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NOTE

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Partitioning of persistent organic pollutants between blubber and blood in killer whales (*Orcinus orca*)

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Persistent organic pollutants (POPs) remain in the environment for extended periods of time and can have long lasting deleterious physiological effects on organisms (Ashraf, 2017). Progressive increases in tissue POP concentrations as a result of direct environmental uptake, such as prey intake, compounded by the inability to metabolize many contaminants, can further disrupt physiological systems (Boon et al., 1994; Lohmann et al., 2007). Top predators, including many marine mammals, experience considerable bioaccumulation of POPs as a result of their high trophic level status and long life expectancy (Gray, 2002; Rowe, 2008). Previous research has shown that chronic exposure to POPs can have detrimental effects on reproduction, growth, and immune and endocrine function in marine mammals (Debier et al., 2005; Desforges et al., 2016; Tanabe et al., 1994).

Many lipophilic POPs are recalcitrant to metabolism and consequently persist in blubber tissue, the predominant lipid storage site in marine mammals (Montie et al., 2008). As such, blubber contains a high contaminant load and is

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therefore the primary sampling substrate for estimating overall contaminant burdens (Tanabe et al., 1981). POPs, however, can be found in a variety of different tissues, including blood, which acts as a lesser repository and mechanism of transport of lipophilic and nonlipophilic contaminants throughout the body (Yordy et al., 2010). While blubber is the primary sampling site for POPs, blood may be a better indicator of bioavailable contaminant concentrations in marine mammals (Yordy et al., 2010). In other words, because blood is in direct contact with target tissues, contaminants in blood are free to elicit adverse effects on endocrine, developmental, immunological, neurological, and reproductive systems (Desforges et al., 2016; Fair et al., 2010; Yordy et al. 2010).

Previous research on marine mammals has investigated patterns of contaminant distribution in the body, but only a few studies have specifically explored the relationship between blubber and blood concentrations (Balmer et al., 2018; Lydersen et al., 2002; Myers & Atkinson, 2012; Peterson et al., 2016; Reddy et al., 1998; Ylitalo et al., 2008; Yordy et al., 2010). Earlier studies on common bottlenose dolphins (*Tursiops truncatus*) and Hawaiian monk seals (*Neomonachus schauinslandi*) demonstrated that blubber concentrations of polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane compounds (DDTs), polybrominated diphenyl ethers (PBDEs), chlordanes (CHLDs), and other contaminants positively correlate with concentrations in the blood (Balmer et al., 2018; Lopez et al., 2012; Reddy et al., 1998; Yordy et al., 2010). This suggests that both blood and blubber can be used to evaluate contaminant burdens in marine mammals. This may be especially useful for studies where animal handling constraints may limit researchers to a specific sample type (Reddy et al., 1998). However, to assess whether this relationship between circulating blood and blubber contaminant levels is ubiquitous for marine mammals, it is essential to assess these relationships in other species.

Killer whales (*Orcinus orca*) are considered to be one of the most contaminated species in the world due to their top trophic level status (Ross et al., 2000). While some populations are already considered endangered or threatened (Fisheries and Oceans Canada, 2011; Krahn et al., 2004), many more may potentially experience declines due to high contaminant burdens (Desforges et al., 2018). This underscores the need to fully understand contaminant concentrations and distributions in the body in order to inform future conservation management strategies (Hickie et al., 2007). Previous research has focused on characterizing the overall toxicant burden across different age classes and geographic locations in both wild and captive killer whale populations (Bennett et al., 2009; Formigaro et al., 2014; Jepson et al., 2016; Krahn et al., 2009; Lawson et al., 2020; Mongillo et al., 2012; Ross, 2006; Ylitalo et al., 2001). Direct comparisons between blubber and serum contaminant levels, however, have not been reported thus far.

To provide preliminary insight into the relationship between blubber POP concentrations and circulating levels in killer whales, we analyzed various POP classes in matched blubber and serum samples taken from two freshly deceased killer whales. We also compared killer whale POP partitioning patterns between blubber and blood with those of bottlenose dolphins, a smaller delphinid. While the sample size of this study was constrained due to difficulties in obtaining suitable samples from large free-ranging killer whales, this study provides preliminary information on the partitioning of persistent organic pollutant congeners in the blubber and blood of killer whales and elucidates the potential risk of circulating contaminants in these top predators.

