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Persistent organic pollutant concentrations in blubber of 16 species of cetaceans stranded in the Pacific Islands from 1997 through 2011

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HIGHLIGHTS

- DDTs, PCBs and chlordanes were predominant in Pacific cetaceans.
- POP classes are stable in cetaceans stranded over the last 15 years for this region.
- Contaminants are compared among cetacean species and to other populations globally.

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ABSTRACT

Persistent organic pollutants (POPs) are toxic man-made chemicals that bioaccumulate and biomagnify in food webs, making them a ubiquitous threat to the marine environment. Although many studies have determined concentrations of POPs in top predators, no studies have quantified POPs in stranded cetaceans within the last 30 years around the Hawaiian Islands. A suite of POPs was measured in the blubber of 16 cetacean species that stranded in the tropical Pacific, including Hawai'i from 1997 to 2011. The sample set includes odontocetes ($n = 39$) and mysticetes ($n = 3$). Median (range) contaminant concentrations in ng/g lipid for the most representative species category (delphinids excluding killer whales [$n = 27$]) are: 9650 (44.4–99,100) for $\sum$DDTs, 6240 (40.8–50,200) for $\sum$PCBs, 1380 (6.73–9520) for $\sum$chlordanes, 1230 (13.4–5510) for $\sum$toxaphenes, 269 (1.99–10,100) for $\sum$PBDEs, 280 (2.14–4190) for mirex, 176 (5.43–857) for HCB, 48.1 (<5.42–566) for $\sum$HCHs, 33.9 (<2.42–990) for $\sum$HBCDs, 1.65 (<0.435–11.7) for octachlorostyrene and 1.49 (<2.07–13.1) for pentachlorobenzene. $\sum$PCB concentrations in these Pacific Island cetaceans approach and sometimes exceed proposed toxic threshold values. Backward stepwise multiple regressions indicated the influence of life history parameters on contaminant concentrations when performed with three independent variables (species category, year of stranding, and sex/age class). No temporal trends were noted ($p > 0.063$), but sex/age class influences were evident with adult males exhibiting greater contaminant loads than adult females and juveniles for $\sum$DDT, $\sum$PCBs, $\sum$CHLs, and mirex ($p \leq 0.036$). POP concentrations were lower in mysticetes than odontocetes for many compound classes ($p \leq 0.003$). p,p'-DDE/$\sum$DDTs ratios were greater than 0.6 for all species except humpback whales, suggesting exposure to an old DDT source. These POP levels are high enough to warrant concern and continued monitoring.

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1. Introduction

Persistent organic pollutants (POPs) are a ubiquitous threat to the marine environment and include polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and organochlorine pesticides (OCPs). These organic pollutants originate from a variety of sources including electrical transformers, pesticides, flame retardants and various other household and industrial items. Transport of organic contaminants through the environment occurs via agricultural runoff (Ross and Birnbaum, 2003), atmospheric transport (Gouin et al., 2004), combustion (Jones and de Voogt, 1999) and ocean circulation (Wania and Mackay, 1995). These persistent, lipophilic, and biomagnifying organic compounds in the environment are extremely stable and resistant to degradation (Godduhn and Duffy, 2003).

In the Hawaiian Islands region, high POP concentrations in fish, Hawaiian monk seals (*Monachus schauinslandi*) and false killer whales (*Pseudorca crassidens*) have raised concerns regarding health impacts of these chemicals on cetacean populations (Brasher and Wolff, 2004; Willcox et al., 2004; Ylitalo et al., 2008, 2009). Cetaceans are particularly

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susceptible to accumulation of POPs, primarily in the blubber (Yordy et al., 2010a), because these species have long lifespans, have large fat deposits and occupy top positions in marine food chains. Although a plethora of studies have quantified POP concentrations in many cetacean species worldwide, only two species, rough toothed dolphins (*Steno bredanensis*, stranded) and false killer whales (free-ranging), have been examined for POP concentrations from the Hawaiian Islands (O'Shea et al., 1980; Ylitalo et al., 2009). The only study to examine POPs in stranded cetaceans for this tropical Pacific region (O'Shea et al., 1980) analyzed samples collected over forty years ago (1968 to 1976). Therefore, determining the concentrations and potential effects of these organic contaminants in tropical Pacific cetaceans is timely and important. This study establishes initial baseline concentrations of POPs in sixteen species of cetaceans stranded in the Pacific Islands region mostly from 2006 to 2011 with three samples dating back to 1997, including one novel species never before analyzed (*Indopacetus pacificus*, Longman's beaked whale). In addition, a preliminary assessment of species category differences within this region was conducted using multiple regression statistics that accounted for temporal changes and expected life history effects of sex and age class on POP concentrations.

2. Materials and methods

2.1. Sample collection

Forty-two blubber samples were obtained from Hawai'i Pacific University's (HPU) marine mammal stranding program (NOAA permit #932-1905). These samples originated from cetacean strandings that occurred in the central and western tropical Pacific regions (Fig. 1). Age class was estimated based on animal weight, total length and maturation of reproductive organs. In order to minimize the impact of postmortem degradation, the samples used in this study were from stranded cetaceans that were code 1 (alive then euthanized) or code 2 (considered freshly dead) (Hofman, 1991). Blubber was sampled from these dead stranded animals at time of necropsy, wrapped in aluminum foil and stored at −80 °C until analyzed for POPs at the National Institute of Standards and Technology (NIST), Hollings Marine Laboratory, Charleston, South Carolina. The sample set represents sixteen cetacean species stranded in the tropical Pacific over 15 years (most from 2006 to 2011 with one from 1997, one from 1998, and one from 2000), encompassing different age classes and sexes (Table 1).

2.2. Analytical methods

2.2.1. Persistent organic pollutants

2.2.1.1. Sample preparation, extraction and cleanup. Blubber subsamples (≈1.0 g) of full depth, but not exceeding a depth of 7 cm, were weighed and manually homogenized using a razor blade in a beaker in a manner to prevent loss of lipids. The blubber was combined with sodium sulfate, transferred to a pressurized fluid extraction (PFE) cell and spiked gravimetrically with internal standard. The internal standard solution contained ^{13}C-labeled PCB congeners (28, 52, 77, 126, 169, 118, 153, 180, 194, 206), 6-F-PBDE 47, PBDE 104, 4′-F-PBDE 160, 4′-F-PBDE 208, ^{13}C-labeled PBDE 209, ^{13}C-labeled pesticides (hexachlorobenzene (HCB), *trans*-chlordane, *trans*-nonachlor, oxychlordane, *p,p*′-DDE, *p,p*′-DDD, *p,p*′-DDT), ^{13}C-labeled methyl-triclosan and ^{13}C-labeled α-, β- and γ-hexabromocyclododecanes (HBCDs). POPs were extracted from blubber samples with dichloromethane using PFE. Total extractable organic content (TEO), as a proxy for lipid content, was determined by removing 50% of the extract gravimetrically, allowing it to dry in an aluminum pan and weighing dried residue. Remaining extracts were cleaned up using size exclusion chromatography with additional clean-up and fractionation of the samples by acidified silica columns in an automated solid phase extraction system. The majority of POPs eluted into the first fraction (F1) followed by the HBCDs in the second fraction (F2).

