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Persistent organic pollutants in fat of three species of Pacific pelagic longline caught sea turtles: Accumulation in relation to ingested plastic marine debris

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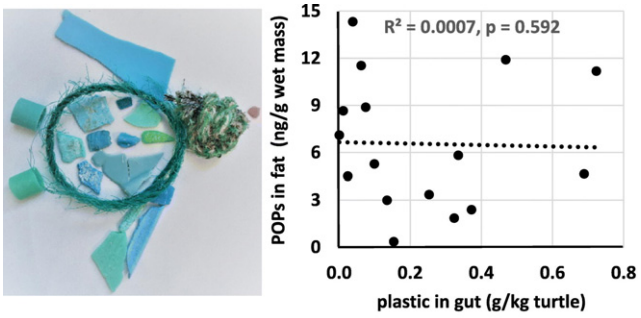
HIGHLIGHTS

- Persistent organic pollutants have rarely been measured in pelagic or Pacific sea turtles.
- Ingesting marine debris may expose sea turtles to persistent organic pollutants.
- We assessed correlations between ingested plastics and fat [POP] in two species.
- GC/MS & LC/MS/MS were used to measure POPs, including brominated flame retardants.
- Results suggest that ingested plastics are a minor source of POP exposure.

GRAPHICAL ABSTRACT



Plastic ingestion not correlated to POPs in sea turtles.

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ABSTRACT

In addition to eating contaminated prey, sea turtles may be exposed to persistent organic pollutants (POPs) from ingesting plastic debris that has absorbed these chemicals. Given the limited knowledge about POPs in pelagic sea turtles and how plastic ingestion influences POP exposure, our objectives were to: 1) provide baseline contaminant levels of three species of pelagic Pacific sea turtles; and 2) assess trends of contaminant levels in relation to species, sex, length, body condition and capture location. In addition, we hypothesized that if ingesting plastic is a significant source of POP exposure, then the amount of ingested plastic may be correlated to POP concentrations accumulated in fat. To address our objectives we compared POP concentrations in fat samples to previously described amounts of ingested plastic from the same turtles. Fat samples from 25 Pacific pelagic sea turtles [2 loggerhead (*Caretta caretta*), 6 green (*Chelonia mydas*) and 17 olive ridley (*Lepidochelys olivacea*) turtles] were analyzed for 81 polychlorinated biphenyls (PCBs), 20 organochlorine pesticides, and 35 brominated flame-retardants. The olive ridley and loggerhead turtles had higher ΣDDTs (dichlorodiphenyltrichloroethane and metabolites) than ΣPCBs, at a ratio similar to biota measured in the South China Sea and southern California. Green turtles had a ratio close to 1:1. These pelagic turtles had lower POP levels than previously reported in nearshore

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turtles. POP concentrations were unrelated to the amounts of ingested plastic in olive ridleys, suggesting that their exposure to POPs is mainly through prey. In green turtles, concentrations of ΣPCBs were positively correlated with the number of plastic pieces ingested, but these findings were confounded by covariance with body condition index (BCI). Green turtles with a higher BCI had eaten more plastic and also had higher POPs. Taken together, our findings suggest that sea turtles accumulate most POPs through their prey rather than marine debris.

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1. Introduction

Persistent organic pollutants (POPs) are man-made chemicals that are extremely persistent, globally transported by atmospheric and oceanic currents, and toxic (Jones and de Voogt, 1999; Wania and Mackay, 1995). POPs include a variety of compounds most of which are lipophilic and biomagnify in food webs, including polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and organochlorine pesticides (OCPs) such as dichlorodiphenyltrichloroethane and its main metabolites (DDTs). The uses of POPs are highly varied, but include pesticides, flame retardants, and other household and industrial purposes (United Nations Environmental Programme, 2017). In many instances POPs are found in environments far from their original source, with transport occurring via the food web, agricultural runoff, atmospheric circulation (Jones and de Voogt, 1999), and ocean circulation (Wania and Mackay, 1995).

POPs have contributed to population declines of several wildlife species, including alligators and birds (Carson, 1962; Fox, 2001; Guillette et al., 1994). In the case of sea turtles, effects of environmental pollutants are still poorly understood (Keller, 2013). The International Union on the Conservation of Nature (IUCN) lists the sea turtle species found in this study as endangered (green [*Chelonia mydas*] globally), vulnerable (olive ridley [*Lepidochelys olivacea*], loggerhead [*Caretta caretta*] turtles), or least concern (green turtles of the Hawaiian subpopulation) (IUCN, 2017). Sea turtles have faced a long list of human impacts affecting their survival, especially direct take and fisheries bycatch, but also anthropogenic chemical contamination (Lutcavage et al., 1997). Although POP concentrations in sea turtles are low relative to other wildlife that feed at higher trophic levels, concentrations have been significantly correlated with several health indicators, including white blood cell counts and some plasma chemistry measurements (Keller et al., 2004b, 2006).

The East Coast of the United States has been the region most extensively studied for POPs in sea turtles (Keller, 2013). While there are data on POP concentrations in sea turtle species inhabiting the Pacific Ocean, knowledge of POP concentrations in sea turtles surrounding the Hawaiian Islands are limited, with only three published studies (Aguirre et al., 1994; Keller et al., 2014a; Miao et al., 2001). Notably, only two of these three studies used methods sensitive enough to detect POPs. Recently, baseline data on POPs have been published for sea turtles in coastal areas of Australia, Japan, Baja California, and Malaysia (Hermanussen et al., 2008; Labrada-Martagon et al., 2011; Richardson et al., 2010; van de Merwe et al., 2010), but data are still lacking in pelagic areas. Additionally, most of these Pacific studies focused on green turtles with only five olive ridleys and five loggerheads having been measured for POPs (Gardner et al., 2003; Richardson et al., 2010). Hence, an assessment of POP exposure in vast regions inhabited by sea turtle species of all size classes is important for better understanding this threat to different life stages and populations of these protected species.

POP exposure occurs mostly through the food web, and broad comparisons across all studies support the general conclusion that POP concentrations follow trophic status and are highest in Kemp's ridley sea turtles (*Lepidochelys kempii*), followed by loggerhead, leatherback (*Dermochelys coriacea*) and finally green turtles (Keller, 2013). Data on olive ridleys have been too limited to rank this species among other sea turtles by POP concentrations. In an 18-year study of diet content analysis of sea turtles captured in American Samoan and Hawaiian pelagic longline fisheries, olive ridleys were found to be opportunistic generalists consuming gelatinous zooplankton and fish, anthropogenic debris (Wedemeyer-Strombel et al., 2015), and often would graze from longline hooks (Work and Balazs, 2002). Juvenile pelagic green turtles captured as bycatch in Pacific longlines were found to be opportunistic, mainly carnivorous, feeding at or near the surface (Parker et al., 2011). Pacific loggerheads fed primarily at the surface on molluscs, hydrozoans and pyrosomes with few deep water prey (Parker et al., 2005). These feeding habits suggest that olive ridleys would likely rank somewhere between Kemp's ridleys and loggerheads for POP concentrations based on their trophic status. Green turtles often rank lowest in POP concentrations (Keller, 2013), but the ranking of green turtles in this study are more difficult to predict. While they are pelagic (farther from contaminant sources) and younger (fewer years to accumulate contaminants), they feed at a higher trophic level than the older neritic herbivorous phase previously analyzed.

