(214 - 349)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% Σhexa-CBs	Female	$(13.0 \pm 1.1)$ 519 + 15	(13.1 2 1.3) $52.8 \pm 1.4$	525 + 19	522 + 16	$525 \pm 14$	$(2111 \ 31.5)$ $489 \pm 26$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	in griena ebb	remarc	(48.6-54.4)	(49.6-55.5)	(46.8-54.9)	(49.3-55.1)	(50.0-54.6)	(43.0-52.6)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	54.0 + 1.4	54.4 + 1.6	49.6 + 2.3	54.1 + 2.0	54.2 + 2.4	51.7 + 2.4	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(51.9-57.3)	(51.1-57.3)	(44.1-53.2)	(49.5-57.1)	(49.0-57.6)	(47.0-55.1)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	% Σhepta-CBs	Female	$22.3 \pm 2.7$	$19.2 \pm 2.7$	$17.4 \pm 2.6$	$21.0 \pm 2.5$	$20.1 \pm 2.6$	$14.4 \pm 1.3$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(16.0-27.2)	(15.5-24.4)	(13.5-22.3)	(16.9-24.9)	(15.7–24.4)	(11.8-16.2)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	$16.8 \pm 1.3$	$16.2 \pm 1.1$	$12.5 \pm 1.3$	$18.3 \pm 2.3$	$16.8 \pm 2.1$	$14.8 \pm 2.8$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(14.7–18.5)	(14.8–18.2)	(10.8–15.1)	(15.1–22.7)	(13.9–21.2)	(10.7–21.1)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% Σocta-CBs	Female	$3.9 \pm 0.9$	$2.7 \pm 0.7$	$1.4 \pm 0.3$	$3.5 \pm 0.8$	$2.9\pm0.7$	$1.1 \pm 0.1$	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$			(2.2–5.4)	(1.8–4.0)	(0.9–2.2)	(2.2–4.8)	(1.9–3.9)	(0.8–1.4)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	$2.7 \pm 0.5$	$2.4 \pm 0.4$	$1.0 \pm 0.2$	$3.2 \pm 0.7$	$2.7 \pm 0.6$	$1.3 \pm 0.4$	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% Spona-CBs	Female	(2.0-3.9) 0.2 $\pm < 0.1$	(2.0-5.5) 01 $\pm < 0.1$	(0.0-1.3)	(2.0-4.0)	(1.7-5.3) 01 $\pm < 01$	(0.7 - 2.2)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	70 ZHOHa CD3	remare	(0.1 - 0.4)	(< 0.1 - 0.2)	$(< 0.1 \pm (0.1)$	(< 0.1 - 0.3)	(< 0.1 - 0.2)	$(< 0.1 \pm (0.1)$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	0.1 + < 0.1	0.1 + < 0.1	< 0.1 + < 0.1	0.2 + < 0.1	(0.1 + < 0.1)	< 0.1 + < 0.1	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			(<0.1-0.3)	(<0.1-0.2)	(<0.1-0.1)	(<0.1-0.3)	(<0.1-0.2)	(<0.1-0.2)	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $	% Σdeca-CBs	Female	0.1 + < 0.1	< 0.1 + < 0.1	0.0 + 0.0	0.1 + < 0.1	<0.1 + <0.1	0.0 + 0.0	
$ \begin{array}{c c c c c c c c c c c c c c c c c c c $			(<0.1-0.3)	(<0.1-0.1)	(0.0-0.0)	(<0.1-0.2)	(<0.1-0.1)	(0.0-0.0)	
$\begin{split} \Sigma CHLs^d & Female & (-0.1-0.1) & (-0.10.1) & (0.0-0.0) & (-0.1-0.1) & (-0.10.1) & (0.0-0.0) \\ (-0.1-0.1) & (-0.10.1) & (0.0-0.0) & (-0.1-0.1) & (-0.10.1) & (0.0-0.0) \\ (-0.1-0.1) & (-0.10.1) & (-0.10.1) & (0.0-0.0) & (-0.1-0.1) & (-0.10.1) & (0.0-0.0) \\ (-0.1-0.1) & (-0.10.1) & (-0.10.1) & (-0.10.1) & (-0.10.1) & (-0.10.1) & (-0.10.1) & (-0.0-0.0) \\ (-0.1-0.1) & (-0.1-0.1) & (-0.10.1) & (-0.10.1) & (-0.10.1) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0) & (-0.0-0.0)$		Male	<0.1 ± <0.1	<0.1 ± <0.1	$0.0 \pm 0.0$	<0.1 ± <0.1	<0.1 ± <0.1	$0.0 \pm 0.0$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(<0.1-0.1)	(<0.1-<0.1)	(0.0-0.0)	(<0.1-0.1)	(<0.1-<0.1)	(0.0-0.0)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	ΣCHLs <sup>d</sup>	Female	$361.7\pm85.1$	$277.9 \pm 41.0$	$196.5 \pm 38.6$	$234.6\pm56.2$	$257.6\pm32.9$	$112.0\pm24.8$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(242.8-532.3)	(211.2-352.6)	(120.5-271.3)	(158.1-373.2)	(207.2-327.9)	(74.9-175.1)	
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$		Male	$578.7 \pm 265.4$	$498.4 \pm 247.9$	$322.2 \pm 93.6$	$484.0 \pm 126.9$	$492.0 \pm 130.3$	$251.8 \pm 74.3$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$			(320.2-1262.2)	(247.2-1061.3)	(187.9-468.7)	(237.1-684.5)	(233.6-729.2)	(111.2-350.7)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	% OxC	Female	$16.3 \pm 2.4$	$17.5 \pm 2.7$	$22.0 \pm 2.9$	$15.8 \pm 2.3$	$17.0 \pm 2.4$	$23.1 \pm 3.0$	