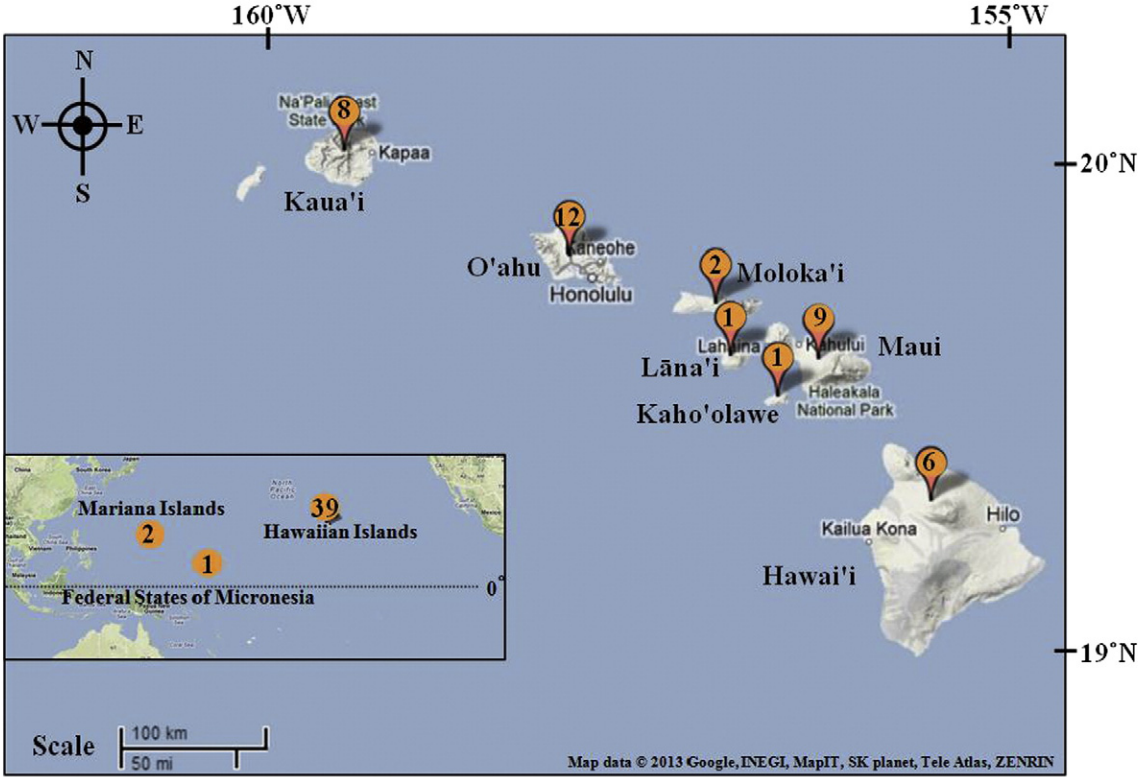

Fig. 1. Location of stranded cetaceans that were analyzed for persistent organic pollutants in the tropical Pacific from 1997 to 2011. Numbers indicate number of individuals that stranded on each island.

Table 1
Stranded Pacific Island cetacean sample list. Code 1 (euthanized) and code 2 (freshly dead) were included in this analysis. Abbreviations are as follows: M = male, F = female, U = unknown, C = calf, J = juvenile, A = adult.

Species	Species abbreviation	Common name	Sample ID	Stranding location	Year of stranding	Species category	Sex	Age class
Feresa attenuata	Fa	Pygmy killer whale	SLP200601	O'ahu	2006	Killer	F	A
F. attenuata	Fa	Pygmy killer whale	KW2009006	Maui	2009	Killer	M	A
Indopacetus pacificus	Ip	Longman's beaked whale	KW2010005	Maui	2010	Beaked	M	J
Kogia breviceps	Kb	Pygmy sperm whale	15320	Kaua'i	2000	Sperm	M	J
K. breviceps	Kb	Pygmy sperm whale	KW2007003	Maui	2007	Sperm	M	A
Kogia sima	Ks	Dwarf sperm whale	KW2009012	Kaua'i	2009	Sperm	M	A
Megaptera novaeangliae	Mn	Humpback whale	15063	Lana'i	1998	Baleen	F	C
M. novaeangliae	Mn	Humpback whale	KW2008003	Maui	2008	Baleen	F	C
M. novaeangliae	Mn	Humpback whale	KW2009002	Kaua'i	2009	Baleen	F	C
Mesoplodon densirostris	Md	Blainville's beaked whale	KW2010012	Maui	2010	Beaked	M	J
Orcinus orca	Oo	Killer whale	KW2008010	Kaua'i	2008	Killer	M	J
Peponocephala electra	Pe	Melon-headed whale	KW2010017	O'ahu	2010	Delphinid	M	J
P. electra	Pe	Melon-headed whale	KW2011002	O'ahu	2011	Delphinid	M	C
P. electra	Pe	Melon-headed whale	KW2011009	Kosrae	2011	Delphinid	F	C
P. electra	Pe	Melon-headed whale	KW2011011	Maui	2011	Delphinid	M	A
Physeter macrocephalus	Pm	Sperm whale	KW2011008	O'ahu	2011	Sperm	F	C
Pseudorca crassidens	Pc	False killer whale	KW2010019	Moloka'i	2010	Killer	F	A
Stenella attenuata	Sa	Spotted dolphin	KW2008005	O'ahu	2008	Delphinid	M	J
S. attenuata	Sa	Spotted dolphin	KW2009015	Hawai'i	2009	Delphinid	M	A
S. attenuata	Sa	Spotted dolphin	KW2010011	Guam	2009	Delphinid	M	C
Stenella coeruleoalba	Sc	Striped dolphin	12470	O'ahu	1997	Delphinid	M	A
S. coeruleoalba	Sc	Striped dolphin	KW2008006	Hawai'i	2008	Delphinid	F	C
S. coeruleoalba	Sc	Striped dolphin	KW2009008	O'ahu	2009	Delphinid	M	J
S. coeruleoalba	Sc	Striped dolphin	KW2009009	Maui	2009	Delphinid	F	A
S. coeruleoalba	Sc	Striped dolphin	KW2009011	Maui	2009	Delphinid	F	C
S. coeruleoalba	Sc	Striped dolphin	KW2010008	Hawai'i	2010	Delphinid	M	A
Stenella longirostris	Sl	Spinner dolphin	KW2007004	O'ahu	2007	Delphinid	M	J
S. longirostris	Sl	Spinner dolphin	KW2007005	O'ahu	2007	Delphinid	F	A
S. longirostris	Sl	Spinner dolphin	KW2008002	Kaua'i	2008	Delphinid	F	A
S. longirostris	Sl	Spinner dolphin	KW2008004	Maui	2008	Delphinid	M	A
S. longirostris	Sl	Spinner dolphin	KW2008009	Hawai'i	2008	Delphinid	F	A
S. longirostris	Sl	Spinner dolphin	KW2008011	Hawai'i	2008	Delphinid	F	A
S. longirostris	Sl	Spinner dolphin	KW2009004	O'ahu	2009	Delphinid	F	C
S. longirostris	Sl	Spinner dolphin	KW2010006	O'ahu	2010	Delphinid	M	J
S. longirostris	Sl	Spinner dolphin	KW2011013	Hawai'i	2011	Delphinid	M	C
S. longirostris	Sl	Spinner dolphin	KW2011018	O'ahu	2011	Delphinid	F	C
Steno bredanensis	Sb	Rough-toothed dolphin	KW2011021	Kaua'i	2011	Delphinid	M	A
Tursiops truncatus	Tt	Bottlenose dolphin	KW2009007	Kaho'olawe	2009	Delphinid	U	A
T. truncatus	Tt	Bottlenose dolphin	KW2011001	Kaua'i	2011	Delphinid	F	C
T. truncatus	Tt	Bottlenose dolphin	KW2011007	Kaua'i	2011	Delphinid	M	A
Ziphius cavirostris	Zc	Cuvier's beaked whale	KW2008008	Moloka'i	2008	Beaked	M	J
Z. cavirostris	Zc	Cuvier's beaked whale	KW2011016	Saipan	2011	Beaked	M	A