Two killer whales (n = 2) from U.S. oceanaria were opportunistically sampled during necropsies in 2017. These subjects were a postreproductive, multiparous adult female (estimated age 41 years) that was euthanized due to a protracted respiratory tract disease and a female calf (3 months), born to another female, that died from acute pneumonia. Both whales were necropsied soon after death, and as a result, the samples collected from these subjects were considered fresh. Since this analysis requires high-quality samples, tissues collected from stranded individuals, which are often degraded due to decomposition, could not be used.

Both animals were examined via routine necropsy using standard procedures. Blood samples were collected via laceration of the axillary artery directly into 10 ml sterile serum blood collection tubes containing active thrombin (BD Vacutainer, Franklin Lakes, NJ). Samples were centrifuged (Sorvall ST 16R centrifuge with a Fisher 75003029 rotor) at 1,500 rpm (377 G) for 10 min and the serum decanted into PFA 7 ml rounded interior vials with screw top closures (Savillex, Eden Prairie, MN). One full-depth blubber sample (from the inner surface of the epidermis to the outer surface of the fascia attached to the muscle; female adult: 6.2 cm; female calf: 3 cm; measured by ruler near the collection site during necropsy) was collected from the dorsal surface, posterior to the dorsal fin from both

individuals. The samples were immediately wrapped in foil and placed in a plastic bag. After collection, serum and blubber samples were stored at −80°C and later shipped overnight to the Northwest Fisheries Science Center in Seattle, WA on dry ice. The frozen samples were stored at -80°C until analysis.

Concentrations of POPs in blubber and serum samples were determined using a gas chromatography/mass spectrometry (GC/MS) method described in Sloan et al. (2014). In this method, killer whale blubber and serum samples were weighed (masses ranging from 0.5 to 2.3 g), mixed with drying agents (sodium sulfate and magnesium sulfate) to remove water, and spiked with a surrogate standard (PCB 103; 75 ng). The samples were then packed in stainless steel sample cells and extracted with dichloromethane using an automated, pressurized solvent extractor. Prior to sample extract cleanup, a subsample of each extract was removed for gravimetric lipid determinations. Highly polar compounds were removed from the extracts using a glass, gravity flow cleanup column (custom made, 22mm id × 25mm length) containing alumina/silica followed by a size exclusion high performance liquid chromatography cleanup step to remove lipids and other biogenic compounds that have been shown to interfere with our GC/MS analyses. The analytes of interest in the cleaned-up sample extracts were separated on a 60m DB-5 GC and measured on a low-resolution MS operating in selected ion monitoring (SIM) mode. Additional details on the labware, equipment, and processes associated with this GC/MS method can be found in Sloan et al. (2014).

As part of our analytical laboratory quality assurance (QA) program (Sloan et al., 2019), each sample set included a solvent (dichloromethane) method blank and a National Institute of Standards and Technology Standard Reference Material (SRM 1945 whale blubber or NIST SRM 1958 human serum). For all sample sets, the method blanks met established QA criteria [no more than five analytes are to exceed 2 × lower limit of quantitation (LOQ) in a method blank]. For the NIST SRMs, concentrations of ≥70% of individual analytes, as well as the gravimetric percent lipid, were within 30% of either end of the 95% confidence interval of the certified values and thus, met our laboratory QA criteria. The percent recoveries of the surrogate standards for field and QA samples ranged from 94% to 125% and met our laboratory quality assurance criteria (percent recovery range 60%–130%).

Analysis included 45 polychlorinated biphenyl (PCB) congeners (17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 82, 87, 95, 99, 101/90, 105, 110, 118, 128, 138/163/164, 149, 151, 153/132, 156, 158, 170, 171, 177, 180, 183, 187/159/182, 191, 194, 195, 199, 205, 206, 208, 209), six dichlorodiphenyltrichloroethane (DDT) compounds (*p,p'*-DDD, *p,p'*-DDE, *p,p'*-DDT, *o,p'*-DDD, *o,p'*-DDE, *o,p'*-DDT), 11 polybrominated diphenyl ether (PBDE) congeners (28, 47, 49, 66, 85, 99, 100, 153, 154, 155, 183), and eight chlordanes (CHLD; *cis*-chlordane, *cis*-nonachlor, *trans*-chlordane, *trans*-nonachlor, nonachlor III, heptachlor, heptachlor epoxide, oxychlordane). For each individual, all compounds that were greater than the LOQ were included in the analysis. Consequently, the compounds included in the analysis varied somewhat by individual, depending on LOQ. The sum of contaminant concentration for each POP class (PCB, PBDE, DDT, CHLD) were calculated by summing concentrations of all measured congeners within each class.