Male $20.0 \pm 2.4$ $20.8 \pm 2.2$ $28.2 \pm 2.3$ $186 \pm 2.0$ $19.6 \pm 1.9$ $24.0 \pm 2.4$ $(17.0-26.2)$ $(17.8-26.1)$ $(24.9-33.5)$ $(14.8-21.3)$ $(15.9-23.1)$ $(20.2-28.3)$ % TCFemale $1.1 \pm 0.2$ $1.3 \pm 0.2$ $1.0 \pm 0.2$ $1.2 \pm 0.1$ $1.2 \pm 0.1$ $1.3 \pm 0.4$ $(0.9-1.6)$ $(1.0-1.7)$ $(0.7-1.4)$ $(0.9-1.6)$ $(1.1-1.6)$ $(1.0-3.2)$ Male $1.0 \pm 0.5$ $1.0 \pm 0.5$ $0.9 \pm 0.4$ $0.9 \pm 0.3$ $1.0 \pm 0.3$ $0.8 \pm 0.3$ $(0.4-1.9)$ $(0.4-1.9)$ $(0.4-1.6)$ $(0.5-1.3)$ $(0.5-1.5)$ $(0.4-1.3)$ % CCFemale $2.9 \pm 0.5$ $3.1 \pm 0.4$ $3.5 \pm 0.7$ $4.1 \pm 1.0$ $3.2 \pm 0.5$ $6.4 \pm 1.7$ $(1.8-3.8)$ $(2.3-3.8)$ $(2.1-4.7)$ $(2.6-6.3)$ $(2.3-4.3)$ $(0.9-8.5)$ Male $2.1 \pm 0.8$ $2.0 \pm 0.8$ $2.8 \pm 1.1$ $2.3 \pm 0.9$ $2.0 \pm 0.7$ $3.1 \pm 1.3$ $(0.7-3.3)$ $(0.8-3.4)$ $(1.4-4.8)$ $(1.1-4.4)$ $(1.1-3.7)$ $(1.2-5.5)$ % TNFemale $67.5 \pm 2.5$ $65.9 \pm 2.4$ $58.6 \pm 2.5$ $66.4 \pm 2.0$ $66.0 \pm 1.9$ $54.7 \pm 2.2$ $(62.0-71.2)$ $(61.0-69.2)$ $(53.5-63.2)$ $(62.0-70.6)$ $(62.2-68.6)$ $(51.0-60.2)$ Male $63.2 \pm 1.9$ $62.6 \pm 2.1$ $52.7 \pm 1.7$ $64.7 \pm 2.2$ $64.1 \pm 2.4$ $55.1 \pm 2.3$ $(59.7-66.8)$ $(57.9-66.0)$ $(50.3-56.8)$ $(61.5-70.0)$ $(55.5-69.4)$ $(51.0-60.0)$ % CNFemale $12.2 $			(13.3-21.9)	(13.5-22.4)	(18.0-28.4)	(12.7-21.8)	(13.9–21.8)	(18.8–28.5)	
% TCFemale $(17.0-26.2)$ $(17.3-26.1)$ $(24.9-3.5)$ $(14.8-21.3)$ $(15.9-23.1)$ $(22.2-2.3)$ % TCFemale $1.1 \pm 0.2$ $1.3 \pm 0.2$ $1.0 \pm 0.2$ $1.2 \pm 0.1$ $1.2 \pm 0.1$ $1.3 \pm 0.4$ $(0.9-1.6)$ $(1.0-1.7)$ $(0.7-1.4)$ $(0.9-1.6)$ $(1.1-1.6)$ $(1.0-3.2)$ Male $1.0 \pm 0.5$ $1.0 \pm 0.5$ $0.9 \pm 0.4$ $0.9 \pm 0.3$ $1.0 \pm 0.3$ $0.8 \pm 0.3$ $(0.4-1.9)$ $(0.4-1.9)$ $(0.4-1.6)$ $(0.5-1.3)$ $(0.5-1.5)$ $(0.4-1.3)$ % CCFemale $2.9 \pm 0.5$ $3.1 \pm 0.4$ $3.5 \pm 0.7$ $4.1 \pm 1.0$ $3.2 \pm 0.5$ $6.4 \pm 1.7$ $(1.8-3.8)$ $(2.3-3.8)$ $(2.1-4.7)$ $(2.6-6.3)$ $(2.3-4.3)$ $(0.9-8.5)$ Male $2.1 \pm 0.8$ $2.0 \pm 0.8$ $2.8 \pm 1.1$ $2.3 \pm 0.9$ $2.0 \pm 0.7$ $3.1 \pm 1.3$ $(0.7-3.3)$ $(0.8-3.4)$ $(1.4-4.8)$ $(1.1-4.4)$ $(1.1-3.7)$ $(1.2-5.5)$ % TNFemale $67.5 \pm 2.5$ $65.9 \pm 2.4$ $58.6 \pm 2.5$ $66.4 \pm 2.0$ $66.0 \pm 1.9$ $54.7 \pm 2.2$ $(62.0-71.2)$ $(61.0-69.2)$ $(53.5-63.2)$ $(62.0-70.6)$ $(62.2-68.6)$ $(51.0-60.2)$ Male $63.2 \pm 1.9$ $62.6 \pm 2.1$ $52.7 \pm 1.7$ $64.7 \pm 2.2$ $64.1 \pm 2.4$ $55.1 \pm 2.3$ $(59.7-66.8)$ $(57.9-66.0)$ $(50.3-56.8)$ $(61.5-70.0)$ $(59.5-69.4)$ $(51.0-60.0)$ % CNFemale $12.2 \pm 0.5$ $12.2 \pm 0.4$ $14.9 \pm 0.7$ $12.5 \pm 0.5$ $12.6 \pm 0.5$ $14.5 \pm 0.8$		Male	$20.0 \pm 2.4$	$20.8 \pm 2.2$	$28.2 \pm 2.3$	$18.0 \pm 2.0$	$19.6 \pm 1.9$	$24.0 \pm 2.4$	
* IC       remate $1.1 \pm 0.2$ $1.5 \pm 0.2$ $1.0 \pm 0.2$ $1.2 \pm 0.1$ $0.1 \pm 0.3$ $0.6 \pm 0.1$ $0.5 \pm 0.5$ $0.8 \pm 0.3$ $0.6 \pm 0.5$ $0.6 \pm 1.1$ $0.5 \pm 0.5$ $0.6 \pm 1.1$ $0.5 \pm 0.5$ $0.6 \pm 1.1$ $0.5 \pm 0.5$ $0.6 \pm 1.1$ $0.6 \pm 0.5$	% TC	Formalo	(17.0-20.2)	(17.8-20.1)	(24.9-33.5)	(14.8 - 21.3) 1.2 + 0.1	(15.9-23.1)	(20.2 - 28.3)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	/o TC	rennale	$1.1 \pm 0.2$	$1.5 \pm 0.2$ (10, 17)	$1.0 \pm 0.2$ (0.7, 1.4)	$1.2 \pm 0.1$	$1.2 \pm 0.1$ (1.1 1.6)	$1.5 \pm 0.4$	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$		Male	(0.3-1.0) 1.0 + 0.5	(1.0-1.7) 1.0 + 0.5	(0.7 - 1.4) 09 + 04	$(0.9 \pm 0.3)$	(1.1-1.0) $1.0 \pm 0.3$	(1.0-3.2) 0.8 ± 0.3	
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$		where	(0.4-1.9)	(0.4-1.9)	(0.4 - 1.6)	$(0.5 \pm 0.5)$	(05-15)	(0.4 - 1.3)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	% CC	Female	$2.9 \pm 0.5$	3.1 + 0.4	$3.5 \pm 0.7$	(0.0 + 1.0) 4.1 + 1.0	$3.2 \pm 0.5$	6.4 + 1.7	
$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$			(1.8–3.8)	(2.3-3.8)	(2.1-4.7)	(2.6-6.3)	(2.3-4.3)	(0.9-8.5)	
$\begin{tabular}{ c c c c c c c c c c c c c c c c c c c$		Male	$2.1 \pm 0.8$	$2.0 \pm 0.8$	$2.8 \pm 1.1$	$2.3 \pm 0.9$	$2.0 \pm 0.7$	$3.1 \pm 1.3$	
$ \begin{tabular}{ c c c c c c c c c c c c c c c c c c c$			(0.7-3.3)	(0.8-3.4)	(1.4-4.8)	(1.1-4.4)	(1.1-3.7)	(1.2-5.5)	
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	% TN	Female	$67.5 \pm 2.5$	$65.9 \pm 2.4$	$58.6 \pm 2.5$	$66.4\pm2.0$	$66.0\pm1.9$	$54.7 \pm 2.2$	
Male $63.2 \pm 1.9$ $62.6 \pm 2.1$ $52.7 \pm 1.7$ $64.7 \pm 2.2$ $64.1 \pm 2.4$ $55.1 \pm 2.3$ (59.7-66.8)         (57.9-66.0)         (50.3-56.8)         (61.5-70.0)         (59.5-69.4)         (51.0-60.0)           % CN         Female $12.2 \pm 0.5$ $12.2 \pm 0.4$ $14.9 \pm 0.7$ $12.5 \pm 0.5$ $12.6 \pm 0.5$ $14.5 \pm 0.8$			(62.0-71.2)	(61.0-69.2)	(53.5-63.2)	(62.0-70.6)	(62.2-68.6)	(51.0-60.2)	
(59.7-66.8)         (57.9-66.0)         (50.3-56.8)         (61.5-70.0)         (59.5-69.4)         (51.0-60.0)           % CN         Female         12.2 + 0.5         12.2 + 0.4         14.9 + 0.7         12.5 + 0.5         12.6 + 0.5         14.5 + 0.8		Male	$63.2 \pm 1.9$	$62.6\pm2.1$	$52.7 \pm 1.7$	$64.7\pm2.2$	$64.1\pm2.4$	$55.1 \pm 2.3$	
% CN Female $12.2 + 0.5$ $12.2 + 0.4$ $14.9 + 0.7$ $12.5 + 0.5$ $12.6 + 0.5$ $14.5 + 0.8$			(59.7-66.8)	(57.9-66.0)	(50.3–56.8)	(61.5-70.0)	(59.5-69.4)	(51.0-60.0)	
	% CN	Female	$12.2 \pm 0.5$	12.2 ± 0.4	14.9 ± 0.7	$12.5 \pm 0.5$	$12.6\pm0.5$	$14.5 \pm 0.8$	

