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The complex issue of chemicals and microplastic pollution: A case study in North Pacific lanternfish☆

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ABSTRACT

Marine plastic debris, including microplastics (<5 mm in size), comprises a suite of chemical ingredients and sorbed chemical contaminants. Thus, microplastics are a potential, and debated, source of anthropogenic chemicals for bioaccumulation and biomagnification. Several studies have investigated the role of microplastics as a vector of contaminants to marine organisms via modeling exercises, laboratory experiments, and field studies. Here, we examined relationships among chemical contaminants and microplastics in lanternfish (family Myctophidae), an important link in marine food webs, from the North Pacific Ocean as a case study from the field. We compared the body burden of several chemical groups (bisphenol A [BPA], nonylphenol [4-NP], octylphenol [4n-OP], alkylphenol ethoxylates [APEs], pesticides, polychlorinated biphenyls [PCBs], and polybrominated diphenyl ethers [PBDEs]) in fish caught within and outside the North Pacific Subtropical Gyre where plastic is known to accumulate. We also tested whether there was a relationship between chemical concentrations in fish and plastic density at each sampling location. Mean concentrations of common plastic constituents (BPA, 4-NP, 4n-OP, APEs, and total PBDEs) were comparable between myctophids collected within and outside the North Pacific Gyre. Pesticides were higher in lanternfish caught outside the gyre and were associated with lower plastic density. Total PCBs were also higher in fish outside the gyre. In contrast, lower chlorinated PCB congeners were higher in fish residing in the accumulation zone and were correlated with higher plastic density. This finding is consistent with other studies demonstrating an association between lower chlorinated PCBs and plastics in biota and suggests that microplastic may be a transport mechanism for some chemicals in nature.

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1. Introduction

The pervasive accumulation of plastic debris in the oceans is seen as an emergent global environmental disturbance with the potential to disrupt vital biological processes and ecosystem functions (Galloway and Lewis, 2016; Global Environment Facility [GEF], 2012). Microplastics (particles < 5 mm) have, like other chemical pollutants, become a global pollutant (Lohmann, 2017; Rochman, 2018). On the ocean surface, it has been estimated that the number of microplastic particles range from 15 to 51 trillion particles and weigh between 93 and 236 thousand metric tons (van Sebille et al., 2015). The wide variation results from the scarcity of data in most of the world ocean, differences in models, and knowledge gaps in sources and fates of microplastics in the ocean (van Sebille et al., 2015). Due to converging ocean surface currents, some of the highest concentrations of microplastics in the open oceans accumulate in subtropical gyres far from land (Barnes et al., 2009; Lebreton et al., 2018; van Sebille et al., 2015).

In addition to being a physical contaminant, microplastic particles are associated with a cocktail of chemicals that include chemical ingredients and those that sorb from environmental media (e.g., water; Rochman, 2015). As such, they can be both a source and sink for chemical contaminants (Engler, 2012; Hirai et al., 2011; Teuten et al., 2009). The propensity for hydrophobic chemicals, including polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane and its metabolites (DDTs), and polybrominated diphenyl ethers (PBDEs) to sorb to microplastic debris along with chemical constituents and additives incorporated in the

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production of plastic materials, such as bisphenol A (BPA), alkylphenols, and PBDEs, makes microplastic debris a potential source of contaminants for bioaccumulation and biomagnification (Hirai et al., 2011; Mato et al., 2001; Rios et al., 2010; Rochman, 2015; Teuten et al., 2009). Because ingestion of microplastic litter has been documented for hundreds of species of wildlife (GESAMP, 2016), questions and concerns have been raised about the role of microplastic litter in transporting toxic pollutants throughout marine food webs.

Ingestion of plastic litter by marine animals is well recognized, and the number of documented species is mounting (Kühn et al., 2015; Werner et al., 2016). The quantity and frequency of ingestion are likely to increase as the influx of plastic litter to the environment increases (Jovanović, 2017). Plastic ingestion poses physical threats for many species (Jovanović, 2017), but in the case of microplastics, the associated chemicals can result in physiological stress (Browne et al., 2013; Rochman et al., 2013, 2014a; Rochman, 2016; Vos et al., 2000) and can potentially biomagnify in the food web (Diepens and Koelmans, 2018).