Due to the small sample size (n = 2), analyses of individual compounds were not feasible. Therefore, contaminants within specific POP classes were combined for some analyses, as previously done in Ellisor et al. (2013).

The partitioning coefficient $\left(\frac{\text{blubber lipid corrected concentration}}{\text{serum lipid corrected concentration}}\right)$ of every compound was calculated to assess its relative distribution between blubber and blood serum. ANCOVAs were used to evaluate whether relationships differed by individual whale. If relationships did not differ, data from both individuals were combined. Linear and nonlinear regression analyses were used to determine relationships between blubber and serum contaminant concentrations within each POP class (PCB, PBDE, DDT, and CHLD).

As a preliminary approach to evaluating differences in partitioning patterns across delphinid species, specific POP concentrations in killer whale blubber samples were incorporated into linear regression equations developed for bottlenose dolphins ($\log \text{concentration}_{\text{plasma}} = b0 + b1 * \log \text{concentration}_{\text{blubber}}$, Yordy et al., 2010). Because the relationships developed for bottlenose dolphins included all age-classes and both sexes combined (Yordy et al., 2010), data for both killer whales were combined in the analysis. Although the two blood matrices (serum and plasma) differ, for example the presence of clotting factors in plasma or slightly higher metabolite concentrations in serum (Folsom et al., 1983; Teahan et al., 2006; Yu et al., 2011), lipid-normalized values are expected to account for variability between the two blood sample types. Furthermore, blubber samples from both the dolphins and killer

whales were taken from the dorsal surface posterior to the dorsal fin, which reduces the likelihood of sample location biases in lipid-normalized blubber samples (Ellisor et al., 2013). Linear regressions were then used to assess relationships between predicted plasma and observed serum values within specific POP classes. Congruence of linear relationships with the ideal 1:1 relationship was assessed by comparing slopes and intercepts (when slopes were not significantly different) using *t*-tests. All graphical and statistical analyses were conducted using SigmaPlot 14.0 software (Systat Software, Inc., San Jose, CA).

Despite the small number of killer whales from which high-quality biological tissues were available, the results from this study are still informative. This is the first study to report POP concentrations in paired blubber and serum samples from female killer whales and provides some preliminary insight into factors influencing circulating contaminant concentrations for this species.

Although we found some differences in the rank order of contaminant class concentrations across individuals, PCBs were the most predominant POP class, followed by DDTs in both blubber and serum from the two killer whale subjects (Table 1). Across all POP classes, blubber and serum contaminant concentrations in the calf were on average 2.6 ± 1.3 *SD* and 1.9 ± 0.9 *SD* times higher, respectively, than in the adult female. The calf's higher POP levels are most likely due to extensive contaminant transfer from her mother and a lack of growth dilution prior to death (Fearnbach et al., 2011; Hickie et al., 2007). In contrast, the 41-year-old female had given birth to four calves over her lifetime and her youngest calf was born 4.5 years prior to her death. POP levels are expected to be lower in multiparous adult females (Hickie et al., 2007).

Cross-study comparisons can be challenging due to differences in analytical methods and variation in individual POPs measured. When accounting for sex and age (when known), blubber contaminant concentrations measured in the present study were lower than levels reported previously for wild stranded and biopsied killer whales (Table 2). Discrepancies in killer whale blubber POP concentrations are related to environmental differences, and in particular, POP concentrations in prey consumed (Krahn et al., 2007; Ross et al., 2000). The results of the present study suggest that these killer whales in human care are fed less contaminated and/or lower trophic level prey, than prey consumed by wild killer whales that reside off heavily industrialized coastlines, such as Japan, Europe, and the west coast of North America (Aguilar & Borrell, 1994; Jepson et al., 2016; Krahn et al. 2007; Krahn et al., 2009; Lawson et al., 2020; Reijnders & de Ruiter-Dijkman, 1995), and even off less industrialized coastlines, such as southeast Greenland (Pedro et al., 2017). Furthermore, contaminant levels in the killer whales from the present study are considerably below the established threshold for sum PCBs in blubber (17,000 ng/g, lw) that were associated with impaired immune function in harbor seals (Ross et al., 1996). In contrast, PCB levels in several wild killer whale

TABLE 1 Lipid content (% wet weight) and sum persistent organic pollutant (POP) concentrations (ng/g, lw) in serum and blubber from a female adult and a female calf killer whale.