#### Table 1 (continued)

		Late molting fast			Early breeding fast			
Туре	Sex	Inner	Outer	Serum	Inner	Outer	Serum	
		(11.2–13.3)	(11.5–13.5)	(13.2–16.5)	(11.2–13.3)	(11.7–13.7)	(13.2–15.9)	
	Male	$13.6 \pm 1.0$	$13.6 \pm 1.0$	$15.4 \pm 1.2$	$13.4 \pm 0.9$	$13.4 \pm 1.2$	$16.9 \pm 1.4$	
		(11.9–15.5)	(12.2–16.3)	(13.8–18.5)	(12.1–15.3)	(11.8–15.8)	(15.5-19.9)	
α-HCH	Female	$7.0 \pm 2.0$	$5.6 \pm 1.2$	$2.7\pm0.7$	$4.8 \pm 1.3$	$4.7 \pm 1.0$	$2.4 \pm 0.8$	
		(3.1-11.7)	(2.8-8.2)	(1.0 - 4.0)	(2.6-7.5)	(2.3 - 7.4)	(1.0-4.1)	
	Male	$7.0 \pm 1.5$	$6.6 \pm 1.6$	$3.8 \pm 1.0$	$5.6 \pm 1.4$	$5.6 \pm 1.6$	$1.7 \pm 0.7$	
		(4.0-9.3)	(3.1-9.4)	(2.2-5.3)	(4.0-7.7)	(2.9-8.8)	(13: 1.0-3.8)	
β-HCH	Female	$36.2 \pm 10.8$	$35.5 \pm 9.5$	$19.2 \pm 6.5$	$26.0 \pm 9.6$	$30.5 \pm 9.3$	$14.6 \pm 4.1$	
		(18.1-60.1)	(19.0-54.1)	(7.7 - 29.8)	(18.8-55.8)	(20.0-57.2)	(7.1-25.5)	
	Male	$61.6 \pm 19.5$	$61.1 \pm 20.8$	$33.6 \pm 11.6$	$50.4 \pm 10.4$	$56.0 \pm 9.3$	$27.8 \pm 5.8$	
		(37.6-119.0)	(37.4-123.9)	(20.2-69.7)	(34.6-70.3)	(41.4-72.1)	(17.0-35.0)	
HCB	Female	$21.7 \pm 4.0$	$22.5 \pm 2.5$	$36.0 \pm 13.1$	$15.4 \pm 3.7$	$19.3 \pm 6.0$	$31.7 \pm 10.4$	
		(10.0-26.3)	(16.9-27.1)	(14.1 - 61.4)	(8.6-21.7)	(10.2 - 30.4)	(7.2-47.6)	
	Male	$31.2 \pm 6.1$	$29.7 \pm 8.1$	$61.3 \pm 23.1$	$21.7 \pm 7.9$	$25.9 \pm 9.0$	$12.5 \pm 8.4$	
		(15.9-43.2)	(14.9 - 42.0)	(15.6-86.7)	(7.5-36.3)	(12.1 - 42.9)	(12: 2.0-30.8)	
ΣPBDEs <sup>e</sup>	Female	$32.1 \pm 14.1$	$27.0 \pm 11.3$	18.2 ± 7.8	$21.2 \pm 11.6$	$24.8 \pm 10.6$	$9.1 \pm 6.1$	
		(1476.0)	(12.2-52.0)	(7.9 - 43.5)	(10.3-61.9)	(13.2-55.6)	(4.4-27.4)	
	Male	$44.9 \pm 31.6$	$43.6 \pm 30.7$	$26.1 \pm 18.8$	$59.9 \pm 68.2$	$63.0 \pm 76.4$	$28.7 \pm 33.8$	
		(18.4-118.7)	(13.2-112.8)	(11.7 - 68.7)	(13.3-264.6)	(14.8-284.7)	(5.4-135.5)	
% BDE-47	Female	$66.3 \pm 5.8$	68.6 ± 7.6	$76.6 \pm 7.4$	$64.3 \pm 7.1$	$66.9 \pm 6.3$	$73.8 \pm 11.4$	
		(55.3-79.2)	(47.1-79.3)	(58.9-88.1)	(47.2-78.6)	(56.3-78.1)	(38.8-87.4)	
	Male	$76.2 \pm 4.4$	$74.5 \pm 7.1$	84.3 ± 5.3	$70.7 \pm 5.5$	$72.4 \pm 3.5$	83.3 ± 4.3	
		(69.9-85.4)	(52.9-84.0)	(74.9-90.3)	(60.6-78.3	(65.6-77.1)	(73.7-90.1)	
% BDE-99	Female	8.7 ± 1.6	$9.9 \pm 5.7$	$6.4 \pm 5.7$	$9.7 \pm 4.7$	8.9 ± 1.6	8.5 ± 9.4	
		(4.2-13.1)	(5.0-28.6)	(0.5-25.8)	(4.2-29.8)	(5.0-12.9)	(1.2-39.1)	
	Male	$4.6 \pm 1.6$	$5.8 \pm 5.1$	$3.4 \pm 3.2$	$7.9 \pm 4.9$	$7.4 \pm 2.6$	$2.5 \pm 2.8$	
		(1.8-7.0)	(2.2 - 23.0)	(0.3-10.3)	(3.1-21.2)	(4.0-13.4)	(0.5-9.8)	
% BDE-100	Female	$10.3 \pm 1.0$	$10.1 \pm 1.2$	$7.3 \pm 1.5$	$9.8 \pm 0.9$	$10.2 \pm 1.2$	$7.1 \pm 2.0$	
		(8.5-12.9)	(7.7-13.1)	(4.3-9.9)	(7.5-11.6)	(8.6-13.1)	(3.7-11.8)	
	Male	$10.3 \pm 1.9$	$10.5 \pm 2.0$	$6.3 \pm 1.1$	$11.1 \pm 3.2$	$10.8 \pm 2.9$	$7.7 \pm 2.2$	
		(7.9-14.8)	(7.8-14.8)	(4.0-8.4)	(7.1-18.9)	(7.5-18.2)	(3.7-11.4)	
% BDE-154	Female	$10.1 \pm 3.9$	$7.4 \pm 3.2$	$6.7 \pm 2.9$	$10.8 \pm 4.2$	9.1 ± 3.8	6.3 ± 3.3	
		(3.9-18.5)	(2.6-15.0)	(2.2 - 11.9)	(3.7-19.7)	(3.0-16.9)	(1.9–13.0)	
	Male	4.7 ± 1.9	$5.2 \pm 1.6$	$2.7 \pm 1.4$	$5.5 \pm 1.9$	$5.1 \pm 1.8$	$3.6 \pm 2.1$	
		(1.4-8.5)	(2.3 - 7.9)	(1.0-6.0)	(2.2-9.3)	(1.9-7.9)	(1.2 - 10.0)	
6-MeO-BDE 47	Female	$2.6 \pm 0.6$	$1.9 \pm 0.4$	$0.8 \pm 0.7$	3.1 ± 0.8	$2.4 \pm 0.7$	$1.0 \pm 0.5$	
		(1.3-4.0)	(1.2-2.9)	(23: 0.2-2.2)	(1.5-4.5)	(1.4-4.4)	(23: 0.2-1.9)	
	Male	$1.7 \pm 0.9$	$1.5 \pm 0.6$	$0.2 \pm 0.7$	$2.2 \pm 1.4$	$1.5 \pm 0.8$	$0.1 \pm 0.6$	
		(0.6-4.0)	(0.4 - 2.8)	(7: 0.2 - 0.8)	(0.6-5.3)	(0.4 - 3.4)	(14.02 - 19)	

<sup>a</sup> The number of samples above the detection limit for a given compound is only presented if the compound was detected in less than 100% of samples.