2.2.1.2. GC/MS analysis. F1 samples were injected three different times with volumes ranging from 1 μL to 20 μL, depending upon which injector was used and the relative concentration in each sample, onto a gas chromatograph mass spectrometer (GC/MS) for varying target constituents. The first method used a programmable temperature vaporization (PTV) inlet operated in the solvent vent mode onto a 5 m × 0.25 mm Restek Siltek guard column (Bellefonte, PA) connected to a 0.18 mm × 30 m Agilent DB-5MS capillary column, 0.18 μm film thickness. PCBs, dichlorodiphenyl–dichloroethane and related compounds (DDTs), octachlorostyrene (OCS), lower-brominated PBDEs, hexachlorocyclohexanes (HCHs), HCB, mirex, oxychlordane, heptachlor and pentachlorobenzene (PeCB) were quantified from this injection using an electron impact (EI) source. With the same inlet, guard and analytical column, the majority of chlordanes (CHLs) and all toxaphenes were quantified using a negative chemical ionization (NCI) source. PBDEs, including the higher-brominated congeners, were quantified from a cool, on-column injection onto a 0.18 mm × 10 m DB-5MS, 0.18 μm film thickness column with a NCI source.

2.2.1.3. LC/MS/MS analysis. For detection of HBCD isomers (α, β, γ), F2 extracts were injected at a volume of 20 μL onto an Agilent Eclipse XDB-C18 guard (4.6 mm × 12.5 mm × 5 μm) and analytical column (3.0 mm × 150 mm × 3.5 μm) connected to an API 4000 (Applied Biosystems, Foster City, CA) with electrospray ionization (ESI) source for liquid chromatography tandem mass spectrometry (LC/MS/MS). An isomer separation method developed for a quaternary HPLC pump (Yu et al., 2008) was modified and optimized for use with a binary pump. Solvent A of the mobile phase was 2.5 mmol/L ammonium acetate in 12.5% water in methanol (volume fraction) and solvent B was acetonitrile. Flow was 0.3 mL/min and the following gradient elution was used: ramped from initial conditions of 90% solvent A to 67% solvent A over the first 12 min, held at 67% solvent A for 3 min, ramped to 0% solvent A over the next 5 min, held at 0% solvent A for 3 min, ramped to 90% solvent A over the next 5 min and held at 90% solvent A for 5 min.

2.2.2. QA/QC and quantification

Samples of Standard Reference Material (SRM) 1945 Organics in Whale Blubber were analyzed as controls. These samples, laboratory procedural blanks and calibration solutions were extracted, processed and analyzed concurrently with the sample set. Six-point calibration curves ranged from 0.05 ng to 1600 ng of the following solutions: SRM 2261 Chlorinated Pesticides in Hexane, SRM 2262 Chlorinated Biphenyl Congeners in Isooctane, SRM 2274 PCB Congener Solution-II in Isooctane, SRM 2275 Chlorinated Pesticide Solution-II in Isooctane, 4 toxaphene congeners (Cambridge Isotope Laboratory), octachlorostyrene (Accustandard), α-, β-, γ-HBCDs (Accustandard) and additional solutions containing 47 PCB and 28 PBDE congeners. POP concentrations were determined using the slope of at least a three point calibration curve that bracketed the peak area ratios observed in each sample. Reporting limits (RL) were calculated as the greater of the nanogram amount of the lowest detectable calibration point or (mean + 3 standard deviations [SD]) nanograms in procedural blanks, all divided by extracted sample mass.

2.2.3. Statistical analysis

Mean, median and standard deviations were generated using the open-source statistics program R (http://cran.r-project/org [R Development Core Team, 2005]) NADA package through Kaplan–Meier, maximum likelihood estimation (MLE) or regression on order statistical (ROS) models as recommended for left censored data (Helsel, 2005). All data were lipid-normalized (ng/g lipid) and summed for the following compound classes: $\sum$ PCBs (polychlorinated biphenyls 8, 18, 28, 29, 31, 44, 45, 49, 50, 52, 56, 63, 66, 70, 74, 79, 82, 87, 92, 95, 99, 101, 104, 105, 106, 107, 110, 112, 114, 118, 119, 121, 127, 128, 130, 132, 137, 138, 146, 149, 151, 153, 154, 156, 157, 158, 159, 163, 165, 166, 167, 170, 172, 174, 175, 176, 177, 178, 180, 183, 185, 187, 188, 189, 191, 193, 194, 195, 196, 197, 199, 200, 201, 202, 203, 205, 206, 207, 208, 209), $\sum$ PBDEs (polybrominated diphenyl ethers 17, 25, 28, 30, 33, 47, 49, 66, 71, 75, 85, 99, 100, 116, 119, 138, 153, 154, 155, 156, 181, 183, 190, 191, 203, 205, 206, 209), $\sum$ DDTs (*o,p′*- and *p,p′*-DDE, *o,p′*- and *p,p′*-DDD, *o,p′*- and *p,p′*-DDT), $\sum$ CHLs (*cis*-chlordane, *cis*-nonachlor, heptachlor, oxychlordane, *trans*-chlordane, *trans*-nonachlor), $\sum$ HBCDs (α-, β- and γ-hexabromocyclododecanes), $\sum$ HCHs (α-, β- and γ-hexachlorocyclohexanes), $\sum$ TOXs (Parlars 26, 32, 50, 62), mirex, hexachlorobenzene (HCB), octachlorostyrene (OCS) and pentachlorobenzene (PeCB).