Individual		Serum	Blubber
Adult	Lipid	0.26	75.2
	∑PCBs	2,700	1,500
	∑DDTs	1,500	1,000
	∑PBDEs	460	250
	∑CHLDs	300	230
Calf	Lipid	0.61	77.9
	∑PCBs	4,900	3,300
	∑DDTs	2,100	2,200
	∑PBDEs	610	400
	∑CHLDs	1,400	880

Note: Abbreviations: PCBs – polychlorinated biphenyls, DDTs – dichlorodiphenyltrichloroethanes, PBDEs – polybrominated diphenyl ethers, CHLDs – chlordanes.

TABLE 2 Comparison of persistent organic pollutant concentrations (POP; ng/g, lw) in the blubber of adult female and calf killer whales from various locations. Mean values for age and contaminant concentrations (±*SD*, when available) are presented. This table includes both fish-eating and marine mammal-eating populations.

Subject	Location	Age	Blubber	Relative to Subject[a]	Reference
Adult – 41	BC, Canada	Adult – U	∑PCBs: 9300(2800)	~6x	Ross et al., 2000
	Washington, USA	Adult – 48(11)	∑PCBs: 26967(24613) ∑DDTs: 45667(46876) ∑CHLDs: 4867(4174) ∑PBDEs: 3267(1793)	~18x ~46x ~21x ~13x	Krahn et al., 2009
	Alaska, USA	Adult – U	∑PCBs: 18180(20028) ∑DDTs: 23326(30510)	~12x ~23x	Lawsen et al., 2020
	Rausu, Japan	Adult – U	∑PCBs: 30600(9990) ∑DDTs: 65800(34237) ∑CHLDs: 22600(9659) ∑PBDEs: 264(72)	~20x ~66x ~98x ~1x	Kajiwara et al., 2006
	UK	Adult – U	∑PCBs: 241980	~161x	Jepson et al., 2016
	Ireland	Adult – U	∑PCBs: 138900	~93x	Jepson et al., 2016
	Canary Islands	Adult – U	∑PCBs: 108130	~72x	Jepson et al., 2016
	St. of Gibraltar	Adult – U	∑PCBs: 215430	~144x	Jepson et al., 2016
	SE Greenland	Adult – U	∑PCBs: 48600 ∑DDTs: 30100 ∑CHLDs: 19300	~32x ~30x ~84x	Pedro et al., 2017
Calf – 3 mo	Rausu, Japan	Calf – few mo	∑PCBs: 51000(18385) ∑DDTs: 145500(77075) ∑CHLDs: 49000(26870) ∑PBDEs: 515(35.4)	~15x ~66x ~56x ~1x	Kajiwara et al., 2006
	Sylt, Germany	Neonate	∑PCBs: 225000	~68x	Schnitzler et al., 2011

[a]Relative to Subject = Reported ∑POP Blubber/∑POP study subject blubber concentration; Abbreviations: U – Unknown age, PCBs – polychlorinated biphenyls, DDTs – dichlorodiphenyltrichloroethanes, PBDEs – polybrominated diphenyl ethers, CHLDs – chlordanes.

populations (Jepson et al., 2016), including the endangered fish-eating Southern Residents, routinely exceed this threshold level for impaired immune function (Herman et al., 2005; Krahn et al., 2007; Krahn et al., 2009; Lawson et al., 2020; Pedro et al., 2017; Ross et al., 2000; Ylitalo et al., 2001).