Laboratory experiments and observational studies in nature have examined plastic debris as a source of hazardous chemicals to organisms. The transfer of PCBs, PBDEs, and nonylphenol, among other chemical contaminants, from multiple types of plastic has been demonstrated in invertebrates (Browne et al., 2013; Chua et al., 2014; Jang et al., 2016), fish (Rochman et al., 2013) and seabirds (Hardesty et al., 2015; Tanaka et al., 2013; Teuten et al., 2009; Yamashita et al., 2011). The accumulation of plastic-associated chemicals in marine biota from higher trophic levels collected in areas of high plastic density has suggested plastic as a source (Fossi et al., 2012, 2014; Gassel et al., 2013; Tanaka et al., 2013), and Rochman et al. (2014b) showed a significant correlation between plastic density and higher brominated PBDEs in wild-caught prey fish.

While ingestion of microplastic may result in bioaccumulation of organic chemicals, models predict that ingested microplastic may also "clean" the organism if a reverse chemical gradient exists (Koelmans, 2015). Several models also predict that the role of microplastic as a source of contamination in nature is likely insignificant relative to other exposure pathways including prey, suspended particulate matter, and dermal absorption, based on the assumption of equilibrium being reached (Gouin et al., 2011; Koelmans et al., 2016). Still, recognized scenarios in which microplastic may play a larger role than predicted do exist, with the North Pacific Gyre being one such "hotspot" (Chen et al., 2018; Hartmann et al., 2017; Lebreton et al., 2018).

To further examine the role of plastics as a vector of chemical contaminants to biota, we present a case study from the North Pacific Gyre. To measure the extent of contamination of microplastics and chemical contaminants in the North Pacific Subtropical Gyre and the potential role of plastic debris as a source of chemicals to wildlife, we sampled lanternfish (family Myctophidae) from two locations: the North Pacific Subtropical Gyre and the California Current. Members of this family are abundant inhabitants of mesopelagic zones worldwide, and comprise a critical component of pelagic food webs as a key prey species. They also play a significant role in carbon and nutrient cycling (Lusher et al., 2015; Wieczorek et al., 2018). We examined tissue concentrations of several chemicals known to sorb to microplastic and/or used as ingredients in plastics, including PCBs, PBDEs, DDTs, alkylphenols and alkylphenol ethoxylates (APEs), and BPA. We sampled fish to test the hypothesis that myctophids in the North Pacific Gyre, even at a great distance from population centers and industrial sources, would be contaminated with anthropogenic chemical contaminants and explore whether some of that chemical exposure may be due to the presence of microplastics.

2. Methods

2.1. Sample collection

Fish from the family Myctophidae were sampled opportunistically during nightly surface trawls. The fish were captured from August 12 through August 28, 2009 using a manta net with a rectangular opening of 16 cm in height by 61 cm in width with a 3 m long 333 μm mesh net. The net was towed port side along the surface and outside the wake of the vessel. Trawls were towed either for one hour or as a series of four consecutive 15-min periods. The latter pattern was adopted midway into the research trip to minimize damage to fish upon collection. During the last trawl (#18), 114 fish were captured in the first two 15-min periods; therefore, we ended the trawl before conducting the last two 15-min tows. Between one and 29 myctophid fish were caught per trawl (except for trawl #18 as described above), removed from the cod end, measured, wrapped in clean foil, and stored in a −20 °C freezer for future analyses. Sampling locations extended from 38° 30.586′N to 34° 04.208′N and 141° 46.66′W to 139° 53.198′W for trawls #6–14, representing the location within the North Pacific Subtropical Gyre, and from 38° 29.487′N, 125° 18.279′W to 38° 28.574′N, 125° 45.844′W for trawl #18, representing the location within the California Current (Fig. 1).