One potential route of exposure of sea turtles to POPs is through ingestion of plastic debris (Teuten et al., 2009). The hydrophobic nature of plastic attracts chemicals, such as POPs, to its surface, and POPs have been found in plastic debris collected from beaches around the world (Endo et al., 2005; Mato et al., 2001; Ogata et al., 2009; Rios et al., 2007), including Hawaii (Heskett et al., 2012). In a companion study to this paper, 92% of the turtles used in this study were found to have ingested plastic debris (Clukey et al., 2017). Furthermore, correlations of chemicals found in seabird fat and their ingested plastics suggest that plastic may be an additional source of exposure to these classes of POPs (Ryan et al., 1988; Tanaka et al., 2013; Yamashita et al., 2011). Specifically, higher brominated PBDE congeners have been found in seabird tissues, which were not present in their prey, but that are incorporated in plastics and textiles as flame-retardants (Tanaka et al., 2013).

Given our limited knowledge about contaminants in the Pacific pelagic zone, we sought to provide baseline contaminant levels of three species of pelagic Pacific sea turtles and spatial trends of contaminant levels in sea turtles inhabiting the Pacific Ocean. In addition, because POPs can accumulate with age or be offloaded from females into eggs (Stewart et al., 2011), we examined POP concentrations between sexes and across lengths and body condition indices of turtles. Additionally, we hypothesized that if ingesting plastics is a significant source of POP exposure, then the amount of ingested plastic would be correlated with the concentrations of POPs accumulated in fat, as seen previously in seabirds (Ryan et al., 1988; Yamashita et al., 2011).

2. Methods

2.1. Sample collection

The U.S. National Oceanic and Atmospheric Administration (NOAA) Pacific Islands Regional Office (PIRO) uses observers on the Hawaiian and American Samoan longline fisheries to collect fisheries catch and bycatch data. Between June 2012 and December 2013, latitude 13.5 °S and 29.6 °N, and longitude 140 °W and 170 °W (Fig. 1), 25 sea turtles (two loggerhead, six green, and seventeen olive ridley turtles) drowned as bycatch in these fisheries were sampled. Observers recorded the capture latitude and longitude and sent the frozen carcasses to NOAA, National Marine Fisheries Service, Pacific Islands Fisheries Science Center

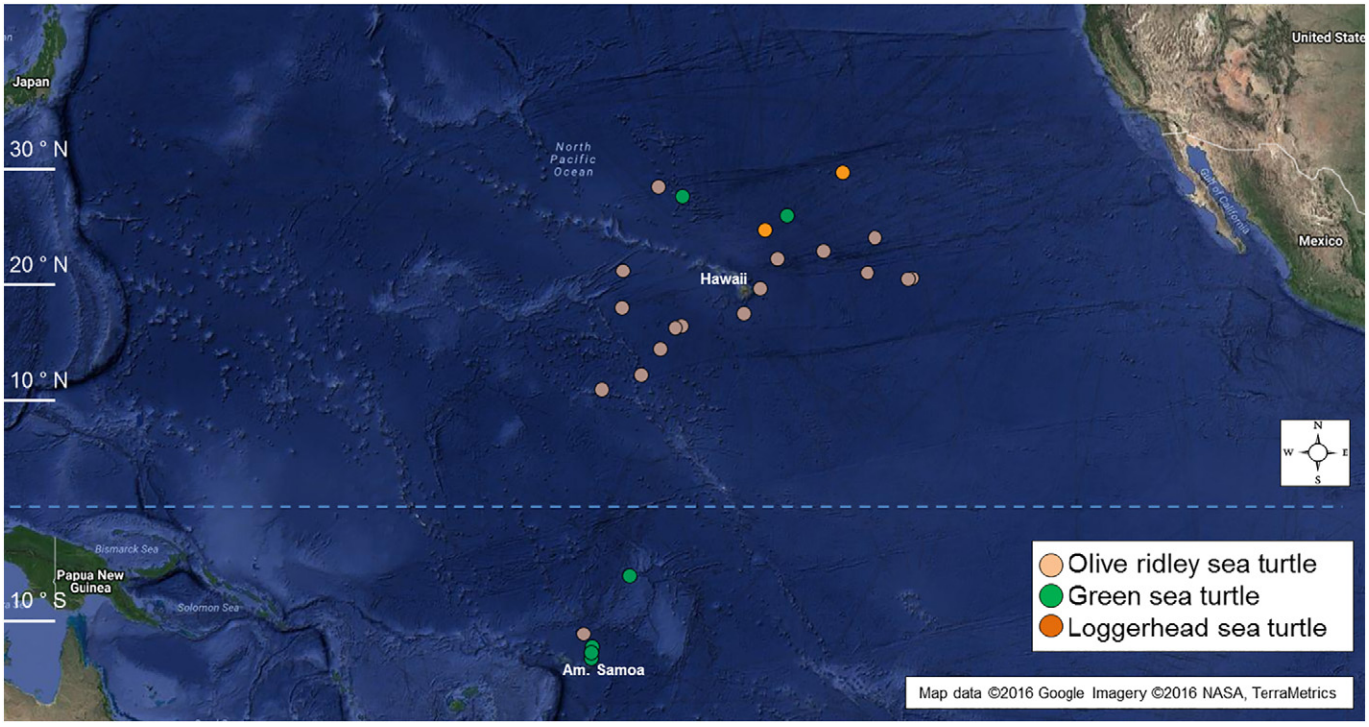

Fig. 1. Capture locations of pelagic Pacific sea turtles sampled in this study. Dashed line is the equator.

in Honolulu, Hawaii, for necropsy. Turtle mass (kg) and straight carapace length (SCL in cm) were recorded. Comprehensive necropsies entailing complete external and internal exams of all organ systems including histology of most organs were performed on all individuals. Body condition at necropsy was classified as poor, fair, good, or excellent based on the appearance of muscle and fat tissue in the inguinal region and under the plastron (Work, 2000). Sex and size class of turtles were determined by visual examination of gross gonadal morphology. Cause of death was determined as forced submergence for all turtles. Body condition index (BCI) can objectively and numerically estimate the quantity of fat in sea turtles (Barco et al., 2016). BCI was calculated as body mass (in kg) divided by the cube of SCL (in cm) and multiplied by 100,000. Fat was sampled from the left inguinal region using hexane-rinsed stainless steel scalpel blade and forceps then stored in a 15 mL Teflon jars. Fat samples were shipped in liquid nitrogen dry vapor shippers (−150 °C) to the National Institute of Standards and Technology (NIST), Hollings Marine Laboratory, Charleston, South Carolina, and curated as part of the Biological and Environmental Monitoring and Archival of Sea Turtle Tissues (BEMAST) project of the Marine Environmental Specimen Bank (Keller et al., 2014b). Samples were cryohomogenized at liquid nitrogen vapor temperatures, and a subsample of each was analyzed for POPs.