Lipid-normalized serum and blubber POP concentrations were positively correlated across all classes. Serum PBDEs, DDTs, and CHLDs were positively, linearly related to corresponding blubber levels. With the exception of CHLDs, for which there was a significant effect of individual as well as an interaction effect, data from the two individuals were combined in linear regressions (DDT: $F_{1,8}$ = 242.8, R^2 = 0.96, n = 10, p < .001; PBDE: $F_{1,13}$ = 60.2, R^2 = 0.81, n = 15, p < .001; CHLD adult: $F_{1,4}$ = 109.6, R^2 = 0.96, n = 6, p < .001; CHLD calf: $F_{1,4}$ = 209.9, R^2 = 0.98, n = 6, p < .001; Figure 1). Slopes did not differ from the 1:1 relationship, with the exception of the regression equation for CHLDs in the calf (T_{10} = 4.1, p = 0.002). Y-intercepts were significantly greater than that of the 1:1 relationship for the PBDEs regression and the regression for CHLDs in the adult (PBDEs: T_{28} = 4.9, p < .0001; CHLDs adult: T_{10} = −8.4, p < .0001) and marginally greater for the DDTs regression (DDTs: T_{18} = 2.1, p = .049; power = 0.52,

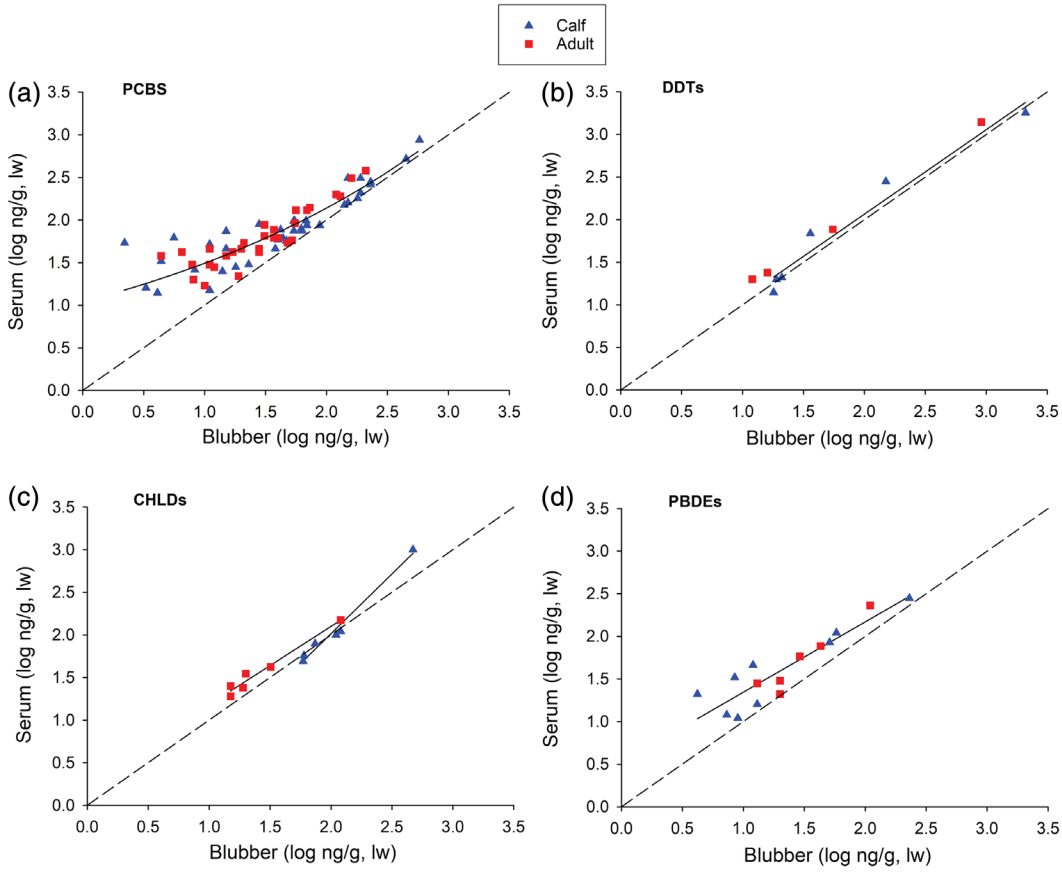

FIGURE 1 Serum persistent organic pollutant congener concentrations in relation to corresponding blubber concentrations for (a) PCB, (b) DDT, (c) CHLD, and (d) PBDE congeners. Each data point represents an individual congener within each specific contaminant group. Data from both individuals were combined for regression analyses (female calf: triangle, adult female: square) except for CHLDs. All regressions are statistically significant (all p < .001) and are indicated by solid lines (PCBs: y = 1.04exp(0.360x), R^2 = 0.82, n = 69; DDTs: y = 0.134 + 0.988x, R^2 = 0.96, n = 10; CHLDs calf: y = −0.788 + 1.40x, R^2 = 0.98, n = 6; CHLDs adult: y = 0.262 + 0.919x, R^2 = 0.96, n = 6; PBDEs: y = 0.519 + 0.826x, R^2 = 0.81, n = 15). Ideal 1:1 relationships (dashed lines) are presented for comparison.