2.2. Taxonomic identification

In the laboratory, myctophid fish were removed from the freezer briefly to extract a piece of caudal fin for genetic identification. Lanternfish were analyzed for mitochondrial DNA gene sequencing of the cytochrome oxidase subunit I (cox1) at the Marine Fisheries Genetics Laboratory, Hatfield Marine Science Center, Oregon State University, Oregon, USA (using the methods in Ward et al., 2005). A small number of samples with unsatisfactory (<95%) similarity to sequences for Myctophidae in the Barcode of Life Database (Ratnasingham and Hebert, 2007) were sent for visual taxonomic identification by P. Davison at the Scripps Institution of Oceanography.

2.3. Plastic density

Plastic density (number of pieces/km^2) at each sampling location was estimated by linearly interpolating a set of three different debris accumulation models. Accumulation patterns from these models, which were either based on the trajectories of drifting buoys in the global ocean (Maximenko et al., 2012; van Sebille et al.,

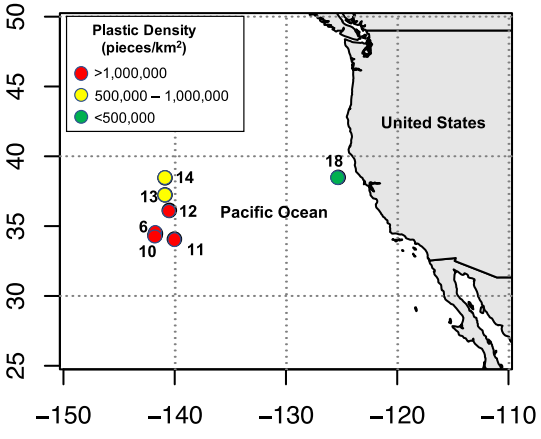

Fig. 1. Map of sampling locations in the North Pacific.

2012), or on numerical simulations with fine-resolution ocean modes (Lebreton et al., 2012), were then regressed onto a set of more than 11,000 manta trawl collections of small floating plastic. This yielded three different estimates of the total amount of floating plastic in the global ocean on a 1 × 1 degrees grid, for the year 2010. Because we were interested in patterns among sampling locations, we used one estimate, since patterns were similar amongst locations. See van Sebille et al. (2015) for a full description of the method and dataset.

2.4. Sample preparation

In the laboratory, thawed fish were dissected to remove the digestive tracts, measured and weighed prior to and following dissection, then rewrapped in foil and stored at −20 °C for chemical analysis.

We collected large numbers of lanternfish, but only 23 were large enough to provide sufficient tissue for chemical analysis as discrete samples. These 23 samples were all collected offshore in the gyre. To achieve adequate tissue mass, 69 smaller lanternfish from the gyre (26–76 mm total length) were combined to make five composite samples of four to 29 fish each using similarly sized fish per sample. It was necessary to select gyre fish collected in multiple trawls from different nights to form composite samples with sufficient tissue mass. Ninety-nine lanternfish collected from the California Current comprised six composite samples, also based on size, with five to 34 fish per sample. Fish samples are shown in Supplementary Material Tables S1-S3.

2.5. Chemical analysis

AXYS Analytical Services Ltd. performed extractions and analyses of fish tissue samples. For each batch, procedural blanks comprised of clean canola oil were sent through the same procedure and analyzed for the same suite of chemicals as each fish sample. The canola oil was food grade; however, the laboratory ran proofs for all methods for which the oil would be used to ensure cleanliness.