2.2. Persistent organic pollutants

2.2.1. Sample preparation, extraction and cleanup

Fat samples were cryo-homogenized using the Retesch Cryomill (Haan, Germany) at 25 Hz for 5 min. Fat subsamples (≈1 g) were combined with sodium sulfate, transferred to pressurized fluid extraction (PFE) cells and spiked gravimetrically with internal standard solution, containing ^{13}C-labeled PCB congeners (28, 52, 77, 126, 169, 118, 153, 180, 194, 206), 6-F-PBDE 47, unlabeled 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, 4,4′-DDE, 4,4′-DDD, 4,4′-DDT), ^{13}C–labeled methyl-triclosan and ^{13}C-labeled α-, β- and γ-hexabromocyclododecanes (HBCDs). All PCB and PBDE congeners were numbered according to IUPAC rules. POPs were extracted using PFE with 3 cycles of dichloromethane (DCM) at 100 °C and 13.8 MPa. Total extractable organic (TEO) content (a proxy for lipid content) was determined by removing 12% of the extract gravimetrically, allowing it to dry in a tared aluminum pan, and weighing dried residue to the nearest 0.01 mg approximately 24 h later. Remaining extracts were cleaned up using size exclusion chromatography (SEC) with 10 mL/min DCM on a PLGel column (600 mm × 25 mm, 10 μm particle size with 100 Å diameter pores, Polymer Labs, Amherst, MA). Extracts were fractionated with acidified silica columns as described in Keller et al. (2009). Fraction One (F1) and Fraction Two (F2) extracts were solvent exchanged to isooctane and methanol, respectively, and each evaporated to 0.2 mL. F1 extracts were injected onto a gas chromatograph mass spectrometer (GC/MS) for all compounds except HBCDs, which were measured from F2 extracts injection onto a liquid chromatograph tandem mass spectrometer (LC/MS/MS). Additional method details are provided in Supplemental Information.

2.2.2. GC/MS analysis

Each F1 extract was injected onto a GC/MS two different times for different target constituents using methods similar to Keller et al. (2014a). Details pertaining to the instrument, oven program, and ions monitored are provided in Supplemental Information (text and Tables S1 and S2). The first injection was performed with an electron impact (EI) source and a programmable temperature vaporization (PTV) inlet operated in the solvent vent mode onto a 5 m × 0.25 mm Restek Siltek guard column connected to a 0.18 mm × 30 m × 0.18 μm film thickness Agilent DB-5MS capillary column. PCBs, selected PBDEs, selected pesticides, and selected additional BFRs were quantified from this injection. PBDEs and selected additional BFRs were quantified from the second injection with a negative chemical ionization (NCI) source and a cool on-column injection of 2 μL onto a 5 m × 0.25 mm Restek Siltek guard column connected to a 0.18 mm × 10 m × 0.18 μm film thickness DB-5MS Agilent analytical column.

2.2.3. LC/MS/MS analysis

HBCDs were quantified using 20 μL injections of F2 extracts as described in Bachman et al. (2014). An Agilent Eclipse Plus XDB-C18

(3.0 mm × 150 mm × 3.5 μm) column on an Agilent 1100-series LC was connected to an electrospray ionization source on an API 4000 MS/MS (Applied Biosystems, Foster City, CA).

2.2.4. QA/QC and quantification

Three replicates of NIST Standard Reference Material (SRM) 1945 "Organics in Whale Blubber" were analyzed as controls. These SRM replicates, laboratory procedural blanks and calibration solutions were extracted, processed and analyzed concurrently with the sample set. Six-point calibration curves ranged from 0.06 ng to 300 ng of compounds found in 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, additional solutions containing 46 PCB and 28 PBDE congeners, the following from Accustandard (New Haven, CT, USA): octachlorostyrene, α-, β-, and γ-HBCDs, pentachlorobenzene, and the following additional BFRs from Wellington Laboratories (Guelph, Ontario, Canada): 1,2-bis(246-tribromophenoxy) ethane (BTBPE), hexabromobenzene (HBB), pentabromoethylbenzene (PBEB), decabromodiphenylethane (DBDPE), 4-methoxy-2,3,3′,4′,5-pentachlorobiphenyl (4-methoxy PCB 107), 4-methoxy-2,2′,3,4′,5,5′-hexachlorobiphenyl (4-methoxy PCB 146), 4-methoxy-2,2′,3,4′,5,5′,6-heptachlorobiphenyl (4-methoxy PCB 187), 6-methoxy-2,2′,4,4′-tetrabromodiphenyl ether (6-methoxy PBDE 47).

The internal standard approach was used to quantify each compound amount. Amounts of each analyte were calculated using the slope and y-intercept of at least a three point calibration curve that bracketed the peak area ratios observed in the samples. Concentrations were determined by dividing the calculated analyte mass by the extracted sample mass. The reporting limits were determined as per Ragland et al. (2011) and are provided in Tables S1 and S2.

2.3. Statistical analysis

All statistical analyses were conducted using the Nondetects and Data Analysis for Environmental Data (NADA) package as recommended for left censored data (Helsel, 2005) in the program R. This approach appropriately handles data with values that are below detection limits. Mean, median and standard deviations were calculated using Kaplan–Meier, maximum likelihood estimation (MLE), or regression on order statistical (ROS) models (Table S5). Mass fractions (ng/g wet mass) of detected compounds were summed for 81 polychlorinated biphenyl congeners (ΣPCBs), 28 PBDE congeners (ΣPBDEs), six DDT and metabolite compounds (ΣDDTs), six chlordanes (ΣCHLs), three HBCD isomers (ΣHBCDs), and three HCH isomers (ΣHCHs) (details provided in Supplemental Information). Mirex, HCB, octachlorostyrene (OCS), and pentachlorobenzene (PeCB) were analyzed individually.

Normality and homoscedasticity of raw and log-transformed data were tested using Shapiro-Wilk and Bartlett tests, respectively. Either parametric (regression by maximum likelihood estimation for left-censored data using the R NADA function cenmle) or nonparametric (empirical cumulative distribution function differences for left-censored data using the R NADA function cendiff) tests were used for differences among turtle species and differences between sexes of olive ridley sea turtles.