which is considered low). The relationship between serum and blubber PCB concentrations was best described by an exponential relationship ($F_{1,68}$ = 316.7, R^2 = 0.82, n = 69, p < .0001; Figure 1A). Only PCB congeners with relatively high blubber concentrations tended to be highly correlated with serum concentrations. These overall significant positive relationships demonstrate that while lipid-normalized serum POP levels were usually greater than corresponding lipid-normalized blubber values, serum POP levels reflect levels in the blubber. Previous studies on polar bears (*Ursus maritimus*), loggerhead sea turtles (*Caretta caretta*), Hawaiian monk seals, and bottlenose dolphins have also found that blood POP levels reflect blubber or fat POP levels (Balmer et al., 2018; Bernhoft et al., 1997; Keller et al., 2004; Lopez et al., 2012; Reddy et al., 1998; Yordy et al., 2010). Indeed, similar to the results from the present study, Yordy et al. (2010) reported that lipid-normalized POP concentrations were highly correlated in delphinid blubber and plasma (R^2 = 0.828 to 0.976 for 18 congener specific relationships determined from samples collected from 56 free-ranging dolphins).

While POP levels are correlated in killer whale blubber and blood, partitioning across the two matrices varied by POP class, congener, and individual whale (Table 3). For both individuals, mean partitioning coefficients were the highest for CHLDs (Adult: 0.72 ± 0.10 *SD*, Calf: 0.98 ± 0.26 *SD*) and DDTs (Adult: 0.66 ± 0.05 *SD*, Calf: 0.91 ± 0.32 *SD*), indicating that these contaminant classes may be more readily transferred between blubber and blood matrices. The lowest mean partitioning coefficient in the adult killer whale was for PCBs (0.50 ± 0.19 *SD*) while the lowest for

TABLE 3 Partitioning coefficients (PC) between blubber (ng/g, lw) and serum (ng/g, lw) persistent organic pollutant (POP) concentrations ($\frac{\text{blubber lipid corrected concentration}}{\text{serum lipid corrected concentration}}$) for a female adult and a female calf killer whale.

	PC			PC			PC	
POP	Adult	Calf	POP	Adult	Calf	POP	Adult	Calf
PCB 17	ND[a]	0.21	PCB 149	0.52	0.88	*p,p*'-DDD	0.71	0.54
PCB 18	0.12	0.33	PCB 151	0.61	1.01	*p,p*'-DDE	0.65	1.17
PCB 28	0.24	0.55	PCB 153/132	0.55	0.67	*p,p*'-DDT	0.67	0.52
PCB 31	0.15	0.21	PCB 156	0.41	0.56	PBDE 28	ND[a]	0.82
PCB 44	0.37	0.68	PCB 158	0.41	0.77	PBDE 47	0.48	0.82
PCB 49	0.43	0.83	PCB 170	0.65	0.55	PBDE 49	0.46	0.81
PCB 52	0.48	0.93	PCB 171	0.59	0.64	PBDE 66	ND[a]	0.61
PCB 66	0.40	0.84	PCB 177	0.67	0.72	PBDE 99	0.56	0.53
PCB 70	0.26	0.32	PCB 180	0.60	0.48	PBDE 100	0.50	0.60
PCB 74	0.43	0.82	PCB 183	0.60	0.57	PBDE 153	0.95	0.26
PCB 82	ND[a]	0.73	PCB 187/159/182	0.68	0.61	PBDE 154	0.67	0.26
PCB 87	0.39	0.72	PCB 194	0.89	0.20	PBDE 155	ND[a]	0.20
PCB 95	0.48	0.94	PCB 195	ND[a]	0.29	*cis*-chlordane	0.79	1.05
PCB 99	0.60	1	PCB 199	0.90	0.31	*cis*-nonachlor	0.76	1.10
PCB 101/90	0.53	0.82	PCB 206	0.86	0.09	HPE[b]	0.60	0.94
PCB 105	0.39	0.79	PCB 208	ND[a]	0.13	nonachlor III	0.79	1.20
PCB 110	0.35	0.69	PCB 209	ND[a]	0.04	oxychlordane	0.57	1.10
PCB 118	0.43	0.90	*o,p*'-DDD	ND[a]	0.95	*trans*-nona[c]	0.80	0.47
PCB 128	0.43	0.78	*o,p*'-DDE	0.60	1			
PCB 138/163/164	0.52	0.87	*o,p*'-DDT	ND[a]	1.29			

