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Baseline

High levels of persistent organic pollutants measured in blubber of island-associated false killer whales (*Pseudorca crassidens*) around the main Hawaiian Islands

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Persistent organic pollutants (POPs) have been measured in tissues of marine mammals since the mid 1960s (Holden and Marsden, 1967; Wolman and Wilson, 1970). These compounds include several pesticides (e.g., DDTs, chlordanes) and industrial chemicals (e.g., PCBs) that are ubiquitous, highly lipophilic and not readily degraded or metabolized. As a result, they can biomagnify to high levels in lipid-rich tissues of top-level marine predators. POPs enter marine waters via direct inputs (e.g., sewage outfalls, industrial and agricultural runoff) as well as from indirect sources (e.g., ocean currents) (Friedlander et al., 2005). Exposure to POPs in marine mammals has been linked to a number of biological effects including reproductive impairment (DeLong et al., 1973; Subramanian et al., 1987), reduced reproductive success (Wells et al., 2005), immune suppression (De Swart et al., 1994; Hammond et al., 2005; Ross et al., 1995) and endocrine disruption (reviewed in O'Hara and O'Shea (2001)). Although many POPs, such as PCBs and DDTs, have been banned for production or use in the US for more than thirty years, some of these compounds are still used in other regions of the world (Fielder, 2008; van den Berk, 2009) and continue to be measured in the tissues of marine mammals throughout coastal regions of the US.

Another class of POPs gaining the attention of environmental scientists and managers are the polybrominated diphenyl ethers (PBDEs). Three different PBDE technical mixtures (i.e., penta-BDE, octa-BDE, deca-BDE) have been manufactured and added as flame retardants to plastics, textiles, clothing, electronic circuit boards and other materials in industrial and developing nations (de Wit, 2002). Deca-BDE is the primary commercial product produced and used in the U.S. as a result of the sole manufacturer phasing out the production of penta-BDE and octa-BDE (U.S. EPA, 2006). Similar to PCBs, these compounds are lipophilic, persistent, and tend to bioaccumulate in marine mammal tissues (de Wit et al., 2004; Ikonomou et al., 2002a, 2002b). Some of the highest levels of PBDEs have been measured in tissues of wildlife and humans from North America due to high volume PBDE use in this region of the world (Hites, 2004; Ikonomou et al., 2002b; LeBeuf et al., 2004). Because these compounds can travel over long distances via atmospheric transport, they have been measured in marine organisms throughout the world, including Antarctica and the Arctic (Corsolini et al., 2006; de Wit et al., 2004). Exposure to PBDEs has been associated with a variety of biological effects (e.g., thyroid disruption, neurobehavioral effects) in laboratory animals (de Wit, 2002) but currently no threshold levels for PBDEs have been established for toxicological effects in marine mammals.

The main Hawaiian Islands include eight volcanic islands (i.e., Hawai'i, Kaho'olawe, Kauai, Lana'i, Mau'i, Moloka'i, Ni'ihua, O'ahu) that are located in the middle of the Pacific Ocean, approximately 1500 miles southwest of the contiguous US. Tourism, defense and agriculture (e.g., production of raw sugar, fresh pineapple) are the primary contributors to the economy of this region (State of Hawaii, Dept. of Business, Economic Development and Tourism, 2008). In addition, coastal development of the main Hawaiian Islands is ongoing and includes conversion of agricultural lands to residences and resorts, as well as expansion of harbor facilities to accommodate large cargo and cruise ships (Friedlander et al., 2005). Activities related to these industries and development processes can be potential sources of POPs to this region. For example, in the 1970's, elevated levels of chlorinated insecticides used to control agricultural pests and termites were reported in water, sediment and aquatic organisms from the main Hawaiian Islands (Bevenue et al., 1972; Tanita et al., 1976).

Around the main Hawaiian Islands, the highest trophic level cetacean regularly encountered is the false killer whale (*Pseudorca crassidens*). Based on observations of predation, individuals from this population appear to feed primarily on large game fish such as mahimahi (*Coryphaena hippurus*), yellowfin tuna (*Thunnus albacares*) and swordfish (*Xiphias gladius*), some of which can be long-lived (Baird et al., 2008). Population estimates for cetaceans within the Hawaiian Exclusive Economic Zone (EEZ) indicate that false killer whales may have the smallest population size of any odontocete within the Hawaiian EEZ (Barlow, 2006). In addition, within this region of Hawai'i there is evidence of population structure for false killer whales, with genetically differentiated insular and offshore populations (Chivers et al., 2007).