<sup>b</sup>  $\Sigma$ DDTs are the sum of *p*,*p*'-DDD, *p*,*p*'-DDE, *p*,*p*'-DDT.

<sup>c</sup> ΣPCBs are the sum of 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).

<sup>d</sup> ΣCHLs are the sum of OxC, TC, CC, TN, and CN.

concentration in blubber and an overall percent lipid of the blubber. Using the percent lipid for full thickness blubber, and both morphometric and ultrasound measurements, we estimated blubber lipid mass for each seal using the steps described in Schwarz et al. (2015). The overall lipid-normalized concentration of each POP compound in blubber was then multiplied by the lipid mass of each seal to obtain a pre- and post-foraging trip blubber burden (mg POP compound in blubber per seal). In order to compare POP bioaccumulation among clusters of seals with varying departure mass and mass gained while foraging, we calculated a burden of POP compound gained per kg of blubber lipid gained over the foraging trip.

#### 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  $3 \times$  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. Animal movement and behavior

To determine if contaminant bioaccumulation varied with foraging behavior, including diving behavior, geography, and stable isotopes (Objective 4), we used a combination of satellite transmitters and time-depth recorders on adult female seals. We used five-minute epoxy to attach satellite transmitters (Wildlife Computers, Bellevue WA: MK-10 AF, SPOT4 and SPOT5) and time-depth recorders (Wildlife Computers, Bellevue WA: MK9 or MK10) to seals at the end of the fasting period on land prior to departure. We recovered instruments when seals returned to land at the end of a foraging trip. One timedepth recorder failed to record accurate information; therefore at-sea foraging behavior in relation to contaminants was restricted to 23 females. We used standard filtering techniques and protocols (Robinson et al., 2012), georeferenced all dives, and interpolated the satellite track to three locations per day (one location every eight hours). From the georeferenced dives and the interpolated satellite tracks, we quantified a set of eight diving and geographic variables: median day foraging dive depth, median night foraging dive depth, 90th percentile depth of day foraging dives, 90th percentile depth of night foraging dives, percent of benthic dives, the average of a dimensionless dive index to quantify proportional use of the water column (dividing the maximum depth of each dive by the bathymetry, obtained from the ETOPO1 1 Arc-Minute Global Relief Model (Amante and Eakins, 2009)), maximum latitude reached during the trip, and median distance to the continental shelf.

We used the eight quantified geographic and diving variables in conjunction with  $\delta^{13}$ C and  $\delta^{15}$ N from elephant seal red blood cells to identify clusters of seal behavior, using a combination of principal components analysis (PCA) followed by hierarchical cluster analysis with the FactoMineR package in R (Husson et al., 2014). This method produces unrotated factors and we used the eigenvalue threshold of 1.0 to retain principal components, which were then input as variables into the cluster analysis. Clusters were identified based on intracluster inertia (using Euclidean distance), although we set the minimum number of clusters at three to achieve finer resolution of groups, based on previous research with a larger number of individuals (Peterson et al., 2015).

#### 2.5. Statistical analysis

For Objective 1, we used linear mixed effects models to test for differences in concentrations of POPs (natural log transformed to meet the assumptions of normality and homogenous variance) in inner blubber, outer blubber, and serum between males and females. We ran mixed effects models separately for all POP compounds at each of two sampling periods (pre-foraging/late molting and post-foraging/early breeding). In each model, we included sex, tissue type, and an interaction between sex and tissue type as fixed effects, with individual seal as a random effect nested within sex. If the interaction was significant, we conducted three contrasts to compare concentrations between males and females in inner blubber, outer blubber, and serum. For Objective 2, we examined correlations among contaminant concentrations at the end of the foraging trip using Pearson's product moment correlation. For correlation analysis, when pairs of concentrations violated the assumption of bivariate normality, both concentrations were natural log-transformed. If this transformation was insufficient to obtain bivariate normality, we used a non-parametric Spearman's rank correlation. We examined the relationship between contaminants separately for each sex because exploratory regression analysis revealed numerous cases where there was either a significant difference in slope or intercept between the sexes. We calculated  $R^2$  to describe the amount of variation statistically explained by each association of compounds. For Objective 3, we used paired t-tests to compare pre-foraging to postforaging contaminant concentrations (for serum, outer blubber, and inner blubber) and blubber burdens in females. For males, we used unpaired t-tests to compare pre-foraging concentrations to post-foraging concentrations. In addition, we used paired t-tests to examine differences in contaminant concentrations (inner blubber, outer blubber, and serum) of four males across their breeding-season fast. Finally, for Objective 4, we used ANOVA to test for differences among the identified clusters of seals for 1) blubber burden gain scaled by lipid gain (mg contaminant gained per kg lipid gained), 2) contaminant concentrations (inner and outer blubber), and 3) the ratio of the concentrations (inner and outer blubber) of the two most prevalent compounds,  $\Sigma$ PCBs and  $\Sigma$ DDTs. We initially used Analysis of Covariance (ANCOVA) with the percent of adipose tissue as a covariate (main effect) for the analysis of concentrations in inner blubber; however, the inclusion of the percent of adipose tissue for early breeding samples was not significant for any contaminant compound (p > 0.279) and was therefore removed from the final analyses. Data were transformed (natural log or reciprocal (1/x) transformation) if necessary to meet the assumptions of normality and homogeneous variance. If clusters were significantly different, we conducted post-hoc pairwise tests with a Holm correction for repeated analysis. All statistical analyses were conducted either in the statistical package R, version 3.1.0 (R Development Core Team, 2012) or JMP Pro 11, and statistical significance was set at  $\alpha = 0.05$ .

#### 3. Results

#### 3.1. Persistent organic pollutant concentrations

All elephant seals had detectable lipid-normalized concentrations of  $\Sigma$ PCBs,  $\Sigma$ DDTs,  $\Sigma$ CHLs and  $\Sigma$ PBDEs in blubber and serum samples before and after their long foraging trip (Table 1). Additionally, HCB,  $\alpha$ -HCH,  $\beta$ -HCH, and 6-MeO-BDE 47 were detected in all blubber samples (Table 1), whereas  $\gamma$ -HCH was only detected in 6% of both inner and outer blubber samples and 2-MeO-BDE 68 was only detected in 22% of inner blubber, but not detected in outer blubber (both compounds only detected during early breeding). Fewer compounds had 100% detectability in serum samples (Table 1). The specific compounds with the highest mean concentrations in serum and blubber of northern elephant seal males and females were  $\Sigma$ DDTs, of which >98% was the DDT metabolite, *p*,*p*'-DDE (Table 1). The specificy (Table 1).

Overall, for the majority of POP compounds pre- and post-foraging trip, male elephant seals had significantly greater concentrations in blubber (inner and outer) and serum than females (see Supplementary Tables 1a-b for the complete reporting of all test statistics). The naturally produced 6-MeO-BDE 47 was the only compound with greater concentrations in both the blubber and serum of females than in males, which was observed at both time periods ( $p \le 0.002$ ). Pre- and postforaging trip, concentrations of  $\Sigma$ DDTs,  $\Sigma$ PCBs,  $\Sigma$ CHLs, and  $\beta$ -HCH were greater in males than females in blubber and serum ( $p \le 0.026$ ). Pre-foraging trip, there was a marginally non-significant difference in the mean concentration of  $\Sigma$ PBDEs in males and females ( $F_{1,36} = 4.0$ , p = 0.054), and post-foraging trip  $\Sigma$ PBDEs were significantly greater in the blubber and serum of males than females ( $p \le 0.008$ ). Pre- and post-foraging trip concentrations of  $\alpha$ -HCH were not significantly different between males and females in blubber ( $p \ge 0.088$ ). In contrast, both pre- and post-foraging trip concentrations of  $\alpha$ -HCH in serum were greater in males than females ( $p \le 0.001$ ). Pre-foraging trip concentrations of HCB in blubber and serum were significantly different between males and females ( $p \le 0.020$ ). However, in contrast with other compounds, post-foraging trip concentrations of HCB were greater for females in serum (p < 0.001), while concentrations were significantly greater in males in inner blubber (t = -2.0, p = 0.050) and nearly significant in outer blubber (t = -1.8, p = 0.068).