Either parametric (regression equation and the likelihood correlation coefficient for left-censored data using the R NADA function cenreg) or nonparametric (Kendall's tau correlation coefficient and associated line for left-censored data using the R NADA function cenken) were used to assess relationships between POP concentrations and TEO, turtle length (SCL), BCI, ingested plastic amounts, and capture location. Six different ways of measuring ingested plastic were tested for correlation to POPs: number of plastic pieces, plastic mass, volume, surface area, body burden (g plastic/kg turtle), and percent of total wet gut content mass comprised of plastic. Capture latitude was tested with locations south of the equator as negative decimal degrees and again with those changed to absolute values to test for distance from equator. We examined sex and size class relationships only in olive ridley turtles, since the loggerhead turtle sample size was too small and all green turtles were immature. To minimize type I errors statistical tests were performed on only compounds that were detected in >50% of the samples, and Bonferroni corrections were used on a p-value of 0.05 (i.e. when 5 compound classes were tested individually, significance was determined as a p-value <0.01).

3. Results and discussion

3.1. Turtle size classes, body condition, and plastic ingestion

Measurements, sex, and body condition of each turtle are shown in Table S3. Both loggerheads were adults, so they may be older than 37 years based on the age at maturity estimate for this population (Ishihara, 2011). Of the 17 olive ridley turtles, just three were immature; the rest were adults. In the Pacific Ocean, olive ridleys are expected to reach maturity at 13 years of age (Zug et al., 2006). All green turtles were immature and estimated at 7 to 11 years old based on comparing their lengths to Hawaiian green turtles that have recruited to nearshore habitat and switched to a more herbivorous diet (Zug et al., 2002). With the exception of two individuals, all other sea turtles were classified as being in good or excellent body condition. One immature female olive ridley was in poor body condition. Plastic ingestion amounts were published previously from these and additional turtles (Clukey et al., 2017). Briefly, ingested plastics were found in one of the two loggerheads (10.8 g), all of the olive ridleys (mean of 6.1 g), and five of the six green turtles (mean of 23.1 g). No adverse impacts from plastic ingestion were observed in these turtles (Clukey et al., 2017).

3.2. POP concentrations in general

The mass fractions of POPs in the replicates of SRM 1945 were on average within 12% of the certified values, providing good confidence in the data from the sea turtle samples. Of the 138 chemicals targeted, 29 individual compounds were above the reporting limit in at least one of the turtles in this study (Tables S4 and S5). ΣDDTs and ΣPCBs were the predominant POPs followed generally by ΣCHLs, α-HBCD, mirex, and then ΣPBDEs (Fig. 2). One olive ridley turtle had an order of magnitude greater ΣDDT concentration (159 ng/g wet mass) than the rest, which ranged from below the reporting limit to 14.3 ng/g wet mass. For one green turtle and one olive ridley turtle, ΣDDT concentrations

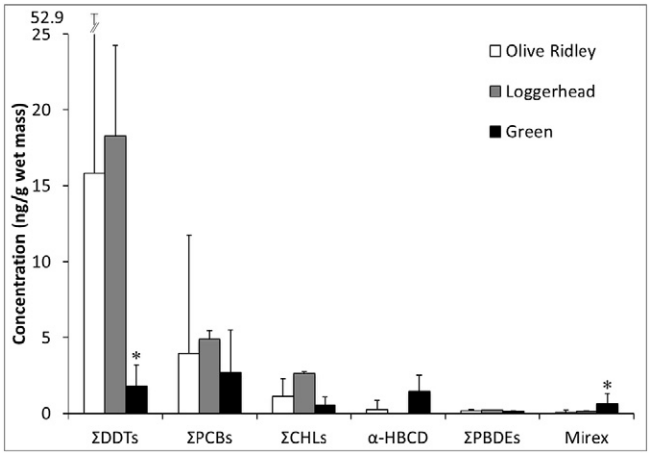

Fig. 2. Mean and standard deviation of persistent organic pollutant concentrations in fat of pelagic Pacific sea turtle species. Dichlorodiphenyltrichloroethanes and its main metabolites (ΣDDTs), polychlorinated biphenyls (PCBs), chlordanes (CHLs), hexabromocyclododecane (HBCD), polybrominated diphenylethers (PBDEs). Asterisk indicates a significant difference between olive ridley and green sea turtles.

were below the reporting limit. The only detected HBCD isomer was α-HBCD with eight turtles having concentrations above the reporting limit and green turtles having the highest mean concentration of 1.46 ng/g wet mass. In addition, hexachlorobenzene was detected in green and loggerhead turtles, while 6-methoxy PBDE 47 was detected only in olive ridley turtles (n = 3) and juvenile green turtles (n = 2) (Tables S4 and S5). DBDPE was detected in only one adult male olive ridley turtle with 1.17 ng/g wet mass. Pentachlorobenzene was detected only in the two loggerhead turtles with an average mass fraction of 0.699 ng/g wet mass. Octachlorostyrene, BTBPE, HBB, PBEB, 4-methoxy PCB 107, 4-methoxy PCB 146 and 4-methoxy PCB 187 were not detected in any of the sea turtles. ΣDDTs were the predominant POP class in olive ridley (median = 6.12 ng/g wet mass) and loggerhead (median = 18.3 ng/g wet mass) turtles, whereas ΣPCBs were the highest of the POP classes in green turtles (1.11 ng/g wet mass) (Table S5 and Fig. 2).

The pelagic sea turtles in this study typically had lower fat concentrations of POPs than those along Baja California, except ΣDDTs were three times higher in pelagic olive ridleys compared to those sampled along Baja California (Gardner et al., 2003) (Table 1). Juvenile green turtles had approximately half the concentration of ΣPBDEs measured in an adult female green turtle off the coast of Queensland, Australia (Hermanussen et al., 2008) (Table 1). While these comparisons are complicated by different life stages and sample sizes of only one, they are the only studies available to compare pelagic to neritic Pacific sea turtles. The comparisons begin to suggest, as expected, that pelagic turtles are less exposed than neritic ones.

In sea turtles along the Southeastern coast of the U.S., ΣPCBs are the predominant POP class followed by ΣDDTs (Keller et al., 2004a; Rybitski et al., 1995) (Table 1). The pelagic Pacific loggerhead and olive ridley turtles from this study had the opposite profile and lower concentrations of both contaminant classes. These differences suggest that sea turtles in the pelagic realm of the Pacific Ocean are not only exposed to much lower contamination but also to a different mixture than turtles inhabiting nearshore areas of the Northwest Atlantic Ocean (Keller et al., 2004a; Rybitski et al., 1995).

Within the central Pacific Ocean, few marine species have been tested for POP concentrations. The available data show that sea turtles are less exposed than marine mammals, especially those at high trophic levels. One of the highest trophic level cetaceans near the Hawaiian Islands, the false killer whale (*Pseudorca crassidens*), averaged (63,000 ± 28,000) ng/g lipid ΣDDTs in adult males and (20,000 ± 4900) ng/g lipid ΣDDTs in subadults (Ylitalo et al., 2009). Average ΣDDTs blubber concentrations of 16 different cetacean species stranded in the Hawaiian Islands were (16,600 ± 30,300) ng/g lipid (Bachman et al., 2014). Lower trophic level Hawaiian monk seals (*Monachus schauinslandi*) from the main Hawaiian Islands averaged 690 ng/g lipid ΣDDTs in adult males, 390 ng/g lipid ΣDDTs in subadults, and 190 ng/g lipid ΣDDTs in adult females (Lopez et al., 2012). Sea turtles in this study averaged 51.9 ng/g lipid or 38.9 ng/g wet mass ΣDDTs. While other species have higher POP concentrations in this region, toxic and sublethal effects of POPs on sea turtles are still not completely understood (Keller, 2013).