In order to examine species differences within this region, statistical tests required taking into account the 15 year time span of sample collection as well as the mix of both gender and age class. Data were tested for normality using Shapiro–Wilk goodness of fit test and when data from a particular contaminant class were not normally distributed they were log-transformed to meet the assumptions of normality (HCB was square root transformed and *p,p′*-DDE/$\sum$ DDTs ratio was transformed with the arcsin of the square root). Backward stepwise multiple regression analyses were performed in JMP 10.0 (SAS Institute, Cary, NC) on concentrations of ten contaminant classes and the ratio of *p,p′*-DDE/$\sum$ DDTs. The backward stepwise multiple regressions were run with the following independent variables: species category, year of stranding and sex/age class. Sex/age class represents sexual maturity within three categories and was coded as categorical data: juvenile (neonates, calves and juveniles for both sexes), adult females and adult males. Species categories were chosen based on family groupings indicated in the National Audubon Society Guide to Marine Mammals of the World (Reeves et al., 2008) as well as trophic ecology aspects and was coded as categorical data. The five phylogenetic categories are baleen whales ($n = 3$), beaked whales ($n = 4$), sperm whales ($n = 4$), delphinids not including killer whales ($n = 27$) and killer whales ($n = 4$). Killer whales included three species (Fa, Oo, Pc), delphinids included six species (Pe, Sa, Sb, Sc, Sl, and Tt), beaked whales included three species (Ip, Md, and Zc), sperm whales included three species (Kb, Ks, and Pm) and baleen whales included one species (Mn) (Table 1 lists the species abbreviations). The year of stranding was the only independent variable within the regressions coded as continuous data. When concentrations were below the RL, half of the RL was used for the multiple regressions and significant statistical findings were verified in the R NADA package.

3. Results and discussion

3.1. POP concentrations and multiple regressions

POP concentrations measured in individual animals are shown in the Supplementary data (Table A.1) and are shown for each species in Table 2. In Table 2, the highest to lowest contaminant classes are shown from the left to right (top congeners are listed in parentheses here): $\sum$ DDTs (*p,p′*-DDE > *p,p′*-DDD + *o,p′*-DDT > *p,p′*-DDT > *o,p′*-DDE > *o,p′*-DDD), $\sum$ PCBs (153 + 132 > 138 > 101 > 149 > 187), $\sum$ CHLs (*trans*-nonachlor > *cis*-chlordane > *cis*-nonachlor > oxychlordane > *trans*-chlordane), $\sum$ TOXs (Parlars 50 > 26 > 62 > 32), $\sum$ PBDEs (47 > 100 > 99 > 154 > 155), mirex, HCB, $\sum$ HCHs (predominantly β-HCH), $\sum$ HBCDs (predominately α-HBCD) and OCS. POPs were detected in 100% of the samples analyzed for $\sum$ PCBs, $\sum$ TOXs, $\sum$ PBDEs; 97.6% for $\sum$ DDTs, $\sum$ CHLs, HCB; 95.2% for mirex, $\sum$ HBCDs; 85.7% for OCS and 83.3% for $\sum$ HCHs and 16.7% for PeCB. Since most samples had non-detectable PeCB (>80%), they were reported only in Table A.1. Species are ranked from greatest to least POP concentrations from top to bottom in Table 2. The killer whale (*Orcinus orca*), false killer whale and pygmy killer whale (*Feresa attenuata*) have the greatest concentrations, while humpback whales (*Megaptera novaeangliae*) have the lowest concentrations. Other delphinid, beaked and sperm whale species generally fell between the killer and baleen whales.

We investigated species category differences while taking into account year of stranding and the various age classes and sexes using multiple regression statistics (Table 3). Statistics such as these have been used in other studies assessing POP concentrations in cetaceans (Tuerk et al., 2005; Hoguet et al., 2013); however, those previous studies focused on a single species rather than grouping several species into categories. Because of low or uneven representation of certain species and life history parameters, this complex data set is not ideal for drawing broad conclusions about these groups overall, so we utilized this approach to help visualize emerging trends and put our observations in perspective with previous reports.

Because multiple regression is sensitive to small sample sizes, regression models were performed with five species categories in an attempt to group similar species and avoid small sample sizes. Sample KW2009007 (bottlenose dolphin, *Tursiops truncatus*) was excluded from these statistical tests due to unknown sex/age class. All regression models for each contaminant class, except OCS, were significant ($p \leq 0.035$; Table 3). Species category accounted for a significant amount of the variability for all POP classes except OCS (Table 3). Baleen whales (in this study only juvenile humpback whales) showed significantly lower contaminant concentrations than the other four odontocete cetacean categories for five compound classes ($\sum$ DDTs, $\sum$ PCBs, $\sum$ CHLs, mirex and $\sum$ HBCDs) and the *p,p′*-DDE/$\sum$ DDTs ratio (Table 3, Fig. 2). Killer whales had higher contaminant concentrations compared to other cetacean categories for $\sum$ DDTs, $\sum$ PCBs, $\sum$ CHLs, mirex and HCB. The pattern of $\sum$ HCHs was different with the baleen whale category having the highest concentrations, statistically higher than sperm and beaked whales ($p = 0.0008$) but not significantly higher concentrations than killer whales and delphinids ($p = 0.0891$) (Table 3). This is not surprising because more volatile contaminants, like HCHs, are more readily transported to higher latitudes (Scheringer, 2008) where this population of humpback whales feeds (Allen and Angliss, 2013). No significant temporal trends were observed, but sex/age class did influence POP concentrations (Table 3).

3.2. Species differences

Species differences observed in this study were expected because of suspected trophic level differences of the species categories. Although trophic levels have not specifically been assigned to cetaceans in this tropical Pacific region, enough evidence from behavioral and necropsy observations (Baird et al., 2006, 2008; Clarke and Young, 1998; Reeves et al., 2009; West et al., 2009) and generalizations about diet composition of many of these species from other regions (Pauly et al., 1998) allowed us to predict that the trophic level order would be: killer > delphinid > (beaked = sperm) > baleen. Most POP concentrations follow this pattern (Table 3, Fig. 2). Killer whales are known to feed at

Table 2
Median (range) and mean ± standard deviation mass fractions (ng/g lipid) of persistent organic pollutants in blubber of 16 species of stranded Pacific Island cetaceans.