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Recently, the population of false killer whales around the main Hawaiian Islands has been estimated at 123 individuals (CV = 0.72) based on a photographic mark-recapture analysis (Baird et al., 2005). There is evidence that this population may have declined substantially over the last 20 years (Reeves et al., 2009). A number of potential causes have been identified, including mortality in the Hawai'i-based long-line fishery (Baird and Gorgone, 2005; Forney and Kobayashi, 2007), reduction in their prey base, and potential health or reproductive effects due to exposure to high levels of POPs (Reeves et al., 2009). Although the details of life history of false killer whales are poorly known, females appear to reach sexual maturity between 8 and 14 years (Purves and Pilleri, 1978), have long calving intervals (estimated at 6.9 years), and may reach 62.5 years (Kasuya, 1986). Males are thought to mature as much as 10 years later and live to 57.5 years (Kasuya, 1986). As a long-lived upper-trophic level predator, false killer whales are likely to accumulate high levels of POPs. Few individuals have been analyzed for POPs, but high levels have been documented in animals that stranded around British Columbia (Baird et al., 1989; Jarman et al., 1996). In the present study, we report concentrations of PBDEs and other POPs measured in biopsy samples collected from nine individuals from the insular population to determine the baseline levels of these contaminants and assess whether their exposure levels to PCBs may be a risk factor for this population.

Field operations were undertaken as part of ongoing studies of odontocetes around the main Hawaiian Islands (see Baird et al., 2008). In July 2008, biopsy blubber samples were collected from nine individual false killer whales from the insular population using a 45 kg pull Barnett RX-150 crossbow and Larsen biopsy tips, measuring 25 mm long and 8 mm wide. A high-density foam collar on the biopsy dart prevented penetration greater than 18 mm. After collection, the biopsy samples were stored in a cooler with ice packs while in the field and transferred to a −20 °C freezer for short-term storage before being stored in a −80 °C freezer. Biopsied individuals were photo-identified, and photographs compared to each other to eliminate duplicate samples, and to the catalog of Baird et al. (2008) to assess population identity and sighting history. Age class (adult/subadult) was assessed in the field based on relative body size and in some cases confirmed based on sighting history. The sex determination of each whale was conducted using zinc finger gene amplification (Chivers et al., 2007). Based on photographs all individuals appeared to be "healthy" (i.e., not emaciated).

Samples were extracted and analyzed for POPs using the gas chromatography/mass spectrometry method of Sloan et al. (2005). Blubber (0.1–0.3 g) was extracted with methylene chloride using an accelerated solvent extractor after the addition of a surrogate standard (PCB 103; 1 ng/μL). This procedure was followed by a clean-up step of the extract on a single stacked, gravity flow silica gel/alumina column to remove any highly polar compounds present in the sample. Using high-performance size exclusion liquid chromatography, the POPs were separated from the bulk lipid and other biogenic material present in each sample, and the cleaned extract was analyzed for POPs using a low-resolution quadrupole GC/MS system equipped with a 60 m DB-5 GC capillary column and a electron impact mass spectrometer in selected ion monitoring mode. The instrument was calibrated using sets of up to ten multi-level calibration standards of known concentrations. Percent lipid and lipid class profiles were determined in biopsy blubber samples using thin-layer chromatography with flame ionization detection (Ylitalo et al., 2005). In this method, each lipid extract sample was spotted on a Type SIII Chromarod and developed in a chromatography tank containing 60:10:0.02 hexane:diethyl ether:formic acid (v/v/v). The lipid classes were separated based on polarity and measured using flame ionization detection. Percent lipid values were calculated by summing the concentrations of five lipid classes (i.e., sterol esters/wax esters, triglycerides, free fatty acids, cholesterol, phospholipids) for each sample, using the mean of two measurements.