POP concentrations were not significantly correlated to TEO (lipid content) in the fat samples (Tables S6 and S7), so we did not normalize the POP concentrations to lipid content for remaining statistical testing.

3.3. POP profiles

In loggerhead and olive ridley turtles, ΣDDTs were the predominant POP followed by ΣPCBs, ΣCHLs, and ΣPBDEs, respectively (Fig. 2), a pattern observed in some other central Pacific wildlife (Bachman et al., 2014; Ylitalo et al., 2009). POPs patterns in sea turtles from the Atlantic Ocean and along the coast of Baja California show that ΣPCBs are higher than ΣDDTs (Gardner et al., 2003; Keller et al., 2004a; Ragland et al., 2011; Stewart et al., 2011). Although it is difficult to accurately determine geographic sources of contamination for these pelagic turtles because of their extensive migrations, others have used ratios of PCBs and DDTs to aid in localizing geographic origin of animals (Le Boeuf et al., 2002). To compare across the Pacific Ocean, we calculated the ΣPCBs:ΣDDTs ratio, which is the reciprocal of the ratio used by Le Boeuf et al. (2002), in the sea turtles and from data published on other biota across the Pacific Ocean (Fig. 3). The ratios in the two loggerhead turtles were 0.24 and 0.32; and in olive ridley turtles the median (range) was 0.232 (0.0295 to 3.26). These low ratios are most similar to marine organisms from the South China Sea and from central California to Ecuador (see references listed in Fig. 3). The ratios in these turtles were approximately two- to 30-times lower than green turtles foraging near Kailua Bay on Oahu (Keller et al., 2014a), tuna in the North Pacific and off of Japan and Taiwan (Ueno et al., 2004), Hawaiian monk seals from the Northwestern Hawaiian Islands (Ylitalo et al., 2008), cetaceans off of Japan (Kajiwara et al., 2006), bivalves in Chile or Peru (Farrington and Tripp, 1995), and harbor porpoises from Washington State (Calambokidis and Barlow, 1991). Interpreting these comparisons is complicated by the broad diversity of species with different diets, migration patterns, and metabolic abilities, as well as different atmospheric transport of PCBs versus DDTs across such a large latitudinal gradient and differences in the number of congeners measured among studies. Regardless, this is the first time this ratio has been mapped across the Pacific Ocean, and a clear and consistent pattern is evident with ratios

Table 1
Persistent organic pollutant levels in fat of sea turtles from current and selected studies. Mean (standard deviation) or range in ng/g wet mass. Ranked generally from highest to lowest by species and location. Loggerhead (Cc), green (Cm), olive ridley (Lo) sea turtles, juvenile (J), adult (A), male (M), female (F), sample size (N), not reported (NR). Dichlorodiphenyldichloroethylene (DDE), dichlorodiphenyltrichloroethane and its main metabolites (ΣDDTs), polychlorinated biphenyls (PCBs), chlordanes (CHLs), polybrominated diphenylethers (PBDEs), hexachlorobenzene (HCB). Below reporting limit (<RL). * indicates a significant difference between olive ridley and green turtles in the current study.

Species	Stage/sex	Location	Year	N	4,4′-DDE	ΣDDTs	ΣPCBs	ΣCHLs	ΣPBDEs	Mirex	HCB	% lipid	Reference
Cc	JAMF	Virginia - North Carolina	1991–1992	20	195 (266)		551 (473)						Rybitski et al. (1995)
Cc	JMF	Core Sound, North Carolina	2000–2001	44	64.9 (64.3)		256 (269)	26.9 (21.3)		4.52 (4.06)	1.13 (2.38)	26.3 (20.6)	Keller et al. (2004a); Keller (2013)
Cc	NR	Baja California	NR	1		<RL	<RL	<RL			<RL		Gardner et al. (2003)
Cc	AMF	Pelagic Pacific	2012–2013	2	14.0 to 22.0	14.1 to 22.5	4.54 to 5.30	2.54 to 2.70	0.211 to 0.227	0.139 to 0.157	1.88 to 2.46	84.5 to 86.5	This study
Lo	JAMF	Pelagic Pacific	2012–2013	17	15.5 (36.9)*	15.8 (37.1)*	3.95 (7.79)	1.13 (1.15)	0.173 (0.0821)	0.0714 (0.158)*	<RL	64.5 (21.6)	This study
Lo	NR	Baja California	NR	1		5.1	18.4	8.1			<RL		Gardner et al. (2003)
Cm	JMF	Baja California	NR	7		<RL to 12.2	<RL to 49.5	<RL to 65.1			<RL		Gardner et al. (2003)
Cm	AF	Queensland, Australia	2004–2006	1					0.2574			78	Hermanussen et al. (2008)
Cm	JMF	Pelagic Pacific	2012–2013	6	1.68 (1.41)	1.80 (1.37)	2.71 (2.80)	0.554 (0.512)	0.150 (0.0315)	0.611 (0.689)	0.251 (0.270)	69.3 (12.7)	This study