Common name	*n*	Total extractable organics (% lipid)	$\sum$DDTs	$\sum$PCBs	$\sum$CHLs	$\sum$TOXs	$\sum$PBDEs	Mirex	HCB	$\sum$HCHs	$\sum$HBCDs	OCS	*p,p′*-DDE/ $\sum$DDTs
Killer whale	1	38.8	171,000	93,200	13,600	6890	938	6330	580	187	213	3.82	0.943
False killer whale	1	46.5	28,200	26,200	4900	2610	1650	1080	364	133	353	5.60	0.830
Pygmy killer whale	2	(24.9–43.8)	(2350–58,200)	(1800–46,300)	(498–7590)	(518–6280)	(45.1–521)	(136–1820)	(145–696)	(<5.86–52.4)	(48.4–248)	(2.36–4.83)	(0.678–0.911)
Melon-headed whale	4	29.8 (3.00–55.8) 29.6 ± 27.5	8860 (7240–99,100) 31,000 ± 45,400	4290 (3440–50,200) 15,600 ± 23,100	1100 (698–9520) 3100 ± 4290	1780 (970–5510) 2510 ± 2040	246 (104–1040) 409 ± 432	210 (169–4190) 1190 ± 2000	343 (141–435) 315 ± 139	94.7 (11.9–276) 119 ± 112	53.6 (<2.42–224) 108 ± 85.4	3.03 (<2.49–7.22) 4.39 ± 2.10	0.843 (0.802–0.924) 0.853 ± 0.054
Striped dolphin	6	34.0 (16.4–48.2) 33.7 ± 10.9	19,700 (13,600–27,500) 20,000 ± 5510	11,600 (6240–24,000) 13,800 ± 7260	2540 (1450–5740) 2830 ± 1570	2290 (1540–5430) 2880 ± 1400	248 (94.8–452) 258 ± 139	741 (280–1170) 725 ± 293	380 (154–857) 419 ± 251	230 (118–566) 281 ± 184	162 (55.6–990) 287 ± 355	3.96 (1.01–11.7) 4.90 ± 3.71	0.834 (0.775–0.890) 0.831 ± 0.039
Rough-toothed dolphin	1	36.0	16,100	13,800	1950	1230	192	829	98.0	33.4	80.0	1.93	0.907
Bottlenose dolphin	3	33.7 (17.9–62.2) 37.9 ± 22.4	11,500 (9650–23,800) 15,000 ± 7700	7689 (7490–20,300) 11,800 ± 7340	1730 (980–2720) 1810 ± 873	1650 (759–2060) 1490 ± 665	1020 (927–1260) 1070 ± 172	500 (285–1030) 605 ± 383	245 (144–745) 378 ± 322	171 (27.9–210) 136 ± 96.0	101 (14.2–130) 81.7 ± 60.3	2.18 (<0.450–8.33) 4.23 ± 4.10	0.917 (0.782–0.928) 0.876 ± 0.081
Longman's beaked whale	1	85.3	12,000	7610	1690	1430	118	139	360	121	62.4	7.10	0.825
Pygmy sperm whale	2	(51.4–62.8)	(3260–13,000)	(2310–8310)	(522–1540)	(313–660)	(16.1–47.7)	(38.0–189)	(156–299)	(<4.07–47.8)	(10.7–27.3)	(4.08–6.17)	(0.800–0.877)
Spotted dolphin	3	54.5 (21.0–67.6) 47.7 ± 24.0	5340 (44.4–30,900) 12,100 ± 16,500	2380 (40.8–11,400) 4610 ± 6000	643 (6.73–1900) 850 ± 963	680 (13.4–1090) 595 ± 543	54.4 (1.99–178) 78.1 ± 90.4	152 (2.14–889) 348 ± 475	72.4 (5.43–159) 78.9 ± 77.0	28.8 (1.68–35.0) 21.8 ± 17.7	29.6 (0.510–33.9) 21.3 ± 18.2	0.185 (<1.33–1.65) 0.673 ± 0.976	0.779 (0.734–0.917) 0.810 ± 0.095
Cuvier's beaked whale	2	81.6 (81.0–82.1)	6180 (3280–9070)	4250 (3130–5360)	640 (545–734)	250 (209–290)	59.4 (53.6–65.2)	104 (82.2–125)	(55.7–116)	8.77 (<3.10–8.77)	56.8 (27.3–86.3)	2.30 (1.83–2.75)	0.784 (0.720–0.848)
Spinner dolphin	10	54.2 (13.8–77.0) 50.8 ± 21.9	2530 (267–15,700) 4190 ± 4890	2090 (427–9730) 3280 ± 3290	533 (77.4–2330) 694 ± 706	611 (66.2–2570) 770 ± 734	559 (46.4–10,100) 1720 ± 3040	107 (21.3–573) 170 ± 197	134 (14.3–235) 127 ± 76.0	29.0 (<5.42–106) 40.5 ± 27.8	20.0 (5.67–220) 38.2 ± 64.6	1.08 (<0.435–2.69) 1.13 ± 0.777	0.777 (0.682–0.919) 0.786 ± 0.072
Sperm whale	1	39.9	4000	1470	445	313	27.2	12.0	108	18.1	12.7	1.38	0.839
Dwarf sperm whale	1	75.6	3030	1900	313	113	25.5	106	79.8	<3.24	17.2	1.30	0.855
Blainville's beaked whale	1	76.8	2480	1450	309	169	30.4	46.2	53.7	8.07	21.6	1.95	0.731
Humpback whale	3	35.5 (0.183–58.2)[a] 31.3 ± 29.2	94.7 (<3390–111) 103 ± 13.9	104 (96.8–661) 287 ± 324	55.9 (<574–60.6) 58.3 ± 4.13	85.0 (60.1–657) 267 ± 338	7.05 (2.08–39.2) 16.1 ± 20.2	<103 (<0.507–2.81) N/A	115 (<336–167) 141 ± 45.0	114 (<1420–157) 135 ± 37.4	0.717 (<36.1–0.859) 0.788 ± 0.123	0.574 (<156–0.608) 0.591 ± 0.029	(0.489–0.521)[b] N/A
All samples	42	46.2 (0.183–85.3) 45.8 ± 22.6	8150 (<3390–171,000) 16,600 ± 30,300	5120 (40.8–93,200) 10,500 ± 17,200	821 (<574–13,600) 1890 ± 2730	864 (13.4–6890) 1500 ± 1720	185 (1.99–10,100) 657 ± 1600	152 (<0.507–6330) 587 ± 1160	159 (<336–857) 237 ± 203	47.8 (<3.10–566) 95.1 ± 122	31.7 (<2.42–990) 95.6 ± 167	1.93 (<0.435–11.7) 2.83 ± 2.61	0.825 (0.489–0.943) 0.800 ± 0.110[c]

[a] The sample with extremely low lipid content was confirmed by analyzing an additional subsample.
[b] One sample was excluded because it was below the reporting limit for $\sum$DDTs ($n = 2$).
[c] One sample was excluded because it was below the reporting limit for $\sum$DDTs ($n = 41$).

Table 3
Species differences, temporal trends and life history influences on POP concentrations in stranded tropical Pacific cetaceans. *p*-Values in bold represent statistical significance ($p < 0.050$). *p*-Values > 0.100 indicate elimination of that independent variable from the model. Where multiple *p*-values are reported, the first *p*-value corresponds to the first "<" sign in the parenthetical result description, the second *p*-value corresponds to the next "<" and so forth. Sex/age classes are: J = juvenile, AF = adult female, AM = adult male.