All solvents and reagents for sample prep were purchased from Fisher Scientific (Fisher Scientific, Fair Lawn, NJ, USA) for analytical chemistry by AXYS Analytical Services Ltd. All solvents and reagents were equal or above pesticide grade. Surrogate standards used for analytical methods included bisphenol A-propane-d6 for analysis of BPA, $^{13}C_6$-4-nonylphenol for analysis of alkylphenols, $^{13}C_6$-NP1EO for analysis of alkylphenol ethoxylates, $^{13}C_{12}$-labeled DDTs (o,p-DDE, p,p-DDE, o,p-DDT, p,p-DDT, and p,p-DDD) for analysis of DDTs, ^{13}C-labeled PCBs (CB#s 1, 3, 4, 15, 37, 54, 77, 81, 104, 105, 114, 118, 123, 126, 155, 156, 157, 167, 169, 170, 180, 188, 189, 202, 205, 206, 208 and 209) for analysis of PCBs, and $^{13}C_{12}$-labeled PBDEs (BDE#s 15, 28, 47, 77, 99, 100, 126, 153, 154, 183, 197 and 209) for analysis of PBDEs. ^{13}C-labeled PCB#s 28, 111 and 178 and ^{13}C-labeled PBDE# 139 were used as clean-up standards during extraction of PCBs and PBDEs, respectively. Recovery standards used for analytical methods included d6-BPA for the analysis of BPA, $^{13}C_6$-NP1EO for the analysis of alkylphenol ethoxylates, $^{13}C_6$-4-NP for the analysis of alkylphenols, $^{13}C_{12}$-PCBs (CB#s 52 and 138) for the analysis of DDTs, $^{13}C_{12}$-PCBs (CB#s 9, 52, 101, 138 and 194) for the analysis of PCBs, and $^{13}C_{12}$-PBDEs (BDE#s 79, 180 and 206) for the analysis of PBDEs.

Homogenized tissues from individual fish or composite samples of multiple fish (Supplementary Material Tables S1-S3) were subsampled for analyses of BPA, alkylphenolics (4-nonylphenol, 4n-octylphenol, 4-nonylphenol monoethoxylate and 4-nonylphenol diethoxylate), pesticides (2,4′-DDT, 4,4′-DDT, 2,2′-DDE, 4,4′-DDE, 2,2′-DDD, 4,4′-DDD), PCBs (CB#s 1–209) and PBDEs (BDE#s 7, 8, 10–13, 15, 17, 25, 28, 30, 32, 33, 35, 37, 47, 49, 51, 66, 71, 75, 77, 79, 85, 99, 100, 105, 116, 119, 120, 126, 128, 138, 140, 153, 154, 155, 166, 181, 183, 190, 196, 197, 203, 204, 206–209). Extraction and analysis procedures were in accordance with AXYS Method MLA-084 for BPA, AXYS Method MLA-080 Rev 2 for alkylphenols and alkylphenol ethoxylates, AXYS Method MLA-028 Rev 6 for pesticides, AXYS Methods MLA-013 and MLA-010 (EPA Method 1668; USEPA, 2010a) for PCBs, and AXYS Methods MLA-013 and MLA-033 (EPA Method 1614; USEPA, 2010b) for PBDEs.

The analyses of BPA and alkylphenols were performed as described in Rochman et al. (2014b) except that 0.67–2.08 g wet weight (ww) and 0.53–1.89 g ww of homogenized tissue were subsampled from each individual fish for analysis of BPA and alkylphenols, respectively. The remaining homogenized tissue samples, approximately 3–4 g ww, were prepared for the analysis of pesticides, PCBs, and PBDEs. Samples were spiked with ^{13}C-labeled surrogate standards and Soxhlet extracted using dichloromethane. Duplicate gravimetric lipid determinations were made using aliquots of this extract. Column chromatography was employed to cleanup extracts using one or more of the following columns: multi-layered acid/base silica, Florisil, alumina, Biobeads, and 4.5% carbon/Celite. Extracts were spiked with isotopically labeled recovery standards prior to instrumental analysis. Pesticides were analyzed by high-resolution mass spectrometric detection (HRMS) with a 40 μL extract volume. The remaining extract for PCBs and PBDEs was spiked with ^{13}C-labeled cleanup standards, concentrated to 20 μL for PCBs and later diluted to 50 μL for PBDEs for analysis by high-resolution gas chromatography (HRGC) with HRMS.