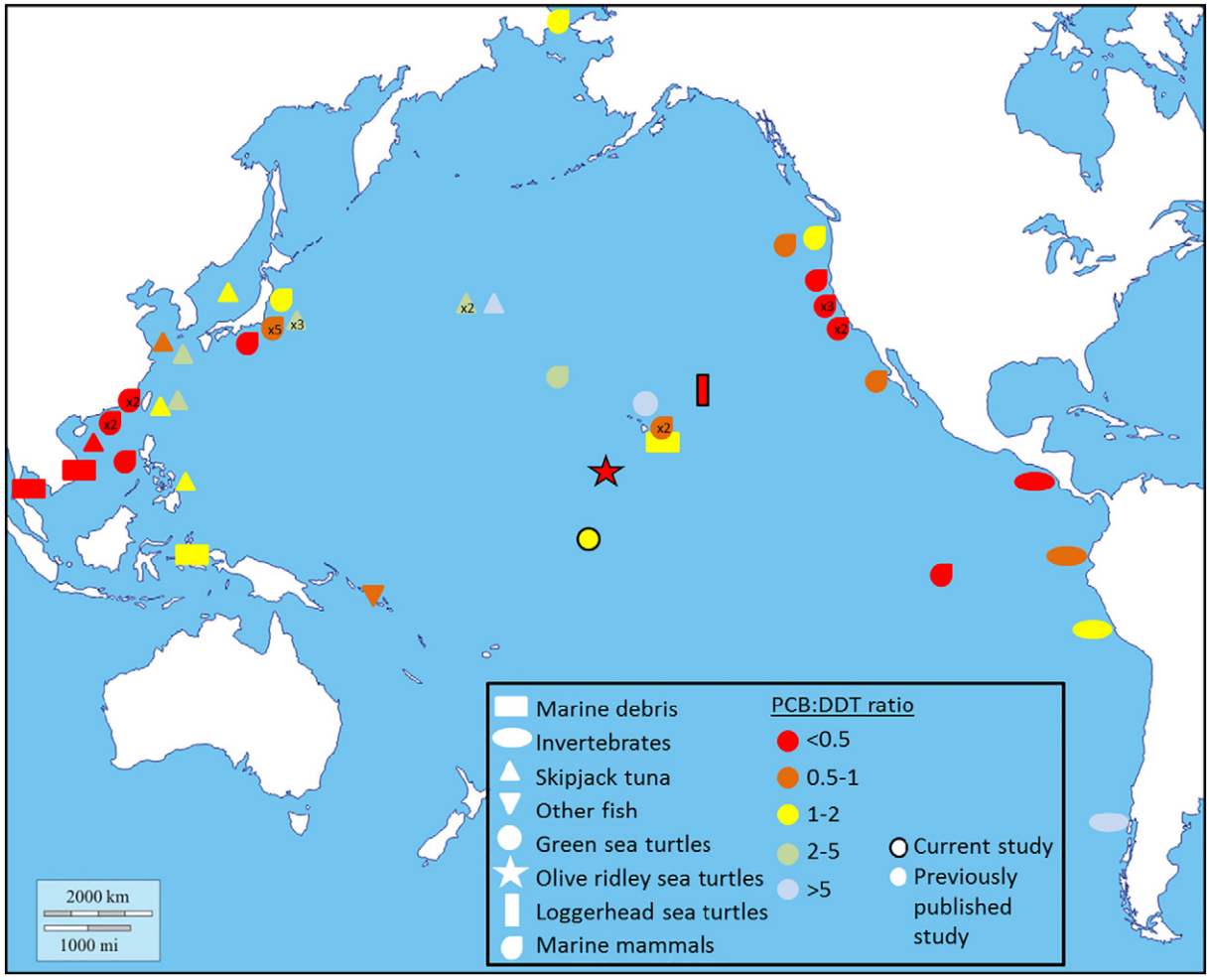

Fig. 3. ΣPCB:ΣDDT ratios (indicated by different colors) in marine biota and debris (indicated by different shapes) from the Pacific Ocean. Numbers inside symbols indicate the number of studies or samples that particular symbol represents. Ratios were calculated using data from the current study (black outline) and those of previously published studies (Alava et al., 2009, 2011b; Bachman et al., 2014; Calambokidis and Barlow, 1991; Del Toro et al., 2006; Farrington and Tripp, 1995; Heskett et al., 2012; Hoguet et al., 2013; Kajiwara et al., 2006; Kannan et al., 2004, 1997; Keller et al., 2014a; Le Boeuf et al., 2002; Ogata et al., 2009; Ramu et al., 2006; Ueno et al., 2004; Ylitalo et al., 2009, 2008).

below 0.5 in the South China Sea and in the East Pacific Ocean from California to Ecuador. Biota further north of these two regions is dominated by ratios that are well above one. The lower ratios appear to occur near more tropical countries, perhaps because of the continued use of DDT for malaria prevention (United Nations Environmental Programme, 2017). Central Pacific biota, including the sea turtles in this study, does not consistently match either pattern; rather, the ratio depends on the species and likely their migratory pathways.

Within each contaminant class, the three species in this study shared similar congener profiles or contaminant composition (Fig. S1). However, green turtles tended to have a higher proportion of *trans* nonachlor. For all species, 4,4′-DDE consisted of 90% to 94% of ΣDDTs with 4,4′-DDT being the only other DDT compound detected. 4,4′-DDE is the most persistent metabolite of the pesticide DDT indicating that Pacific pelagic sea turtles are exposed to older, rather than recent usage of 4,4′-DDT (Aguilar, 1984).

In all three species the predominant PCBs congeners were 138, 146, 153 + 132, and 180 + 193 (Fig. S1). This is similar to sea turtles in the Atlantic Ocean except that pelagic Pacific turtles had higher proportions of PCB 146 and lower proportions of PCB 187 and PCB 199 (Ragland et al., 2011). Globally, PBDE 47 is the PBDE congener in the highest concentration in most wildlife (Hites, 2004), and it was nearly the only PBDE congener detected in these sea turtle samples. Only one immature male olive ridley turtle had detectable concentrations of PBDEs 153 and 154, while PBDE 47 was below the reporting limit (Table S4).

3.4. Species differences

Though loggerhead turtles had higher concentrations of ΣDDTs, ΣPCBs, and ΣCHLs than the other two species, sample size ($n = 2$) prevented statistical comparisons. Statistically significant differences in concentrations for some compounds were observed between olive ridley and green turtles (Fig. 2, Table S5). Olive ridleys showed significantly higher levels of PCB 149 ($p < 0.001$ but not ΣPCBs) and 4,4′-DDE (and ΣDDTs, $p = 0.008$) whereas green turtles showed significantly higher levels of mirex ($p < 0.001$). Most of the olive ridley turtles (82%) were adults while all six green turtles were immature. The different size classes and or differences in prey selection may have resulted in these species differences. In Baja California ΣPCBs concentrations were highest in loggerheads followed by olive ridley and then green turtles (Richardson et al., 2010), similar to the trend we saw in pelagic turtles.

3.5. Sex and length relationships

No differences between any POP was evident between sexes in olive ridleys (14 females, 3 males) ($p > 0.05$) despite most of them being adults. However, a positive correlation between SCL and ΣCHLs concentrations was apparent (Table S6), suggesting bioaccumulation through age. Even when the turtle with the smallest SCL and poorest body condition was excluded, ΣCHLs showed a linear fit with SCL (ΣCHLs =

$0.245 * SCL - 13.3$; $R^2 = 0.231$; tau = 0.417; $p = 0.027$). The increasing concentrations are not surprising as our samples span immature and adult sea turtles, and bioaccumulation with age is commonly seen in other marine organisms (Borrell et al., 1995).

3.6. Geographic comparison of POP concentrations

POP concentrations were not correlated with capture location in olive ridleys ($p > 0.05$; Table S6), but were positively correlated with latitude (Fig. 4), absolute value of latitude, and longitude in green sea turtles (Table S7). POP concentrations in green turtles increased further northeast and with greater distance from the equator. The green turtles captured in the Northern Hemisphere were approximately three to six times higher in concentration of ΣDDTs and ΣPCBs than those captured in the Southern Hemisphere (Fig. 4). This difference between the hemispheres has been observed frequently in samples of air, water, and biota, because greater amounts of POPs were used in the Northern Hemisphere (Connell et al., 1999). Similar increases in sea turtle POP concentrations moving away from the equator have been observed along the east coast of the U.S. and are thought to be due to greater human density and use of compounds farther north (Alava et al., 2011a; Ragland et al., 2011). This trend in the Pacific Ocean is also likely related to global distillation, the geochemical process by which certain semi-volatile chemicals, like POPs, are transported from warmer regions to colder regions through evaporation and condensation (Simonich and Hites, 1995). The process is repeated in hops with latitude, giving it the name grasshopper effect, carrying chemicals thousands of kilometers in a matter of days towards the poles (Gouin et al., 2004).