#### 3.2. Correlations between contaminants

We found significant, positive relationships between concentrations of most, but not all, compounds in male and female elephant seals in blubber and serum after the foraging trip, upon arrival to the colony at the start of the breeding season (Supplementary Tables 2a-c). In males and females,  $\sum$  DDTs, and  $\sum$  PBDEs had significant, positive correlations (p < 0.05, Supplementary Tables 2a–c) in inner blubber (males:  $R^2 = 0.83$ ; females:  $R^2 = 0.85$ ), outer blubber (males:  $R^2 =$ 0.77, females:  $R^2 = 0.88$ ) and serum (males:  $R^2 = 0.77$ ; females:  $R^2 =$ 0.59). Similarly, correlations between  $\sum$  DDTs and  $\sum$  PCBs were strong for males in all tissue types ( $R^2 \ge 0.83$ , p < 0.001). However, in females, less variation was statistically explained by the association of these compounds in inner blubber ( $R^2 = 0.37$ , p = 0.001) and serum ( $R^2 =$ 0.46, p < 0.001), and there was no significant relationship in outer blubber ( $R^2 = 0.11$ , p = 0.115; Supplementary Tables 2a–c). Additionally, in both males and females,  $\sum$  CHLs had significant, positive correlations with  $\sum$  DDTs and  $\sum$  PCBs in inner blubber ( $R^2 \ge 0.44$ ), outer blubber  $(R^2 \ge 0.40)$ , and serum  $(R^2 \ge 0.42)$ . In females,  $\sum$  CHLs were significantly and positively correlated with  $\sum$  PBDEs in inner blubber, outer blubber, and serum ( $R^2 \ge 0.17$ ,  $p \le 0.044$ ), whereas these two compounds in males were only significantly correlated in the inner blubber (rho = 0.53, p = 0.047) and not in the outer blubber or serum. In males,  $\sum$  PCBs were significantly correlated with  $\sum$  PBDEs in inner blubber ( $R^2 = 0.61$ , p < 0.001), outer blubber ( $R^2 = 0.56$ , p = 0.001)

and serum ( $R^2 = 0.49$ , p = 0.004), but in females were only significantly correlated in inner blubber ( $R^2 = 0.18$ , p = 0.036) and serum ( $R^2 =$ 0.18, p = 0.041), and not in outer blubber ( $R^2 = 0.04$ , p = 0.345) (Supplementary Tables 2a–c). For males and females,  $\alpha$ -HCH and HCB were positively correlated in all tissue types ( $R^2 \ge 0.30$ , p < 0.021). In females but not males,  $\beta$ -HCH was positively correlated with  $\sum$  CHLs in blubber and serum ( $R^2 \ge 0.18$ ,  $p \le 0.037$ ; Supplementary Tables 2a–c)

#### 3.3. Change in contaminant concentrations and blubber burdens

#### 3.3.1. Female concentrations

Using paired pre- and post-foraging trip tissue samples from adult females, we detected significant changes in almost all contaminant concentrations in blubber (inner and outer) and serum across the foraging trip (Fig. 2; Table 1). Additionally, the percent of lipid in inner blubber and serum significantly increased across the foraging trip (t = 4.67, df = 23, p < 0.001; t = 2.18, df = 23, p = 0.040, respectively; Table 1), but the percent of lipid in outer blubber did not significantly change between those time periods (t = 0.32, df = 23, p = 0.753; Table 1). The majority of POP concentrations in elephant seal females decreased across the foraging trip, with 6-MeO-BDE 47 the only contaminant to increase in concentration within the blubber while seals were foraging (Supplementary Table 3).

Concentrations of  $\sum$  DDTs,  $\sum$  PCBs,  $\sum$  CHLs,  $\sum$  PBDEs, and  $\beta$ -HCH significantly decreased across the long foraging trip in inner blubber, outer blubber, and serum (p < 0.044; Table 1; Supplementary Table 3). Concentrations of HCB and  $\alpha$ -HCH decreased in inner blubber and outer blubber (p < 0.023) but not in serum (p > 0.160; Supplementary Table 3). Concentrations of 6-MeO-BDE 47 increased within inner and outer blubber (p < 0.001) but serum concentrations did not significantly change (Supplementary Table 3). Due to the low detectability of  $\gamma$ -HCH and 2-MeO-BDE 68, we did not test for changes in concentration of these compounds across the foraging trip.

#### 3.3.2. Male concentrations

Using unpaired samples for adult males, we detected very few changes in POP concentrations across the foraging trip and we did not detect any overall differences in lipid percent; however, the variability in male contaminant concentrations was substantially higher than that observed in females (Table 1; Supplementary Table 4). In male serum, only concentrations of  $\sum$  CHLs, HCB, and  $\alpha$ -HCH decreased across the foraging trip (p < 0.034; Supplementary Table 4). In blubber, changes were only observed between pre-and post-trip concentrations of HCB and  $\alpha$ -HCH in inner blubber (p < 0.015; Table 1; Supplementary Table 4). The only contaminant observed in males to increase in concentration across the foraging trip was 6-MeO-BDE 47 measured in serum (t = -3.10, df = 15.6, p = 0.007; Table 1). Conversely, when paired tissue samples from four males were examined across the breeding fast (48-53 days of fasting between samples; Supplementary Table 5), concentrations of  $\sum$  DDTs,  $\sum$  PCBs,  $\sum$  CHLs, HCB, and  $\beta$ -HCH significantly increased in inner blubber, outer blubber, and serum (p < 0.042; Supplementary Tables 5–6). Concentrations of  $\sum$  PBDEs increased in serum across the breeding fast (t = 5.0, p = 0.016), but did not change in blubber ( $p \ge 0.087$ ; Supplementary Table 5). Similarly, concentrations of  $\alpha$ -HCH increased in serum across the breeding fast (t = 5.7, p = 0.011) but did not change in blubber. Concentrations of 6-MeO-BDE 47 did not significantly change across the breeding fast in blubber or serum (Supplementary Table 5).

#### 3.3.3. Female blubber burdens

In adult females, blubber burdens of  $\sum$  PCBs,  $\sum$  DDTs,  $\sum$  CHLs,  $\sum$  PBDEs, HCB,  $\alpha$ -HCH,  $\beta$ -HCH, and 6-MeO-BDE 47 significantly increased across the foraging trip, based on paired pre- and postforaging trip blubber samples (p < 0.001; Fig. 2; Table 2). Elephant seal females gained a mean  $\pm$  SD (range) of 25.1  $\pm$  13.4 mg (4.1– 56.3 mg)  $\sum$  PCBs, 40.1  $\pm$  18.4 mg (13.9–82.9 mg)  $\sum$  DDTs, 7.8  $\pm$ 4.0 mg (2.0–15.9 mg)  $\sum$  CHLs, and 0.7  $\pm$  0.4 mg (0.2–1.5 mg)  $\sum$  PBDEs between sampling periods, over the long foraging trip (Table 2). These increases resulted in female elephant seals arriving to the colony with approximately 60.5  $\pm$  16.5 mg  $\sum$  PCBs, 103.2  $\pm$ 31.0 mg  $\sum$  DDTs, 19.8  $\pm$  4.8 mg  $\sum$  CHLs, and 1.8  $\pm$  0.7 mg  $\sum$  PBDEs in their adipose tissue (Table 2).