All blubber contaminant concentrations are reported in ng/g, lipid weight. Sum PCBs (∑PCBs) includes the sum of congeners 17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 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 and 209. Sum DDTs (∑DDTs) is the sum of *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDT and *p,p′*-DDT. Sum chlordanes (∑CHLDs) is the sum of heptachlor, heptachlor epoxide, oxychlordane, *gamma*-chlordane, nona-III-chlordane, *alpha*-chlordane, *trans*-nonachlor and *cis*-nonachlor. Sum PBDEs (∑PBDEs) is the sum of congeners 28, 47, 49, 66, 85, 99, 100, 153, 154 and 183. Additional POPs analyzed in the current study include hexachlorobenzene (HCB), β-hexachlorocyclohexane (β-HCH), aldrin, dieldrin, mirex and endosulfan I.

As part of a performance-based quality assurance program (Sloan et al., 2006), a method blank and a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM®

Table 1
Concentrations of ∑CHLDs, ∑DDTs, ∑PCBs and ∑PBDEs measured in biopsy blubber samples of false killer whales from the main Hawaiian Islands sampled in July 2008.

Sample ID	Sex/age class	Collection date	Percent lipid	Lipid weight (ng/g)			
				∑CHLDs[a]	∑DDTs[a]	∑PCBs[a]	∑PBDEs[a]
RWB2008Jul26.02	Subadult – female (S)	7/26/2008	16	2900	16,000	14,000	2400
RWB2008Jul26.03[b]	Subadult – male	7/26/2008	41	3200	23,000	24,000	2900
		Mean ± SD	29 ± 18	3100 ± 210	20,000 ± 4900	19,000 ± 7100	2700 ± 350
RWB2008Jul16.01	Adult male (M)	7/16/2008	18	4100	83,000	33,000	780
RWB2008Jul16.04	Adult male	7/16/2008	16	4900	43,000	33,000	1600
		Mean ± SD	17 ± 1.4	4500 ± 570	63,000 ± 28,000	33,000 ± 0	1200 ± 580
RWB2008Jul16.03	Adult female (F)	7/16/2008	23	190	1200	1000	26
RWB2008Jul16.05	Adult female	7/16/2008	36	1300	8300	11000	1700
RWB2008Jul16.06	Adult female	7/16/2008	35	430	2500	2200	120
RWB2008Jul16.07[b]	Adult female	7/16/2008	12	140	1200	1100	<LOQ[c]
RWB2008Jul26.06	Adult female	7/26/2008	16	310	1800	2100	260
		Mean ± SD	24 ± 11	470 ± 480	3000 ± 3000	3500 ± 4200	420 ± 720
p Value[d]			0.6393	**0.0074**	**0.0029**	**0.0101**	0.1832
Tukey–Kramer HSD[e]			–	M,F; S,F	M,F; S,F	M,F; S,F	–

[a] Individual compounds summed are reported above.
[b] Mother/offspring pair.
[c] <LOQ for the sum indicates concentrations of all compounds included in the sum were below their individual limits of quantitation. For each <LOQ, a value of zero was used to calculate the mean and standard deviation of the mean.
[d] Significant differences (ANOVA, $p < 0.05$) in POPs and lipid concentrations based on age class are shown in bold.
[e] Unlike letters indicate significant differences using Tukey–Kramer honestly significant difference (HSD) test ($p < 0.05$).

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1945) were analyzed with the false killer whale blubber samples. Concentrations of individual analytes measured in SRM® 1945 were in excellent agreement with the reference values published by NIST. Other quality control samples met established laboratory criteria. POP concentrations were $\log_{10}(x+1)$ transformed and percent lipid values were arcsine square root transformed to increase the homogeneity of variance. Analysis of variance (ANOVA) and the Tukey–Kramer honestly significant difference test (HSD) were used to compare mean concentrations of POPs among three age/sex classes (subadult whales (both males and females), adult females, adult males) (Zar, 1999). The level of significance used for all statistical tests was $\alpha \leqslant 0.05$. All statistical analyses were completed using JMP Statistical Software (SAS Institute, Inc., Cary, NC).