2.6. Instrumental analyses

Analyses were performed as described in Rochman et al. (2014b). Briefly, BPA and alkylphenolics were analyzed using a high performance liquid chromatograph coupled to a triple quadrupole mass spectrometer (LC-MS/MS), and PCBs and PBDEs were analyzed using a HRMS coupled to an Agilent 6890 + gas chromatograph. In addition, analysis of pesticides was performed on a high resolution GC/MS, operated at a static (8000) mass resolution (10% valley) in the electron ionization (EI) mode using multiple ion detection (MID).

2.7. Quality assurance and quality control

All surgical tools were solvent rinsed in acetone and hexane between dissections of each individual fish. All extraction tools were rinsed in acetone, toluene, dichloromethane and methanol. Analysis glassware was cleaned with water and detergent, baked at 350 °C for 10 h and solvent rinsed (3 × each methanol, toluene, hexane) before use. Each analysis batch (up to 20 field samples of a single sample matrix) included at least two quality control samples: a procedural blank and a known or control sample of a similar matrix as the field samples. Laboratory procedural blanks for extraction and spiked matrix blank (canola oil) were extracted and run with the analytical batch. DDTs were undetected in extraction blanks. Blank levels of BPA, alkylphenols, alkylphenol ethoxylates, PCBs, and PBDEs measured in extraction blanks were subtracted from the reported concentrations extracted from samples.

The following quality control criteria were used to guarantee correct identification of BPA: BPA must have an LC retention time within 0.1 min of the ^{13}C-labeled BPA, it must have a parent ion with a specific mass to charge ratio, and that parent ion must produce 2 daughter ions of specific mass to charge ratio, a signal-to-noise ratio was greater than 3 for the parent to daughter ion transitions and the retention time must be within ±0.4 min of the

predicted retention time from the mean determination from the Initial Calibration. To guarantee correct identification of alkylphenols and alkylphenol ethoxylates, the following quality control criteria were used: each target analyte must have a parent ion with a specific mass to charge ratio, and that parent ion must produce daughter ions of specific mass to charge ratio, LC retention times must be within ±0.4 min of the predicted retention time from the mean determination from the daily bracketing calibration standard, and signal-to-noise ratio was greater than 3 for the parent to daughter ion transitions. The following quality control criteria were used for correct identification of DDTs: peak responses must be at least three times the background noise level, the relative retention time must be within the predicted window, peak centroids for the quantification and confirmation ions must coincide within two sec, and the relative ion abundance ratios must be within 20% of the theoretical. The quality control criteria identified in Section 16.0 of EPA Method 1668 and Section 16.0 of EPA Method 1614 were used to guarantee correct identification of PCBs and PBDEs respectively: the signals for the specified mass to charge ratio for the quantification and confirmation ions must be present and must maximize within the same 2 MS scans, the ratio of their intensities must be within 15% of the specified ratio, their relative retention time (with respect to the labeled surrogate) must be within method specifications and signal-to-noise ratio was greater than 2.5.

Quantification of all analytes was carried out by isotope dilution internal standard where exact labeled analogue standards are available or by internal standard quantification using the labeled analogue of a related compound when exact labeled analogues are not available. This quantification protocol produces analytical results that are corrected for any losses during sample workup and any fluctuations of instrumental response due to the presence of sample matrix in the extracts. The reported concentrations of BPA, alkylphenols, APEs, DDTs, PCBs, and PBDEs are recovery corrected based upon the recovery efficiencies of surrogate standards.

The limit of quantification (LOQ) ranged from 0.5 to 1.8 ng/sample (ww) for BPA, 0.5–7.7 ng/sample (ww) for alkylphenols, 0.5–2.4 ng/sample (ww) for alkylphenol ethoxylates, 0.001–0.13 ng/sample (ww) and 0.02–5.1 ng/sample (lw) for DDTs, 0.1–3.4 pg/sample (ww) and 1.8–132 pg/sample (lw) for PCBs, to 0.25–94 pg/sample (ww) and 3.5–1480 pg/sample (lw) for PBDEs. LOQs represent signal to noise greater than 2.5. Note that larger limits for PBDEs are mainly attributed to BDE209. The higher LOQs for BDE-209 are due to small sample sizes.