#### 3.4. Geography and foraging behavior

Adult female northern elephant seals (N = 23) were at sea for a mean  $\pm$  SD (range) of 224  $\pm$  4 days (217–233 days), with the distal location of the trip located 3364  $\pm$  1062 km (range 1126–5206 km) from the Año Nuevo colony (Fig. 3). The median distance of individual seals from the continental shelf ranged from 47 to 1204 km, with maximum distances from the continental shelf of 188-1646 km. The maximum latitude reached by individuals ranged from 42.0 to 59.2° N. Stable carbon and nitrogen isotope values quantified in red blood cells were  $-19.5 \pm$ 0.3% (-20.1 to -18.8%) for  $\delta^{13}$ C and  $14.4 \pm 0.9\%$  (13.4 to 16.5%) for  $\delta^{15}$ N. The median depth of foraging dives for individual seals was 633  $\pm$ 51 m (539–760 m) during the day and 486  $\pm$  22 m (440–532 m) during the night. The 90th percentile of foraging dives for individual seals was  $776 \pm 93$  m (641–938 m) during the day and  $653 \pm 43$  (578–740) during the night. Individual seals only had 4.8  $\pm$  3.5% (10.7–35.5%) of total dives identified as benthic with an overall mean dive index (quantifying proportional use of the water column) of  $15.5 \pm 6.3\%$  (10.7–35.5%).

Adult female elephant seals were clustered into three groups, the preset minimum number of groups, based on their foraging behavior, including geographic location variables, diving behavior variables, and both carbon and nitrogen isotopes (Fig. 3). The three clusters could be broadly classified into a northerly cluster (N = 4), a cluster enriched in <sup>13</sup>C and <sup>15</sup>N isotopes (hereafter the isotopes cluster) (N = 3), and an offshore cluster (N = 16). Based on the variables important in cluster creation, seals in the northerly cluster were characterized by a more northerly maximum latitude (57.9 ± 1.7° N), used a greater proportion of the water column (dive index = 24.8 ± 8.1%), had shallower median day and night foraging dive depths (556 ± 22 m and 459 ± 13 m,



**Fig. 2.** Change in POP concentrations and burdens over a foraging trip. Paired pre- and post-foraging trip samples for 24 adult female elephant seals show the change in concentration (full blubber cores) and the change in blubber burden (mg) of three anthropogenic and one naturally produced (6-MeO-BDE 47) POPs across an approximately seven month long foraging trip. All eight compound groups measured in this study significantly decreased in concentration across the foraging trip, except for 6-MeO-BDE 47. Burden, however, increased for all compounds. Note that the axes labels are not the same for all compounds. All eight compounds are shown in Supplementary Fig. 1.

#### Table 2

Mean  $\pm$  SD (range) of the estimated blubber burden (mg) pre- and post-foraging trip in adult female northern elephant seals (*Mirounga angustirostris*), the mean burden of contaminant gained over the approximately seven month long, post-molting foraging trip, and the percent increase in the blubber burden over the foraging trip. Statistical results are for paired t-tests comparing the change in the burden of POPs in the blubber between pre-and post-foraging trip (N = 24).

Contaminant type	Pre-foraging burden (mg)	Post-foraging burden (mg)	Gain (mg)	% increase in burden	DF	t-Value	p-Value
ΣDDTs <sup>a</sup>	$63.1\pm25.7$	$103.2 \pm 31.0$	$40.1\pm18.4$	$71.2\pm37.2$	23	- 10.67	< 0.001
	(33.7-146.4)	(59.3-179.4)	(13.9-82.9)	(17.4–162.2)			
ΣPCBs <sup>b</sup>	$35.3 \pm 7.6$	$60.5 \pm 16.5$	$25.1\pm13.4$	$73.3 \pm 37.3$	23	-9.22	< 0.001
	(21.9-51.0)	(40.2-106.1)	(4.1-56.3)	(9.2-173.4)			
ΣCHLs <sup>c</sup>	$12.0 \pm 2.8$	$19.8 \pm 4.8$	$7.8\pm4.0$	$69.1 \pm 39.1$	23	-9.63	< 0.001
	(8.0-17.0)	(12.1-32.9)	(2.0-15.9)	(12.2-185.3)			
α-HCH	$0.2 \pm 0.1$	$0.4 \pm 0.1$	$0.1\pm0.1$	$72.1 \pm 65.4$	23	-6.48	< 0.001
	(0.1-0.4)	(0.2-0.6)	(-0.1-0.4)	(-21.2-214.7)			
β-HCH	$1.4 \pm 0.6$	$2.2 \pm 0.8$	$0.8\pm0.4$	$68.4 \pm 38.7$	23	-10.40	< 0.001
	(0.6-3.0)	(1.4-4.6)	(0.3-1.6)	(16.7-190.9)			
HCB	$0.9 \pm 0.2$	$1.4 \pm 0.4$	$0.5\pm0.4$	$72.1 \pm 67.6$	23	-6.33	< 0.001
	(0.5-1.3)	(0.6-2.4)	(-0.2 - 1.5)	(-21.3-288.3)			
ΣPBDEs <sup>d</sup>	$1.1 \pm 0.6$	$1.8 \pm 0.7$	$0.7\pm0.4$	$69.8 \pm 46.5$	23	-8.81	< 0.001
	(0.5-3.1)	(0.9–3.4)	(0.2-1.5)	(12.4–182.0)			
6-MeO-BDE 47	$0.1 \pm < 0.1$	$0.3 \pm 0.1$	$0.2\pm0.1$	$214 \pm 108.4$	23	-11.52	< 0.001
	(<0.1-0.1)	(0.1-0.4)	(<0.1-0.3)	(40.5-525.0)			

<sup>a</sup> ΣDDTs are the sum of *p*,*p*'-DDD, *p*,*p*'-DDE, *p*,*p*'-DDT.

<sup>b</sup> ΣPCBs are the sum of 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).

<sup>c</sup> ΣCHLs are the sum of OxC, TC, CC, TN, and CN.

<sup>d</sup> ΣPBDEs are the sum of 7 PBDE congeners (IUPAC numbers: BDE 28, 47, 99, 100, 153, 154, and 183).

respectively), had shallower 90th percentile of dive depths during the day (687 ± 31 m), and had a smaller median distance to the continental shelf (197 ± 96 km) than the overall mean value for all seals. Seals in the isotopes cluster were more enriched in <sup>13</sup>C ( $-18.9 \pm 0.04\%$ ), more enriched in <sup>15</sup>N (16.4 ± 0.04‰), and had a deeper median depth of foraging dives at night than the overall mean for all seals. The seals in the offshore cluster had an average maximum latitude of 47.8 ± 3.7° N, used a smaller proportion of the water column (dive index = 12.2 ± 1.2%), had a smaller proportion of benthic dives (3.6 ± 1.6%), had a greater median distance to the continental shelf (880 ± 225 km), and had a deeper median and 90th percentile of foraging dive depths during the day (652 ± 34 m and 812 ± 81 m, respectively) than the overall mean for all seals.

Both the burden of contaminant (mg) gained per kg lipid gained over the foraging trip and contaminant concentration (inner and outer blubber) differed among clusters for some compounds (Fig. 3). The mg POP gained per kg lipid gained over the foraging trip differed among clusters for  $\sum$  DDTs,  $\sum$  PBDEs, and 6-MeO-BDE 47 ( $F_{2,20} = 7.14$ , p = 0.005;  $F_{2,20} = 6.47$ , p = 0.007;  $F_{2,20} = 21.35$ , p < 0.001, respectively; Fig. 3). Pairwise tests revealed that the isotope cluster had greater  $\sum$  DDTs and  $\sum$  PBDEs gained per kg lipid gained than the offshore cluster (p =0.004 and p = 0.008), but was not significantly different from the northerly cluster (p = 0.088 and p = 0.221), and the northerly cluster was not significantly different than the offshore cluster (p = 0.222 and p = 0.221; Fig. 3). The mg gain of 6-MeO-BDE 47 per kg lipid gained was lower for the northerly cluster than the isotope or offshore clusters (p < 0.001), which were not different from each other (p = 0.476).