[a]ND = Not determined due to concentrations below detection limits.
[b]HPE = Heptachlor epoxide.
[c]*trans*-nona = *trans*-nonachlor.

the calf was for PBDEs (0.55 ± 0.25 *SD*). Partitioning coefficients for individual contaminant compounds were highly variable across the two whales (Table 3). Interestingly, as a whole, POP partitioning ratios were greater in the calf compared to the adult female. For the calf, the partitioning coefficients of 29 individual POPs approached (>0.70) or were >1, while for the female, partitioning coefficients of only nine contaminants fell within this range. These preliminary results suggest that, despite a high degree of variability, partitioning coefficients for killer whales are typically <1, which means that lipid-normalized POP concentrations are greater in the serum than in the blubber.

While two previous studies reported that, for the most part, POP concentrations are higher in dolphin blubber than in blood (partitioning coefficients >1; Tanabe et al., 1981; Yordy et al., 2010), a more recent study on bottlenose dolphins (Balmer et al., 2018) reported that, similar to the current study, most POP concentrations were higher in blood compared to blubber (partitioning coefficient <1). This finding was also reported for 8 out of the 14 measured contaminant compounds in an earlier study on bottlenose dolphins (Reddy et al., 1998). Several factors could explain the variability in partitioning coefficients across studies as well as across individuals. The killer whales in this study both experienced health complications, but on different time scales: the female had a chronic health condition while the calf had an acute illness. These circumstances may have increased lipid mobilization in order to sustain vital bodily functions, consequently increasing serum POPs (Debier et al., 2006). Additionally, blood lipid content can strongly influence serum POP levels (O'Brien et al., 2015). The serum lipid content for the killer whale calf (0.61% lipid content) was greater than that of the adult female (0.26% lipid content) as well as the plasma lipid contents reported for bottlenose dolphins (0.23%–0.32% from Balmer et al., 2018; 0.41%–0.53% from Yordy et al., 2010). Thus, variability in blood lipid content may be responsible, in part, for the disparate results across delphinid studies and between the two killer whales in the present study.

Relationships between observed killer whale serum POP concentrations and predicted plasma POP concentrations (from equations developed for bottlenose dolphins from Yordy et al., 2010) were positively correlated for all contaminant classes (PCBs: $F_{1,44} = 102.9$, $R^2 = 0.69$, $n = 46$, $p < .001$; DDTs: $F_{1,6} = 352.7$, $R^2 = 0.98$, $n = 8$, $p < .001$; PBDEs: $F_{1,6} = 25.0$, $R^2 = 0.77$, $n = 8$, $p = .002$; CHLDs: $F_{1,6} = 65.8$, $R^2 = 0.90$, $n = 8$, $p < .001$). This was somewhat expected because both killer whales and bottlenose dolphins belong to the delphinid family, and presumably several physiological characteristics are conserved across the two species (Baird, 2000). However, while these findings suggest similarities in POP partitioning patterns across the two species, the dolphin-derived equations ubiquitously underpredicted killer whale serum concentrations (Table 4).

Despite underestimating measured serum levels, certain POP concentrations were better predicted than others (Table 4). Dissimilar to the other POP classes, the regression equation (slope and y-intercept) for CHLDs was not significantly different from that of the 1:1 relationship, which suggests that the CHLD equations for dolphins could be suitable for female killer whales. As CHLDs are readily mobilized by killer whales and other cetaceans (Hoekstra et al., 2003; Wolkers et al., 2007), blood CHLD concentrations may be more universally predicted from blubber levels, compared to other POP classes. The small sample size in the present study limits our ability to thoroughly evaluate the suitability of using equations developed for dolphins to predict partitioning patterns in various age/sex classes of killer whales, but it is also possible that partitioning patterns may be species-specific. Additional studies with larger sample sizes are warranted as well as direct comparisons between serum and plasma to adequately investigate species-specific partitioning patterns.