Compounds	Model	Species category	Year of stranding	Sex/age class
$\sum$DDTs	**0.0002**	**0.0004**, **0.0368** (baleen < sperm, beaked, delphinid < killer)	>0.100	**0.0162** (J, AF < AM)
$\sum$PCBs	<**0.0001**	**0.0001**, **0.0152** (baleen < sperm, beaked, delphinid < killer)	>0.100	**0.0078** (J, AF < AM)
$\sum$CHLs	**0.0013**	**0.0031**, **0.0205** (baleen < sperm, beaked, delphinid < killer)	>0.100	**0.0311** (J, AF < AM)
$\sum$TOXs	**0.0024**	**0.0024** (baleen, sperm, beaked < delphinid, killer)	>0.100	>0.100
$\sum$PBDEs	<**0.0001**	<**0.0001** (baleen, sperm, beaked < delphinid, killer)	>0.100	>0.100
Mirex	<**0.0001**	<**0.0001**, **0.0298**, **0.0073** (baleen < sperm, beaked < delphinid < killer)	>0.100	0.0863, **0.0361** (J < **AF** < **AM**)
HCB	**0.0351**	**0.0351** (beaked, baleen, sperm, delphinid < killer)	>0.100	>0.100
$\sum$HCHs	**0.0034**	**0.0008**, 0.0891 (**sperm**, **beaked** < **killer**, **delphinid** < baleen)	>0.100	>0.100
$\sum$HBCDs	**0.002**	**0.0006**, 0.0508 (**baleen** < **sperm**, **delphinid**, **beaked** < killer)	>0.100	>0.100
OCS	0.0626	>0.100	0.0626 (decreasing)	>0.100
p,p′-DDE/$\sum$DDTs	<**0.0001**	**0.0010** (baleen < beaked, delphinid, sperm, killer)	>0.100	**0.0008** (AF, J < AM)

the highest trophic level, feeding on other marine mammals, fish and cephalopods (Baird et al., 2006). Hawaiian killer whales (*O. orca*), in particular, have been known to feed on cephalopods (either directly or indirectly) as well as humpback whales (Baird et al., 2006). False killer whales are known to feed on large game fish predominately in surface waters, even preying on tuna hooked in long line fisheries (Baird et al., 2008; Reeves et al., 2009). Therefore, it was not surprising that this species category had the highest statistically significant concentrations of $\sum$DDTs, $\sum$PCBs, $\sum$CHL, mirex and HCB, as well as the highest non-significant concentrations of $\sum$HBCDs. They were tied for highest with delphinids in $\sum$PBDEs and $\sum$TOXs, and were third highest in $\sum$HCHs. The other delphinids eat mostly high-trophic level, epipelagic fish, mesopelagic fish and some squid (Pauly et al., 1998; Clarke and Young, 1998). Hawaiian sperm and beaked whales are known to feed predominately on mesopelagic squid and occasionally on small fishes as indicated by stomach content analyses (Clarke and Young, 1998). Similar observations have been made in pygmy sperm whale (*Kogia breviceps*) stomachs (West et al., 2009). Species feeding at these depths may also ingest prey with intermediate to high contaminant loads since mesopelagic fish and deep sea squid have been considered sinks for OCPs in the north Pacific deep oceans (Takahashi et al., 2010). Humpback whales that winter in Hawai'i are known to feed on small fishes and euphausiids in the North Pacific (Allen and Angliss, 2013), which is a lower trophic level and different foraging grounds than species in the other four categories. The contaminant concentration differences among species categories (Table 3, Fig. 2) are very likely related to these trophic level differences, but could also be influenced by latitudinal migration patterns and depth of foraging.

Contaminant profiles were investigated among the species categories (Fig. 3). The most obvious difference among species categories is that baleen whales have higher proportions of lower chlorinated PCBs (tetra and pentachlorobiphenyls) as well as lower brominated PBDEs (PBDE 47) when compared to the other cetacean categories. Baleen whales also showed greater proportions of DDD, the more hydrophilic metabolite of DDT and less of the long term storage compound *p,p′*-DDE. These differences in contaminant profiles are most likely attributed to location of foraging grounds and age class of the sampled humpback whales. Humpback whales wintering in Hawai'i return to their feeding grounds in higher latitudes along the western coasts of U.S. and Canada, in the Gulf of Alaska and Bering Sea (Allen and Angliss, 2013). More volatile and smaller compounds, like HCHs, HCB, lower chlorinated PCBs and lower brominated PBDEs, as well as DDD compared to DDT are more readily transported atmospherically to these higher latitudes (Scheringer, 2008), providing a greater proportion of these compounds in humpback whales. Additionally, all three humpback samples were from juveniles, so their POP profiles are more likely influenced by these lower molecular weight and less hydrophobic compounds that are more readily transferred maternally (Yordy et al., 2010b).

3.3. Sex/age class relationships

Using the backward stepwise multiple regression, the gender and age class influences on POP concentrations in this study (Table 3) are consistent with previous reports. Adult males exhibited higher concentrations than adult females and juveniles for the top three contaminant

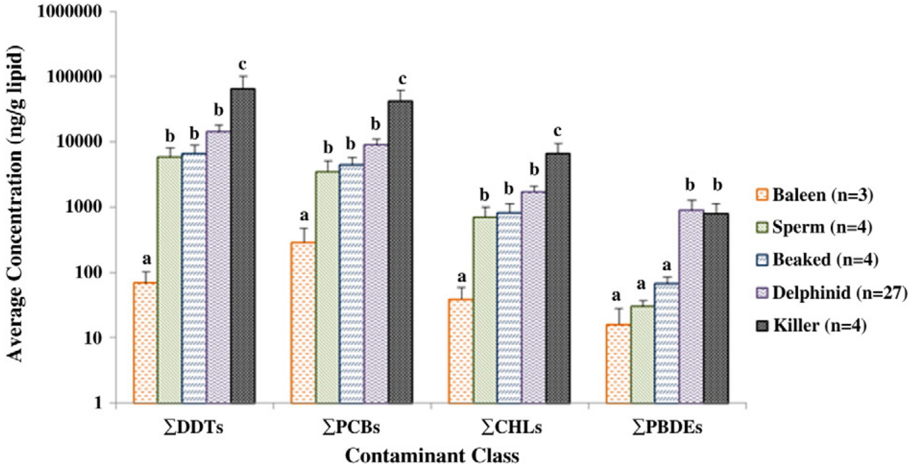

Fig. 2. Trends in selected high prevalence POP classes among stranded cetacean species groups in the tropical Pacific. Different letters above columns indicate significant differences between cetacean categories as indicated by backward stepwise multiple regression, standard error is graphed.