Concentrations of  $\sum$  DDTs,  $\sum$  PBDEs, and 6-MeO-BDE 47 differed among clusters in both inner blubber ( $F_{2,20} = 7.77$ , p = 0.003;  $F_{2,20} = 8.4$ , p = 0.002;  $F_{2,20} = 15.23$ , p < 0.001, respectively; Fig. 3) and outer blubber ( $F_{2,20} = 10.67$ , p < 0.001;  $F_{2,20} = 13.60$ , p < 0.001;  $F_{2,20} = 5.03$ , p = 0.017;  $F_{2,20} = 10.35$ , p < 0.001, respectively; Fig. 3), similar to patterns demonstrated using the burden of these four contaminants gained per kg lipid gained. Pairwise tests revealed similar differences as described above between clusters for inner and outer blubber for all four of these contaminants (Supplementary Materials). There was no detectable difference in the mg contaminant gained per kg lipid gained over the foraging trip for  $\sum$  PCBs,  $\sum$  CHLs, HCB,  $\alpha$ -HCH, or  $\beta$ -HCH among the clusters of elephant seals.

The ratio between the two most prevalent contaminant compounds,  $\sum$  DDTs and  $\sum$  PCBs, was significantly different between clusters of elephant seal females for inner and outer blubber ( $F_{2,20} = 11.6$ ,

p < 0.001 and  $F_{2,20}$  = 12.0, p < 0.001, respectively). Ratios of  $\sum$  DDTs to  $\sum$  PCBs ranged from 1.0 to 3.4 in inner blubber and 1.2–3.8 in outer blubber. For both inner blubber and outer blubber, the ratios were significantly greater in the isotopes cluster than the northerly cluster (p < 0.026) and the offshore cluster (p < 0.001), which were not significantly different from each other (p < 0.216) (Fig. 4). While we could not assign male elephant seals to geographic foraging regions because they were not satellite tracked, males had  $\delta^{13}$ C values ranging from − 19.2 to − 16.5‰ and had ratios of  $\sum$  DDTs to  $\sum$  PCBs ranging from 1.1 to 3.2 in inner blubber and 1.3–3.8 in outer blubber, overlapping with the ratios observed in females (Fig. 4).

#### 4. Discussion

Adult northern elephant seals revealed variation in bioaccumulation of POPs with sex and foraging behavior. All females and males had measurable concentrations of DDTs, PCBs, CHLs, and PBDEs in blubber and blood, demonstrating the legacy of these contaminants across an extensive expanse of the mesopelagic northeast Pacific Ocean.

For adult female and male elephant seals, the predominant POP compounds in blubber and serum were  $\sum$  DDTs, followed by  $\sum$  PCBs, which is in contrast with free-ranging adults from several other northeastern Pacific marine mammal species. Although some individual elephant seals had ratios of  $\sum$  DDTs to  $\sum$  PCBs close to 1, no elephant seals had greater concentrations of  $\sum$  PCBs than  $\sum$  DDTs (Fig. 4). In contrast, concentrations of  $\sum$  PCBs were greater than  $\sum$  DDTs in the blubber of Steller sea lions (Eumetopias jubatus) from Alaska and monk seals (Neomonachus schauinslandi) from the Hawaiian Islands (Lion et al., 1996; Lopez et al., 2012; Willcox et al., 2004). Additionally, female resident killer whales (Orcinus orca) from the Kenai Fjords in Alaska (Ylitalo et al., 2001) and belugas (Delphinapterus leucas) from Cook Inlet, Alaska (Hoguet et al., 2013) also had greater concentrations of  $\sum$  PCBs than  $\sum$  DDTs in blubber. In blubber of male harbor seals from the Gulf of Alaska, concentrations of  $\sum$  PCBs were greater than  $\sum$  DDTs; however, the relationship between these two compounds was less clear for females (Wang et al., 2007). In contrast to these species but similar to northern elephant seals from the present and previous studies (Debier et al., 2012), concentrations of  $\sum$  DDTs were greater than  $\sum$  PCBs in the blubber of bottlenose dolphins (*Tursiops* truncatus) in San Diego Bay (Reddy et al., 2001) and California sea lions (Zalophus californianus) found along the California coast (Blasius and Goodmanlowe, 2008; Greig et al., 2007; Kannan et al., 2004;



Fig. 3. Clusters of seal behavior along with corresponding POP concentrations and blubber burdens. Northern elephant seals (*Mirounga angustirostris*) were clustered into three groups based on geographic variables, diving variables, and stable isotope ratios ( $\delta^{13}$ C and  $\delta^{15}$ N). The clusters in the map and the boxplots have corresponding colors. The top row of boxplots shows clusters by the burden of contaminant (mg) gained for each kg of lipid gained between sampling periods, pre- and post-foraging trip. The bottom row shows lipid normalized contaminant concentrations in inner blubber. Letters indicate significant differences between clusters.  $\sum$  PCBs were not significantly different between clusters, but were included as a reference. Untransformed data are shown here, although, when necessary, concentrations were transformed to meet assumptions of normality and homogenous variance.

Ylitalo et al., 2005), although the only published concentrations of these compounds in adult California sea lion blubber are for stranded animals.

Geographic variability in the relative distribution of  $\sum$  DDTs to  $\sum$  PCBs in the northeastern Pacific may partially explain why elephant seals differ from more northerly- and offshore-distributed species. Sea otters (*Enhydra lutris*) sampled in Alaska had greater  $\sum$  PCBs than  $\sum$  DDTs in serum, whereas otters sampled in California had greater  $\sum$  DDTs than  $\sum$  PCBs (Jessup et al., 2010). Similarly, harbor porpoises (*Phocoena phocoena*) and humpback whales (*Megaptera novaeangliae*) sampled along the west coast of North America demonstrated

increasing concentrations of  $\sum$  PCBs relative to  $\sum$  DDTs in blubber with increasing latitude (Calambokidis and Barlow, 1991; Elfes et al., 2010). Synthesis of these studies provides strong evidence for past and current distributional gradients of  $\sum$  DDTs relative to  $\sum$  PCBs in marine predators across the northeastern Pacific Ocean. In the present study, we observed a lower ratio of  $\sum$  DDTs to  $\sum$  PCBs for the northerly and offshore seals than the seals foraging closer to the coast and further south (Fig. 4). However, despite female elephant seals foraging a significant distance from shore across a large latitudinal range of the northeastern Pacific, the relationship of  $\sum$  DDTs to  $\sum$  PCBs for all seals



**Fig. 4.** Ratios of  $\sum$  DDTs to  $\sum$  PCBs in elephant seals. Ratios of  $\sum$  DDTs to  $\sum$  PCBs in relation to  $\delta^{13}$ C values from adult female and male (sub-adult 4 and adult age classes) northern elephant seals (*Mirounga angustirostris*) sampled at the Año Nuevo colony upon returning from a foraging trip. Females are grouped based on a cluster analysis, using diving behavior, geography and stable C and N isotopes. Males were not satellite tracked.

appeared less similar to their northerly or highly offshore (i.e., monk seals from Hawaii) pinniped counterparts and more similar to marine mammals from southerly latitudes in the North Pacific.

POPs have not previously been quantified in free-ranging adult male northern elephant seals, and the two most prevalent POP compounds were  $\sum$  DDTs and  $\sum$  PCBs. Mean concentrations of  $\sum$  DDTs in males from the present study were greater than  $\sum$  DDTs in free ranging adult harbor seals (Phoca vitulina) from the Gulf of Alaska (Wang et al., 2007) and adult male Hawaiian monk seals from multiple colonies (Lopez et al., 2012; Willcox et al., 2004). In contrast, the mean concentrations of  $\sum$  PCBs in male Steller sea lions captured in British Columbia (Alava et al., 2012) and male Steller sea lions from the Bering Sea and Prince William Sound (Wang et al., 2011) were greater than the mean concentrations of  $\sum$  PCBs we observed in elephant seals. It should be noted, however, that direct comparison of  $\sum$  PCBs is difficult due to the variability in congeners quantified in each study. Mean  $\sum$  PCBs in elephant seal males were similar to male monk seals from the main Hawaiian Islands (Lopez et al., 2012), although other colonies of monk seals (Midway Island and French Frigate Shoals) demonstrated increased localized contamination of PCBs (Willcox et al., 2004; Ylitalo et al., 2008).