The most abundant POPs measured in biopsy blubber of false killer whales from the main Hawaiian Islands were DDTs and PCBs, with concentrations ranging from 1000 to 83000 ng/g, lipid (Table 1). PBDEs, chlordanes, β-HCH, dieldrin, HCB and mirex were also measured in these whales but at much lower concentrations than DDTs and PCBs (Table 1, Fig. 1). Endosulfan I and aldrin, on the other hand, were below the LOQ for all animals analyzed in the current study. Recent studies have reported measuring relatively low levels of PCBs, DDTs, PBDEs and other contaminants in aquatic organisms from the main Hawaiian Island region (Brasher and Wolff, 2004; Kimbrough et al., 2009; Miao et al., 2001; Orazio et al., 2003; Xu et al., 2009; Yang et al., 2008) but no data have been previously available for false killer whales or their presumed prey. Consumption of contaminated prey is the primary route of exposure for marine mammals (Aguilar et al., 1999).

Age class and sex appeared to influence the concentrations of POPs measured in the false killer whales. Mean levels of ∑CHLDs, ∑DDTs and ∑PCBs were significantly different among the three age/sex classes of whales, with adult females having lower values than those measured in adult males and subadults (Table 1). Similarly, the mean PBDE concentration in adult females was lower than those in subadults and adult males, but these differences were not significant at $\alpha = 0.05$ level (Table 1). Examination of POPs measured in blubber samples of a mother–offspring pair (RWB2008Jul16.07 and RWB2008Jul26.03) also showed that the levels of ∑CHLDs, ∑DDTs, ∑PCBs (Table 1) and mirex (data not shown) were at least an order of magnitude higher in the subadult male offspring than those measured in his mother. A number of contaminant studies on odontocetes have also reported lower POP levels in adult females compared to those measured in blubber of adult males and juveniles (subadult) (Hansen et al., 2004; Tilbury et al., 1999; Wells et al., 2005; Westgate et al., 1997; Ylitalo et al., 2001) due to the transfer of lipids and the POPs associated with these lipids from mother to calf during gestation and lactation (Aguilar and Borrell, 1994; Gardner et al., 2007). In contrast, males continue to accumulate these compounds throughout their lives.

One finding of interest in the current study was that subadult whales had the highest mean level of ∑PBDEs measured in these animals; however, these differences were not significant ($p = 0.1832$) among the age classes of whales (Table 1). The subadults also had elevated mean HCB, β-HCH and dieldrin concentrations (Fig. 1) compared to those measured in adult males and females but only dieldrin levels were significantly different ($p = 0.0496$) among the age classes. In recent killer whale (*Orcinus orca*) studies, higher levels of ∑PBDE, HCB and ∑HCHs have been reported in blubber of juvenile fish-eating individuals ("Southern Residents") compared to those determined in adult males and females from the same population (Krahn et al., 2007b, 2009). The elevated levels of PBDEs, HCB, β-HCH and dieldrin measured in the blubber of the subadult false killer whales may be due to differences in prey items or feeding rates, as well as variations in metabolism and excretion of these lipophilic compounds compared to adults (Aguilar et al., 1999). These findings of elevated contaminant levels in subadult whales are a concern as these animals are still developing biologically and may be at higher risk to deleterious effects associated with exposure to these compounds than adults in the same population.

The percent lipid measured in the biopsy blubber samples of the false killer whales ranged from 12–41% and contained primarily triglycerides (>84%) and wax esters (<16%). These percent lipid values are comparable to those reported in biopsy blubber samples of Eastern North Pacific killer whales that were analyzed by the same quantitation method (Herman et al., 2005; Krahn et al., 2007a,b, 2009; Ylitalo et al., 2001). Similar to our findings, Litchfield et al. (1975) reported that blubber of false killer whales contained both wax esters (4%) and triglycerides (96%).

The mean concentrations of ∑PCBs, ∑DDTs and ∑CHLDs measured in biopsy blubber of adult male false killer whales in the present study are much lower than those reported previously in blubber of two adult male false killer whales (Table 2) that stranded in British Columbia in the late 1980s (Baird et al., 1989; Jarman et al., 1996). These findings may be due to differences in analytical methodologies or variations in contaminant levels in presumably "healthy" wild-ranging whales (current study) vs. stranded animals that may have been in poor health. In addition, it is also probable that differences in contaminant levels in feeding ranges (waters of main Hawaiian Islands vs. west coast of North America) and sample collection years (2008 vs. 1987/1989) as well as variations in the ages of animals sampled also contributed to these differences in POPs levels observed for the false killer whales (Aguilar et al., 1999). In contrast to ∑PCBs, ∑DDTs and ∑CHLDs, the levels of mirex – a POP used as a fire retardant and insecticide in the US until 1978 (ASTDR, 1995) – measured in whales in the