The mean (range%) recoveries of the surrogate standards ranged from 69% (66–71) for BPA, 77% (66–98) for alkylphenols, 68% (59–75) for alkylphenol ethoxylates, 98% (92–103) for DDTs, 79% (44–104) for PCBs to 70% (47–98) for PBDEs. The recovery of spiked matrix blank samples ranged from 22 to 67% for BPA, 74–93% for alkylphenols, 57–87% for alkylphenol ethoxylates, 54–77% for DDTs, 84–101% for PCBs to 89–103% for PBDEs.

2.8. Statistical analyses

We compared chemical concentrations between lanternfish collected in and out of the subtropical gyre, that is, those collected offshore and those from the California Current. We included both composite and discrete samples from the gyre so that comparisons would not be limited to single- versus multiple-fish samples. The size range of fish in the composite samples from the gyre was also comparable to that of the fish in the California Current samples. To measure differences among chemical body burdens for each chemical class in fish sampled in and out of the gyre, all concentrations were $[x]^{1/6}$ transformed to achieve normality, and compared using a Two-Sample T-test Assuming Unequal Variances (Microsoft Excel, 2010), which is recommended for smaller sample sizes and when variances are unequal, as was the case for some of the chemicals analyzed (Ruxton, 2006).

We performed simple linear regression with untransformed data to examine the correlation between modeled plastic density estimates at each sampling location and chemical body burden in fish tissues. Only the 23 samples of individual myctophids, which represented discrete sampling locations within the accumulation zone (locations #6 and #10–14), were used in addition to the six composite samples of lanternfish collected at location #18 in the California Current.

2.9. Analysis of plastic in stomach

Six lanternfish collected in the gyre were selected for examination of stomach contents as part of a method development study for extraction and identification techniques of microplastics in fish. This method is described in detail in Wagner et al. (2017). It was not a goal of that study to measure total numbers of microplastics per fish. Therefore, the analysis provides a partial, not thorough survey of plastic particles in the myctophid stomachs. The results provide a proof-of-concept that microplastics were ingested by lanternfish used in this study. Complementary methods were used to characterize plastic type, size, and morphology of microplastics using optical microscopy, scanning electron microscopy plus energy-dispersive X-ray spectroscopy (SEM/EDS), and Fourier-transform infrared micro-spectroscopy (FTIR). Stomachs were dissected and visually screened for ingested polymer particles under a stereo-zoom microscope. Rigid particles >1 mm, visually resembling microplastics, were placed in a clean glass petri dish for SEM. Stomach contents were extracted using pulsed ultrasonic extraction and filtered onto 10 μm pore size polycarbonate (PC) filters. Representative subsamples were obtained from each PC filter by cutting out randomly selected, 1 cm^2 squares. Particles were randomly selected from each filter subsample for analysis. No attempt was made to measure fractions of filter areas analyzed or total numbers of microplastics per fish. FTIR was conducted on individual, 15 μm–5 mm particles using a Thermo Electron Nicolet Continuum FTIR Microscope with 10× optical objective, 15× IR objective, and a Nexus 470 spectrometer.

3. Results

3.1. Taxonomic identification

Several species of myctophids were identified both within and outside the gyre. However, nearly all samples were *Myctophum nitidulum* and *Tarletonbeania crenularis* collected in the gyre and in the California Current, respectively. Only these two species were used in subsequent analyses.

3.2. Plastic density among locations

Estimates of plastic density were highest for locations #6 and #10–12 (>1,000,000 pieces/km^2 and up to 1,585,300 pieces/km^2; Fig. 1). Density estimates at locations #13 and 14 were lower (927,670 and 709,000 pieces/km^2, respectively) but over six times higher than for location #18 (114,650 pieces/km^2; Fig. 1). As such, we expect fish caught in the subtropical gyre to be exposed to much greater concentrations of plastic debris than fish caught in the California Current.