3.7. Correlations with body condition index

Body condition index can be a useful calculation to objectively and numerically estimate the quantity of fat a turtle has, though its reliability has been disputed (Barco et al., 2016). BCI and TEO were not correlated with one another for green turtles ($p = 0.397$; Fig. 5A) or for the larger sample size of olive ridley turtles ($p = 0.072$) when the smallest, poorest body condition turtle was excluded. The same finding was reported for immature loggerhead turtles from North Carolina (Keller et al., 2004a). This might suggest that BCI is not a reliable indicator of the amount of fat, but the majority of the turtles in both studies were of good or excellent body condition when assessed visually. This limited range of BCIs may reduce the ability to see significant correlations. BCI was correlated positively with the amount of plastic ingested by green turtles (Clukey et al., 2017) (Fig. 5A). This does not mean that turtles eating more plastic are healthier, rather that fatter turtles ate more of both plastic and prey. Turtles that eat more food may be expected to have higher POP concentrations, and the significantly positive correlations between BCI and concentrations of ΣPCBs (Fig. 5A), ΣCHLs, and mirex appear to confirm this relationship (Table S7). ΣPBDEs and a-HBCD were not significantly correlated with BCI (Table S7).

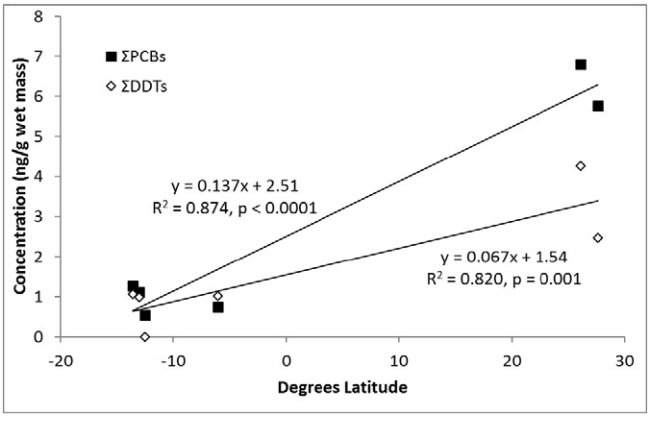

Fig. 4. Correlations of capture latitude with fat concentrations of polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane and its main metabolites (ΣDDTs) in pelagic Pacific green sea turtles.

3.8. Correlations with plastic ingestion

Within the olive ridley turtles, no correlations were observed between POP concentrations and six different methods of measuring ingested plastics (Table S6). The lack of relationship indicates that little to none of the variability in POPs stored in the fat of olive ridleys is explained by the amount of ingested plastics currently in their gut. Within the smaller sample size of green turtles, one significant positive correlation was observed between concentrations of ΣPCBs and the number of plastic pieces ingested (Fig. 5A, Table S7). No correlations were observed with the other five approaches for measuring amount of plastic ingested: mass, volume, or surface area of ingested plastics, body burden (g plastic per kg turtle) or % of gut contents consisting of plastic (Table S7).

The largest route of exposure to POPs for sea turtles is through ingestion versus dermal or inhalation (Keller, 2013). The ingestion route can include ingestion of natural prey, associated sediments, seawater, and/or plastic debris. Sediment can be ruled out for these particular turtles since they are likely foraging in waters too deep for them to reach the seafloor. Seawater is negligible since it will have orders of magnitude lower concentrations of lipophilic POPs than prey or plastics. Transfer of sorbed POPs from ingested plastics into animals has been hypothesized, but few studies have proven it, fewer have considered the relative exposure from plastics versus prey, and none have examined the pharmacodynamics of POPs on plastics within the gut of vertebrates (Rochman, 2015; Teuten et al., 2009).

Three studies offer correlative evidence that POPs transfer to wildlife. These demonstrated that greater amounts of ingested plastics relate to greater concentrations or altered profiles of particular POPs (ΣPCBs, higher brominated PBDEs, and lower chlorinated PCBs, respectively) in seabird tissues (Ryan et al., 1988; Tanaka et al., 2013; Yamashita et al., 2011). The current study is the first of this type for sea turtles, but findings differed from the seabird studies. In olive ridley turtles, no POP related to plastic ingestion, not even the lowest chlorinated congener detected, PCB 99. Higher brominated PBDE congeners were not detected in any individual of any turtle species, so these particular compounds were not substantially leaching from the ingested plastics as suggested in seabirds (Tanaka et al., 2013). Within green turtles, we inspected whether lower chlorinated PCB congeners were more strongly correlated with number of ingested plastic pieces than higher chlorinated congeners, but this pattern seen previously in seabirds (Yamashita et al., 2011) was not apparent in the sea turtles. Two principal limitations of correlative studies like these should be acknowledged. First, they cannot prove cause and effect. Second, they cannot account for time lags without assumptions. For instance, the plastics found in the guts of these turtles were consumed approximately three to six weeks prior to capture (Clukey et al., 2017), but the POPs in their fat had presumably accumulated over their entire lives, approximately a decade for the olive ridleys and green turtles. This time lag requires us to assume either that individual turtles are consistent in their plastic ingestion or that fat quickly assimilates recent dietary exposure. Both assumptions are plausible based on evidence. As creatures of habit, individual sea turtles are known to specialize in a particular prey item (Vander Zanden et al., 2010); thus, it is reasonable that a turtle that tends to eat plastic debris may always tend to eat plastics. And, POP concentrations in sea turtle fat significantly correlate to POP concentrations in blood (Keller et al., 2004a), suggesting the fat can reflect recent exposure.