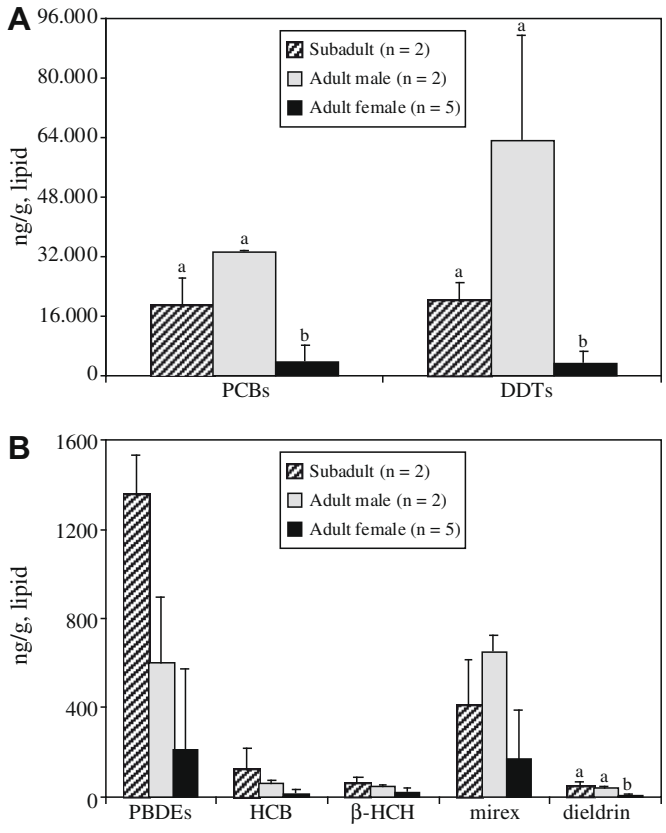

Fig. 1. Mean (±SD) concentrations of ∑PCBs and ∑DDTs (A) and ∑PBDEs HCB, β-HCH, mirex and dieldrin (B) measured in biopsy blubber samples of adult and subadult false killer whales from the main Hawaiian Islands. Bars with unlike letters differ significantly; Tukey–Kramer HSD test, $p < 0.05$.

Table 2
Concentrations of persistent organic pollutants measured in blubber of adult male marine mammals from the west coast of North America.

Species	Collection region	Collection year(s)	*n*	Percent lipid	Lipid weight (ng/g)					Reference
					Mirex	∑CHLDs	∑DDTs	∑PCBs	∑PBDEs	
False killer whale	Hawai'i	2008	2	17 ± 1.4	610 ± 510	4500 ± 570	63,000 ± 28,000	33,000 ± 0	1200 ± 580	Current study
False killer whale	British Columbia	1987, 1989	2	91 ± 2.8	260 ± 99	15,000 ± 710	1,000,000 ± 1400,000	45,000 ± 7500	Not analyzed	Jarman et al. (1996)
Harbor porpoise	British Columbia	1991/1993	5	80 ± 10	Not analyzed	Not analyzed	Not analyzed	Not analyzed	810 ± 740	Ikonomou et al. (2002b)
California sea lion	Southern California	1994/2006	5	38 ± 41	Not detected	4900 ± 7700	2300,000 ± 2900,000	290,000–440,000	55,000 ± 79,000	Blasius and Goodmanlowe (2008) and Meng et al. (2009)
Offshore killer whale	Alaska	2003/2004	4	18 ± 3.7	760 ± 180[a]	16,000 ± 2300	420,000 ± 100,000	110,000 ± 22,000	3300 ± 940	Krahn et al. (2007a)
Resident killer whale	Alaska	2003/2004	40	25 ± 11	170 ± 64[a,b]	6200 ± 2400	21,000 ± 12,000	13,000 ± 5900	76 ± 70	Krahn et al. (2007a)
Resident killer whale	Puget Sound, WA	2004/2007	10	22 ± 6.1	190 ± 72[a]	8600 ± 3300	82,000 ± 38,000	56,000 ± 46,000	4400 ± 1800	Krahn et al. (2007b, 2009)

[a] Data from J. Bolton, Pers. Comm.
[b] $n = 37$.

current study were more than two times higher than those found in the false killer whales that stranded in British Columbia (Baird et al., 1989; Jarman et al., 1996). These differences in mirex levels between the two whale groups may be due to the more extensive use of this pesticide in Hawai'i (used to control mealy bugs in pineapple fields) compared to the west coast of North America (ASTDR, 1995; UNEP/FAO, 2005).