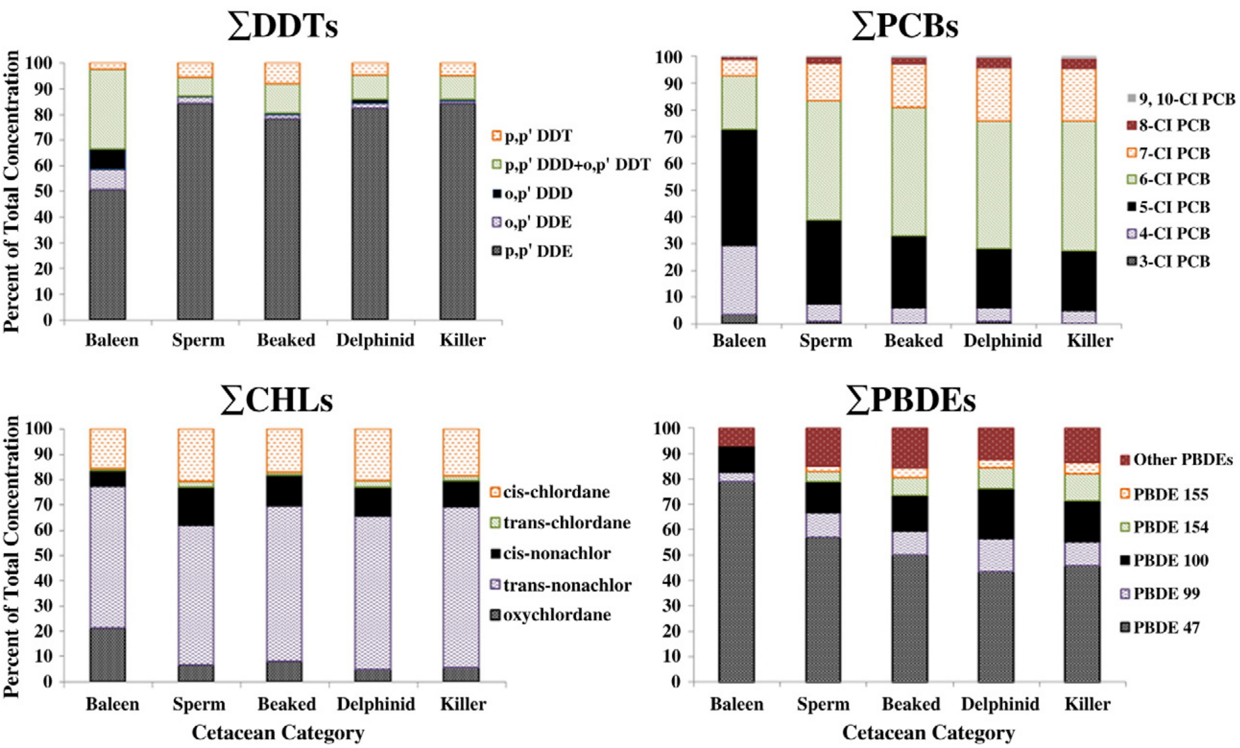

Fig. 3. Contaminant composition profiles across five species categories (baleen, sperm, beaked, delphinid and killer) of stranded cetaceans from the Pacific Islands.

classes ($\sum$DDTs, $\sum$PCBs and $\sum$CHLs), mirex and *p,p′*-DDE/$\sum$DDT ratio. This sex and age class difference is likely due to maternal transfer of POPs from adult females to their offspring (Yordy et al., 2010b; Hansen et al., 2004; Krahn et al., 2009), while males and females prior to their first pregnancy bioaccumulate POPs through time (Marsili et al., 1995).

3.4. Geographic comparisons of POP concentrations

Studies chosen for comparison to the current data were predominantly from the Pacific Ocean, represent fresh frozen marine mammal blubber samples and utilized POPs quantification methodology with clear quality control/assurance. To reduce the complexity of comparisons, only adult male delphinids including killer whales from this study were compared to males published from other studies (Table 4). In general, cetaceans from this study had similar POP concentrations compared to cetaceans from Japan (Kajiwara et al., 2006), but higher levels than cetaceans from the Philippines (Kajiwara et al., 2006) and Hawaiian monk seals (Lopez et al., 2012) (Table 4). Ranges of $\sum$PCBs, $\sum$DDTs, and $\sum$PBDEs were higher in cetaceans from this study compared to those from Alaska, with $\sum$CHLs being similar in both regions (Hoguet et al., 2013). Ranges overlapped with free-ranging false killer whales from Hawai'i (Ylitalo et al., 2009).

Aside from compounds in Table 4, it is interesting to note high concentrations of the HBCD flame retardant in stranded cetaceans from this study. The concentration range (<2.42–990 ng/g lipid) overlapped with marine mammals from European regions of known elevated use of technical HBCD (Law et al., 2006) as well as the western Pacific Ocean (e.g., South China and Japan) (Lam et al., 2009), but was higher than those reported from California sea lions (*Zalophus californianus*) (<0.4–96 ng/g lipid) (Stapleton et al., 2006).

Taking a closer look at humpback whales, POP concentrations in the stranded calves from this study (Table 2) overlap with or are on the lower end of the ranges from free-ranging adult males in Alaska (Elfes et al., 2010). Compared to adult males sampled in California, the Hawaiian calves were similar in $\sum$HCHs but on the lower end of the

Table 4
A comparison of persistent organic pollutant mass fractions (ng/g lipid) in the blubber of odontocetes from the Northern Hemisphere of the Pacific Ocean. Adult male concentrations are reported for the Central Pacific and male concentrations representing unknown age classes are reported from West and North Pacific.

	Central Pacific			West Pacific		North Pacific
	Main Hawaiian Islands, USA	Main Hawaiian Islands, USA	Hawaiian Islands, USA	Philippines	Japan	Eastern Chukchi Sea, Alaska, USA
Species	Seven delphinid species Various	False killer whale (*Pseudorca crassidens*)	Hawaiian monk seal (*Monachus schauinslandi*)	Spinner dolphin (*Stenella longirostris*)	Melon-headed whale (*Peponocephala electra*)	Beluga whale (*Delphinapterus leucas*)
Sample number	Adult male ($n = 8$)	Adult male ($n = 2$)	Adult male ($n = 15$)	Male ($n = 3$)	Male ($n = 5$)	Male ($n = 26$)
Collection year	1997–2011	2008	2000–2010	1996	2001	1989–2006
	Median (range)	Median (range)	Mean ± SE	Mean (range)	Mean (range)	Median (range)
$\sum$PCBs	17,050 (7690–50,200)	33,000 (33,000–33,000)	1800 ± 400	3600 (2600–5400)	24,000 (15,000–30,000)	4860 (2190–9070)
$\sum$DDTs	23,400 (9730–99,100)	63,000 (43,000–83,000)	690 ± 150	16,000 (15,000–17,000)	27,000 (18,000–33,000)	4310 (1350–8240)
$\sum$PBDEs	395 (94.8–1560)	1200 (780–1600)	120 ± 39	36.0 (20–64)	320 (300–340)	12.8 (4.33–32.2)
$\sum$CHLs	2335 (1420–9520)	4500 (4100–4900)	190 ± 100	540 (340–920)	4100 (3600–5400)	3830 (1300–6720)
Reference	This study	Ylitalo et al. (2009)	Lopez et al. (2012)	Kajiwara et al. (2006)	Kajiwara et al. (2006)	Hoguet et al. (2013)

range for the other POPs (Elfes et al., 2010). The lower concentrations in Hawaiian samples are likely caused by sex and age class differences, rather than geographical differences because humpback whales wintering in Hawai'i are part of the Central North Pacific stock that feed off of Alaska (Allen and Angliss, 2013).