3.3. Chemical body burden

3.3.1. Chemical concentrations

PCBs, PBDEs, and DDTs were detected in all samples from all

locations. The other chemicals analyzed were detected in some of the samples from all locations in and out of the gyre with the exception of 4n-octylphenol, which was not observed in the five composite samples collected in the gyre (Table 1; Supplementary Table S4). The mean chemical concentrations in composite versus discrete samples of fish from the gyre showed similar results; therefore, we combined the results for all gyre fish to compare to fish from the California Current for comparisons in and out of the gyre (Table 2).

3.3.2. Comparisons of chemical body burden inside and outside of the gyre

Mean concentrations of BPA, 4-NP, 4n-OP, and APEs were not significantly different between myctophids collected within the subtropical gyre (i.e., more plastic) and out of the gyre (i.e., less plastic), as shown in Fig. 2.

Significantly higher concentrations of total PCBs were found in lanternfish from the California Current compared to those collected in the gyre ($t[32] = 3.6$, $p < 0.001$). In contrast, significantly greater concentrations of lower chlorinated PCB congeners (mono to tetra) were found in lanternfish collected in the gyre versus in the California Current ($t[20] = 4.4$; $p < 0.001$). The sum of PBDEs did not differ between lanternfish from in and out of the gyre ($p > 0.05$). Concentrations of higher brominated PBDEs (183–209) were significantly greater in lanternfish residing outside the gyre in the California Current ($t[15] = 3.3$, $p < 0.01$). The sum of DDTs was significantly higher in lanternfish residing outside the gyre in the California Current ($t[24] = 10.5$, $p < 0.0001$). Results from comparisons of all mean chemical concentrations in and out of the gyre are shown in Fig. 2.

3.3.3. Relationship between plastic density and chemical concentrations in fish

We observed relationships between plastic density and fish tissue concentrations for certain chemical groups. Lower chlorinated PCBs (mono to tetra) were significantly greater in locations with greater plastic density ($R^2 = 0.159$, $p = 0.032$; Fig. 3g). A similar but insignificant trend was observed with 4n-OP and plastic density ($R^2 = 0.108$, $p = 0.081$; Fig. 3c). Greater concentrations of higher brominated BDEs, sum of 183–209, were associated with lower plastic density ($R^2 = 0.232$, $p = 0.009$; Fig. 3i). DDTs were also significantly greater where plastic density was lower ($R^2 = 0.488$, $p < 0.0001$; Fig. 3e). No relationship was observed between plastic density and tissue concentrations of BPA, APEs, 4-NP, total PCBs, or total PBDEs ($p > 0.05$). Results from linear regression analyses for all chemicals are shown in Fig. 3.

3.4. Stomach contents

Four particles in two fish were identified as microplastics by FTIR. A sample from lanternfish M080, collected in trawl #12, contained one 750-μm piece of polyethylene (PE), and lanternfish M143, sampled in trawl #13, had three pieces of microplastic comprised of 1) PE, 750 μm 2) polypropylene (PP), 4 mm, and 3) a combination of PE and PP, 750 μm. The prevalence of microplastic in this small sample set of lanternfish stomachs was 2/6 (33%). Furthermore, only portions of fish stomach contents were examined; therefore, the number of microplastics identified is likely only a partial count. Fish M143, with three pieces of plastic identified from its stomach, had a greater concentration of lower chlorinated PCBs (11 ng g^{-1} lw) compared to the other fish (0.87–4.04 ng g^{-1} lw).

4. Discussion

All chemicals analyzed were found in fish collected from the gyre and the California Current.

In comparison to other studies of contaminants in myctophid fish (Table 3), mean concentrations of PCBs and DDTs were lower in the North Pacific Gyre than in the western North Pacific (Takahashi et al., 2010; de Brito et al., 2002), but PBDE levels were comparable to those in lanternfish in the western North Pacific (Takahashi et al., 2010) and Antarctica (Borghesi et al., 2009). Compared to myctophids in the South Atlantic accumulation zone, concentrations of APEs, BPA, and total PCBs were similar, but alkylphenols were higher in the North Pacific and total PBDEs were higher in South Atlantic fish (Rochman et al., 2014b).