We compared the POPs levels measured in the current study with those reported recently in other fish-eating marine mammal species from the west coast of North America (Blasius and Goodmanlowe, 2008; Ikonomou et al., 2002b; Krahn et al., 2007a,b, 2009; Meng et al., 2009) because no contemporary POP data have been published on cetaceans from the Hawaiian Island region (O'Shea et al., 1980). Mean ∑CHLDs, ∑PCBs and ∑DDTs concentrations measured in the Hawaiian false killer whales were lower than the values reported for California sea lions (*Zalophus californianus*) from southern California (Blasius and Goodmanlowe, 2008), offshore killer whales sampled in Alaska (Krahn et al., 2007a) and "Southern Resident" killer whales (Krahn et al., 2007b, 2009) but were higher than those measured in Alaskan resident killer whales except for ∑CHLDs (Krahn et al., 2007a). The mean level of ∑PBDEs (Table 2) measured in the Hawaiian false killer whales was higher or comparable to those reported in Alaskan resident killer whales (Krahn et al., 2007a) and harbor porpoises (*Phocoena phocoena*) that stranded in urban harbors of British Columbia (Ikonomou et al., 2002b) but was lower than the mean values measured in blubber of California sea lions that stranded in southern California (Meng et al., 2009), as well as Southern Resident (Krahn et al., 2007b, 2009) and offshore killer whales (Krahn et al., 2007a). These findings were expected as offshores and "Southern Residents", as well as California sea lions, appear to spend a portion of their time feeding on fish from highly urbanized areas (e.g., Puget Sound, Washington, central and southern California coasts) based on observational field data and contaminant levels and/or ratios, whereas the false killer whales, harbor porpoises and Alaskan resident killer whales primarily consume prey from less contaminated regions of the eastern North Pacific (e.g., main Hawaiian Islands, Vancouver, British Columbia, Eastern Aleutian Islands). A possible source of PBDEs to the Hawaiian coastal region is effluent from wastewater treatment plants as a number of plants discharge to the coastal ocean in Hawai'i (Friedlander et al., 2005) and appreciable levels of these compounds have been measured previously in wastewater effluents (de Boer et al., 2003; North, 2004). However, PBDEs may be entering this marine ecosystem via other sources that have not yet been identified.

Accumulation of high tissue levels of POPs has been associated with biological and physiological effects in marine mammals (O'Hara and O'Shea, 2001). For example, Kannan et al. (2000) recommended a safe upper PCB threshold concentration of 17,000 ng/g, lipid for PCBs in blubber based on a number of studies that measured various toxicological endpoints (e.g., thyroid hormone concentrations) and PCB concentrations. Three out of nine animals sampled in the current study had ∑PCBs that exceeded this threshold value. Our findings indicate that some of these animals are exposed to PCB levels that may affect their health. In addition to PCBs, these animals are also exposed to other classes of toxic POPs that may increase their risk to adverse effects.

The current study is the first to report blubber concentrations of POPs, including PBDEs, in free-ranging false killer whales, and the first for any free-ranging cetaceans from the Hawaiian Islands. Wide ranges of POP concentrations were measured in these animals, with DDTs and PCBs being the most abundant. Similar to previous cetacean studies, age class and sex influenced the levels of POPs measured in the whales. Interestingly, subadult false killer whales had higher levels of some classes of POPs (e.g., ∑PBDEs,

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dieldrin, HCB) compared to the other sampled animals. Although the POP concentrations measured in the false killer whales in the current study were generally equal to or lower than those reported for false killer whales that stranded in British Columbia or fish-eating marine mammals from the west coast of North America, some of the animals in the current study were exposed to PCB levels that could potentially affect their health. Due to the small size of this whale population and their life history strategies (e.g., long-lived, time to maturation), continued monitoring of POPs is essential in assessing the health and viability of these animals.

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