We observed greater concentrations of most POP compounds in males versus females, similar to previous studies on other pinniped species (Barron et al., 2003; Lopez et al., 2012; Wang et al., 2007). This is not surprising, given that females offload a large proportion of their contaminant burden to their offspring, with some transfer occurring during gestation but the primary transfer occurring through lactation (Debier et al., 2012; Frouin et al., 2012; Vanden Berghe et al., 2010). For northern elephant seals, most females give birth to a pup every year and offload a significant proportion of their contaminant burden to their pup (Debier et al., 2012; Peterson et al., 2014). In addition to reproductive explanations for differences in contaminant concentrations between the females and males, male elephant seals also differ from the majority of female elephant seals in foraging location and diving behavior, as well as potential differences in prey selection (Le Boeuf et al., 2000). The only compound found in greater concentrations in the blubber of females was the naturally produced 6-MeO-BDE 47, which may be

attributed to the offshore nature of female foraging in contrast with the coastal nature of male foraging (Le Boeuf et al., 2000; Simmons et al., 2007). This hypothesis was additionally supported within females, with the greatest concentrations of 6-MeO-BDE 47 observed in the cluster of females identified as foraging furthest away from the continental shelf (Fig. 3). Limited information exists regarding sources or accumulation patterns of MeO-BDEs in the northeastern Pacific (Alonso et al., 2014). It is unlikely that differences in age between males and female elephant seals caused the differences we observed, as reproductive aged males likely overlapped in age with the older females we sampled (Condit et al., 2014).

The difference in correlations between females and males among compounds may indicate either differential geographic distributions or varying rates of metabolism or reproductive transfer among compounds. For example, correlations  $\sum$  DDTs and  $\sum$  PCBs in blubber and serum were weaker, especially for females, than the strong correlations observed for both females and males between  $\sum$  DDTs and  $\sum$  PBDEs. In addition,  $\sum$  CHLs were well correlated with  $\sum$  PCBs and less so with  $\sum$  DDTs or  $\sum$  PBDEs. While there is evidence for selective maternal transfer of some POP compounds over others (Debier et al., 2003; Vanden Berghe et al., 2012) and the potential exists for differential metabolism of certain POP compounds over others, these physiological processes would likely be consistent among elephant seals (Louis et al., 2015). Thus, variability in geographic distribution of POPs or differences in foraging behavior associated with geography are more likely explanations for the patterns we observed. Long range transport potential varies among POP compounds, with PCBs (specifically, the less chlorinated congeners) more likely to reach regions farther from their initial source than either DDTs or PBDEs (Lohmann et al., 2007; Wania and Dugani, 2003). This was supported by female elephant seals from our study because they returned from a wide range of foraging areas with similar bioaccumulation of  $\sum$  PCBs, indicating ubiquity in distribution, whereas bioaccumulation of  $\sum$  DDTs and  $\sum$  PBDEs was highest in animals that foraged closer to the continental shelf (Fig. 3). In male seals, all three compounds were highly correlated, which may be attributed to the more uniform, coastal foraging behavior of males. Additionally, the timeline of use for these compounds may contribute to their environmental distribution. Whereas worldwide manufacture of PCBs ceased in the early 1990s (Breivik et al., 2002), the use of PBDEs is more recent in some regions and thus these compounds may not have yet reached more remote locations, and the use of DDT for insect control in parts of the world (including Mexico and some regions of Asia) continues or was recently phased out (Lohmann et al., 2007).

Foraging behavior influenced bioaccumulation of some POP compounds in mesopelagic foraging adult female northern elephant seals. In previous research on epipelagic foraging marine predators from the North Pacific Ocean, intraspecific variability in POP concentrations was observed in shearwaters (Calonectris leucomelas) (Ito et al., 2013), skipjack tuna (Katsuwonus pelamis) (Ueno et al., 2003), humpback whales (Elfes et al., 2010), and Hawaiian monk seals (Lopez et al., 2012), and attributed to differences in geographic location. However, the influence of foraging behavior on POP concentrations may depend on foraging characteristics beyond geography. Our cluster analysis of female seals, characterized using geography, stable isotope ratios ( $\delta^{13}$ C and  $\delta^{15}$ N), and diving behavior, showed that  $\sum$  DDTs and  $\sum$  PBDEs were greatest in the seals that foraged further south, closer to the continental shelf, and were more enriched in  $\delta^{13}$ C and  $\delta^{15}$ N (Fig. 3). Of note, one female in this cluster foraged in a geographic region overlapping with females from the offshore cluster; however, her stable isotope ratios and her  $\sum$  DDTs and  $\sum$  PBDEs were different from the offshore females, providing evidence for an interplay between the multiple components of foraging behavior that influence contaminant bioaccumulation. The seals that foraged more northerly and at shallower depths, near the Subarctic Gyre, had the lowest concentrations of 6-MeO-BDE 47, suggesting that regions in the north Pacific may vary in the production of this naturally-produced brominated compound.

Our study showed that elephant seals ingest a suite of POPs while foraging in the mesopelagic North Pacific. The influence of body condition on blubber concentrations of POPs (Debier et al., 2012; Hall et al., 2008; Myers and Atkinson, 2012) makes it challenging to use concentrations in blubber to track temporal changes in POPs in marine predators that fluctuate markedly in body condition as a consequence of annual life history events. Concentrations of POPs in previous studies on elephant seals decreased in response to improved body condition and mass gain, which often occurs during foraging (Peterson et al., 2014), and increased while fasting (Debier et al., 2012). For our study, contaminant concentrations in female elephant seals decreased over the foraging trip, but blubber burdens of all compounds increased, indicating ingestion of POPs during the foraging trip at a higher rate than they were being metabolically processed or transferred to a developing fetus.

We provided an important comparison between POP tissue concentrations and blubber burdens in a free ranging marine predator across the duration of an approximately seven month foraging trip. Concentrations of POP compounds in blubber (inner and outer) decreased while animals gained mass during their foraging trip, with the exception of 6-MeO-BDE 47, whereas the absolute burden of all POP compounds in blubber increased across the foraging trip. Our results demonstrate that regardless of any differences in foraging characteristics, all female elephant seals gained POPs while foraging. To account for varying animal size and foraging success, we did not directly compare blubber burden with clusters of seal foraging behavior, but instead used the ratio of POP burden gained to the lipid mass gained while foraging. Importantly, the differences among clusters of elephant seal foraging behavior for POPs were consistent regardless of whether we examined concentrations in blubber (inner or outer) at the end of the foraging trip or the burden of contaminant gained per kg of lipid gained. These observations suggest that, in the case of elephant seals sampled at the end of a foraging trip, contaminant concentrations alone can be used to indicate the influence of recent foraging behavior on POP exposure.

As mesopelagic predators, northern elephant seals utilize a large portion of the northeastern Pacific Ocean, and all individuals demonstrated POP bioaccumulation. The inability of males to offload POPs to offspring possibly coupled with differences in foraging behavior from females resulted in greater bioaccumulation of POPs in the males, as seen in other marine mammal species. Tissue correlations and individual burdens suggest uneven distribution of major POP compounds in marine food webs across the foraging range of northern elephant seals. Our calculation of blubber burdens for adult females allowed us to distinguish ingestion of POPs over the course of an extensive foraging trip, despite mass dilution effects on tissue concentrations. We showed that the deep-ocean foraging strategy of elephant seals does not eliminate their risk for POP exposure and further indicates that legacy POPs can reach mesopelagic food webs.

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#### Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx. doi.org/10.1016/j.scitotenv.2015.06.097.

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