Experimental studies have shown that ingesting plastics that have sorbed POPs can transfer POPs to the tissues of invertebrates and fish (reviewed in (Rochman, 2015; Teuten et al., 2009)). However, the

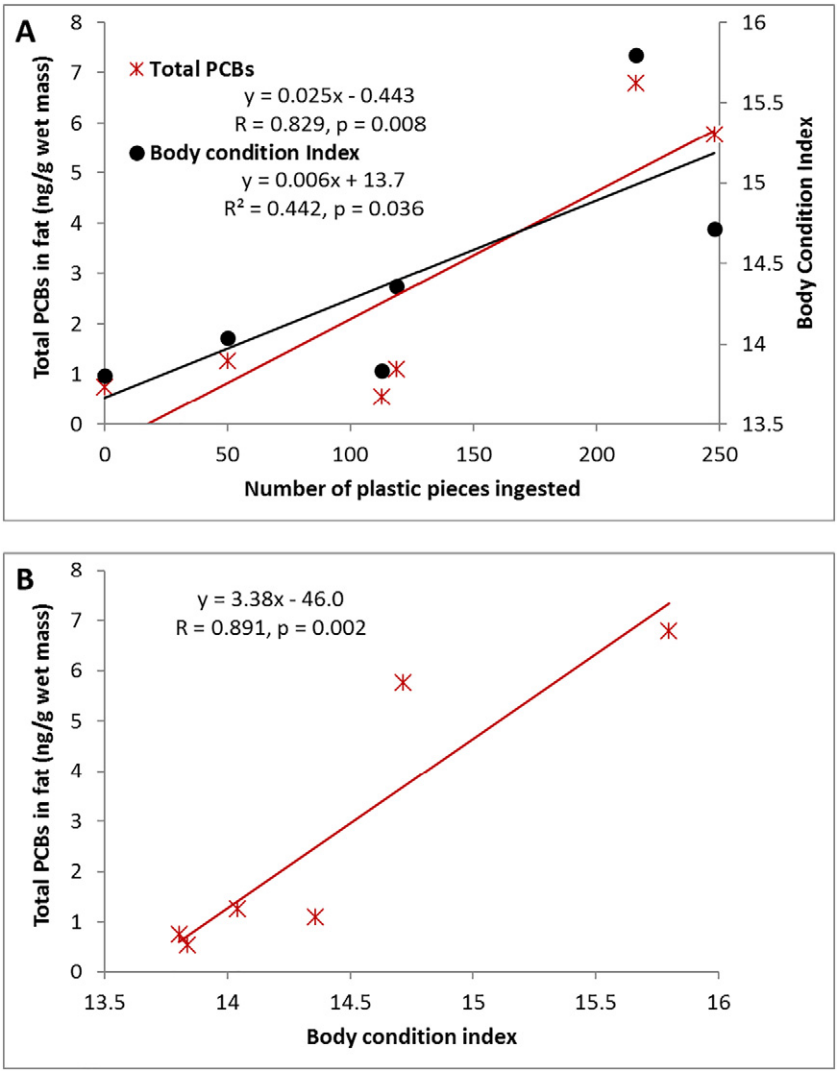

Fig. 5. Inter-correlations among polychlorinated biphenyl (PBC) concentrations in fat, (A) amount of plastic debris ingested and (B) body condition index (kg/cm^3 × 100,000) in pelagic Pacific green sea turtles.

opposite is also possible. In lugworms (*Arenicola marina*), ingested plastics can scavenge phenanthrene and decrease the body burden of this compound. Mass balance studies and pharmacodynamics have not been performed to determine the relative amounts of POPs from natural prey versus plastics; nor have studies examined if plastics moving through the gut of a vertebrate could sorb POPs and serve as an elimination pathway through fecal excretion.

While we know sea turtles eat large amounts of anthropogenic debris (Balazs, 1985; Clukey et al., 2017), it must be a relatively small portion of the overall diet of pelagic Pacific sea turtles in order to maintain the good body condition observed in the turtles in the current study. Thus, it is highly probable that these POP concentrations originated mostly from their prey, resulting in few or no correlations between plastic ingestion amounts and POP accumulation in fat. Additional circumstantial evidence suggests positive covariation among BCI, ingested plastic amounts (Clukey et al., 2017), and POP concentrations in green turtles (Fig. 5A and B). These relationships do not allow us to decipher the source of POP concentrations in the higher exposed turtles from more food (thus higher BCI) or more plastic ingested (possibly because they are ingesting more mass regardless of what they select to eat); however, they call into doubt that plastics are the main route of exposure. Another way to assess the source of POPs is by comparing POP profiles in the turtles to either those in their pelagic prey or in pelagic marine debris. Unfortunately, no studies have analyzed pelagic invertebrates or small fish, the natural prey of these sea turtle species, from this region of the Pacific Ocean for POP concentrations. Without these data, we are left with comparing POP profiles found in plastic marine debris collected from Hawaiian beaches (Heskett et al., 2012; Rios et al., 2007) to those in the sea turtle fat. The assumption from this approach is that greater similarity in profiles would indicate a greater chance that plastic debris is a source of POPs exposure in sea turtles. Four plastic samples from each study were analyzed for POPs, and their average PCB:DDT ratios were estimated at 1.85 (Heskett et al., 2012) and 138 (Rios et al., 2007). The variability between studies and among samples is too large to make solid conclusions, but these ratios are more similar to the green turtle ratio (1.10) and substantially higher than those seen in olive ridley (0.32) and loggerhead sea turtles (0.27). Much more work is needed to determine if plastics are a source or a sink for POPs in marine species.

Taken together, the data in this study suggest that sea turtles in the pelagic Pacific are accumulating POPs most likely from their natural prey, not from ingested plastics. That said, green turtles may have the largest possibility of accumulating POPs from ingested plastic debris. Interestingly, green turtles on average had the highest concentration of α-HBCD compared to the other two species. HBCD is a widely used brominated flame retardant in foam insulation, textiles, and electronics (United Nations Environmental Programme, 2017). However, no significant correlations were found between HBCD and the amounts of plastic

ingested. Future studies examining POP exposure through ingested plastics in this region should focus on green turtles to increase the sample size.

4. Conclusion

Sea turtles captured as bycatch in the Hawaiian and American Samoan longline fisheries provided the largest POPs evaluation to date for three species of pelagic Pacific sea turtles. Despite sample size limitations for loggerhead and green turtles, we were still able to demonstrate expected trends in POP concentrations among the species and geographic locations. Importantly, we established some of the first POP concentrations in tissues of olive ridley sea turtles.

These data will aid scientists and managers in addressing environmental health, global monitoring of POPs and in conservation and management strategies of sea turtles on a greater scale. Whether these turtles are at risk of nearing the toxic threshold for POPs is difficult to determine especially since sea turtles' sensitivity to POPs is not fully understood. Nevertheless, these sea turtles have POP concentrations lower than other central Pacific marine species and lower than sea turtles found near coastal regions.

The hypothesis that ingested plastic can increase levels of POP was not supported by this study; however, our low sample size limited statistical power, especially for green turtles. The majority of POP accumulation detected in these pelagic Pacific sea turtles most likely came from their natural prey. However, the issue of contamination transfer from marine debris should not be disregarded and should be investigated further, especially in pelagic green turtles. The fact that these pelagic turtles had such a high frequency of debris ingestion should highlight a greater concern of the wider problem of plastic pollution in the ocean.

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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.

Appendix A. Supplementary data

Detailed data and information on individual turtles and statistical outcomes are provided in one Supplementary material file. Supplementary data associated with this article can be found in the online version, at http://dx.doi.org/10.1016/j.scitotenv.2017.07.242.

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