A juvenile male killer whale with the highest overall concentration of POPs measured in this study (Table 2) was compared to free-ranging Southern resident males (ages 4–21 years) from the North Pacific (British Columbia through Washington state) (Krahn et al., 2009). The stranded Hawaiian killer whale had higher concentrations of $\sum$PCBs, $\sum$DDTs and $\sum$CHLs and similar or lower concentrations of $\sum$PBDEs, $\sum$HCHs and HCB than the Southern resident population (Krahn et al., 2009). Another highly contaminated animal was a 24-year old female false killer whale (Table 2). She had concentrations of $\sum$PCBs and $\sum$DDTs well above the range of other free-ranging adult females from the same endangered insular population (Ylitalo et al., 2009). Female false killer whales are hypothesized to reach maturity around 8 to 14 years and live up to about 60 years (Ylitalo et al., 2009), so this animal was likely sexually mature. These two highly contaminated animals were both emaciated and showed signs of adrenal pathologies. Adrenal and kidney lesions were observed in the killer whale. Likewise, enlarged adrenal glands were seen in the false killer whale, and the cause of stranding was described as adrenocortical dysfunction leading to chronic debilitation. It is plausible that the high contaminant exposure may have contributed to the health issues in these animals, since enlarged adrenal glands and increased corticosterone production have been seen in laboratory studies exposing animals to PCBs (Matthews et al., 1978) and altered adrenal physiology in marine mammals exposed to high organochlorine levels (Vos et al., 2000).

Compared to other more contaminated sites, stranded cetaceans from this region, even the killer whale, exhibit $\sum$PCBs well below concentrations reported from male bottlenose dolphins near an EPA Superfund Site (highest concentration of 2870 μg/g lipid) in Brunswick, Georgia (Balmer et al., 2011). Schwacke et al. (2002) derived a toxic threshold for marine mammal health at 14.8 μg/g lipid for $\sum$PCBs. The overall median value reported for all stranded tropical Pacific cetaceans from this study (5.12 μg/g lipid) was below the threshold, but seven (16.7%) animals (killer whale, false killer whale, pygmy killer whale, melon-headed whale (*Peponocephala electra*), striped dolphin (*Stenella coeruleoalba*) and bottlenose dolphin) had concentrations above the threshold, indicating that cetaceans from this region may be experiencing toxic effects from PCB exposure.

3.5. Ratio of *p,p′*-DDE/$\sum$DDT

DDT was used extensively in Hawai'i during the 1940s and although banned in the United States in 1972, DDT is still being used in developing countries primarily for malaria control (WHO, 2011). In fact, Hawai'i harbors substantial amounts of this pesticide as indicated by three Oahu soil sites which contain concentrations that rank in the top 5% of DDT reported in the nation (Brasher and Wolff, 2004). In all stranded cetaceans from this Pacific region, the most persistent metabolite, *p,p′*-DDE, was the predominant DDT compound detected. All individuals (except one humpback calf) had detectable amounts of parent DDT compound ranging from 1.3% (killer whale) of the total DDT concentration to as much as 22.4% (spinner dolphin, *Stenella longirostris*). In contrast, only two out of 79 (2.5%) Hawaiian monk seals had detectable *p,p′*-DDT concentrations (Ylitalo et al., 2008). In other studies, Hawaiian monk seals contained either no detectable *p,p′*-DDT (Willcox et al., 2004) or no more than 0.67% of total DDT (Lopez et al., 2012). This shows that the stranded cetaceans have a higher proportion of parent *p,p′*-DDT than monk seals from this region. The majority of marine mammal studies assess exposure to recent use of the parent DDT pesticide using a ratio of *p,p′*-DDE/$\sum$DDT, and Aguilar (1984) predicted that as new releases of technical DDT into the environment decrease, this ratio may stabilize around 0.6 in marine mammal tissues. All of the 16 species, except humpback whales, had ratios above 0.6 (Table 2). While humpback whale samples were below this threshold, the remaining proportions of their $\sum$DDT concentrations are not made of *p,p′*-DDT, they were the DDD isomers. When comparing DDT ratios with other tropical Pacific marine mammals, cetaceans in the current study were similar to Galapagos sea lion pups (*Zalophus wollebaeki*) (mean ± SE, 0.827 ± 0.028) (Alava et al., 2011) and greater than stranded delphinids from a metropolitan Atlantic coastal region of Brazil (mean, 0.56) (Santos-Neto et al., 2014). These comparisons indicate that cetaceans near remote Pacific islands are exposed to less recent application of the parent *p,p′*-DDT pesticide than those feeding in tropical, urbanized coastal mainland environments.

4. Conclusions

This unique sample set of stranded cetacean blubber tissues ($n = 42$) from the tropical Pacific region from 1997 thru 2011 provides the largest POPs evaluation for this geographical region since 1980 (O'Shea et al., 1980; Ylitalo et al., 2009). The samples represent a diverse mixture of species ($n = 16$), age classes and sexes. We acknowledge the complexity of this data set that originates from the opportunistic stranding of individuals from the diverse populations inhabiting this region. Yet despite the challenges inherent in this sample set, we demonstrate preliminary species category trends and life history influences (age class and sex) on POP concentrations, while detecting no significant temporal trends over this time period. Importantly, we established initial concentration values for POPs in the blubber of both baleen and toothed whales. The species category, age class and sex differences observed were expected and are supported by evidence elsewhere in the literature related to influences of trophic level, bioaccumulation through age, maternal offloading and atmospheric transport.

POP levels quantified in this remote global region are high enough to warrant continued monitoring of these toxic organic pollutants, as supported by the large number of cetaceans above the $\sum$PCBs toxic threshold. Additional research should focus on examining the health effects from these pollutants, especially in the more exposed species categories, as well as the analysis of prey species from this region. The results of this study should contribute to a framework for others addressing environmental health, assist in the global monitoring of POPs and serve to promote long term conservation and management strategies for these relatively understudied Pacific cetacean stocks.

Disclaimer

Certain commercial equipment, instruments, or materials are identified in this paper to specify adequately the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

Conflict of interest

None.

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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.2014.04.073.

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