In conclusion, this is the first study to report partitioning of persistent organic pollutants in the blubber and blood (serum) of killer whales. Because blood and blubber POP levels are positively correlated, as they are in bottlenose dolphins (Yordy et al., 2010), this preliminary study provides evidence that POP concentrations quantified in blubber biopsies reflect circulating POP concentrations in killer whales. While this study was conducted on animals in managed care, these preliminary findings are relevant to assessing risk of POP exposure to killer whale populations elsewhere. For example, members of the endangered Southern Resident killer whale population have greater blubber contaminant loads (Krahn et al., 2007, 2009; Ross et al., 2000) compared to other North Pacific fish-eating killer whale populations and are therefore likely to have correspondingly higher levels in their blood. As this population may also be prey limited (Krahn et al., 2004), individual Southern Resident killer whales may also have high rates of

TABLE 4 Difference between observed and predicted persistent organic pollutant (POP) blood concentrations (Δobs-pred[a], log ng/g lw). Observed concentrations were determined from serum samples collected from a female adult and a female calf killer whale in this study. Predicted values were calculated from POP concentrations determined in matching blubber samples using regression equations developed by Yordy et al. (2010) relating bottlenose dolphin blubber and plasma contaminant data.

	Calf	Adult		Calf	Adult
POP	Δobs-pred	Δobs-pred	POP	Δobs-pred	Δobs-pred
PCB 18	0.69	1.18	PCB 183	0.49	0.48
PCB 44	0.41	0.59	PCB 194/205	1.34	0.38
PCB 49	0.31	0.50	PCB 195	0.80	ND[b]
PCB 52	0.33	0.64	PCB 199	0.85	0.37
PCB 66/70	0.40	0.67	PCB 206	1.48	0.40
PCB 74	0.33	0.53	PCB 208	1.48	ND[b]
PCB 87	0.37	0.56	*cis*-chlordane	0.18	0.34
PCB 99	0.23	0.47	*trans*-chlordane	0.65	0.38
PCB 105	0.34	0.67	*cis*-nonachlor	0.20	0.26
PCB 110	0.32	0.60	oxychlordane	0.19	0.40
PCB 118	0.19	0.47	*o,p'*-DDE	0.45	0.74
PCB 128	0.28	0.49	*p,p'*-DDE	0.23	0.47
PCB149	0.20	0.44	*p,p'*-DDT	0.63	0.57
PCB 151	0.29	0.57	*o,p'*-DDT/*p,p'*-DDD	0.59	0.52
PCB 153/132	0.36	0.45	PBDE 47	0.23	0.45
PCB 156	0.73	0.92	PBDE 99	0.47	0.45
PCB 170	0.49	0.42	PBDE 153	0.86	0.26
PCB 177	0.38	0.43	PBDE 154	1.03	0.58

[a]Δobs-pred = observed serum concentration (log ng/g lw) – predicated plasma concentration (log ng/g lw).
[b]ND = not determined due to the concentration being below detection limits.

lipid mobilization, which can increase circulating contaminant levels (Debier et al., 2006) and the likelihood of contaminants to disrupt physiological pathways, potentially causing detrimental health issues (Fair et al., 2010; Krahn et al., 2009; Ross et al., 1996) and subsequent population-wide effects (Desforges et al., 2018). Due to the limitations of this study, further investigation is required to understand factors that dictate partitioning of POPs in killer whale blubber and blood. Regardless of the limitations, this study provides important new data on partitioning of POPs in the blood and blubber of killer whales, and importantly, demonstrates that persistent organic pollutant levels in blood serum are highly correlated with levels in the blubber.

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AUTHOR CONTRIBUTIONS

Molly McCormley: Data curation; formal analysis; investigation; methodology; visualization; writing-original draft; writing-review & editing. **Dawn Noren:** Conceptualization; data curation; formal analysis; funding acquisition; investigation; methodology; project administration; resources; supervision; visualization; writing-original draft; writing-review & editing. **Gina Ylitalo:** Data curation; funding acquisition; methodology; resources; supervision; validation; writing-original draft; writing-review & editing. **Judy St. Leger:** Data curation; resources; writing-review & editing.

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