Models predict that fish living outside the gyre, in the California Current, will be exposed to lower plastic density. We found some chemicals in fish tissues to be associated with lower plastic density (DDTs, PCBs, sum of BDEs 183–209), documenting that plastic is not a main source of all chemicals to myctophid fishes. This trend was not the same for all chemicals analyzed in this study, however, as some chemicals were correlated with higher plastic density (lower chlorinated PCBs). Furthermore, many of the chemicals analyzed were found at comparable levels regardless of plastic density (BPA, APEs, 4-NP, total PCBs, total PBDEs), demonstrating the complexity of the system.

The significantly higher concentrations of DDTs and total PCBs in myctophids collected outside the gyre could be explained by the proximity of these fish to the west coast of the U.S.A. where sediment contamination with DDTs and PCBs is well-known (USEPA, 2012; Yamashita et al., 2018; Zeng and Tran, 2002). In general, higher contaminant concentrations are expected near coastal areas where there is substantial historical production and use of these chemicals (Engler, 2012).

Higher concentrations of BDEs 183–209 in fish living outside the gyre may suggest recent exposure with minimal debromination of congeners in the California Current (Stapleton et al., 2004) and might also be explained by proximity to land-based sources with recent and/or historical production and use. In 2008, prior to this study, legislation in California prohibited the manufacture, processing, and distribution of products containing the commercial formulations pentaBDE and octaBDE (AB 302, Chan, 2003) such that only the higher brominated formula (decaBDE) could be used in the state. The greater concentrations of higher brominated BDEs outside the gyre, suggesting that plastic is not a measurable source of PBDEs to fish in this case study, are in contrast with previous

Table 1
Percent of chemicals detected.

Sample Location	Number of Fish	Mean % Lipid	Percent of Samples with Chemical Detection						
			BPA	APEs	4-NP	4n-OP	Total PCBs	Total PBDEs	DDTs
Gyre	23	5.37	52	57	70	39	100	100	100
Gyre	5[a]	2.82	20	40	20	0	100	100	100
California Current	6[a]	3.40	17	33	33	17	100	100	100

[a] Indicates composite samples.

Table 2
Mean chemical concentrations for samples collected from the North Pacific Gyre and the California Current.

Chemical	Fish and Source Location			
	Individuals—Gyre	Composites—Gyre	Combined[a] Samples—Gyre	Composites— California Current
	Mean Concentration (Standard Deviation)			
BPA (ng g^{-1} ww)	1.08 (1.38)	0.686 (1.53)	1.01 (1.39)	1.28 (3.14)
APEs (ng g^{-1} ww)	2.50 (3.16)	2.09 (3.08)	2.43 (3.10)	1.75 (3.48)
4n-OP (ng g^{-1} ww)	0.803 (1.17)	0 (0)	0.66 (1.10)	0.033 (0.082)
4-NP (ng g^{-1} ww)	10.6 (11.7)	7.18 (16.1)	9.98 (12.3)	11.3 (24.8)
Sum DDTs (ng g^{-1} lw)	17.8 (16.1)	19.6 (8.90)	18.1 (15.0)	61.3 (11.1)
Total PCBs (ng g^{-1} lw)	22.4 (17.0)	22.2 (5.59)	22.4 (15.5)	28.8 (2.82)
Lower Cl PCBs (ng g^{-1} lw)	3.95 (2.21)	0.785 (0.192)	3.39 (2.34)	1.03 (0.450)
Sum PBDEs (ng g^{-1} lw)	2.31 (3.14)	3.57 (1.61)	2.53 (2.95)	5.76 (6.53)
BDEs 183–209 (ng g^{-1} lw)	1.20 (2.28)	3.51 (1.57)	1.61 (2.33)	5.22 (6.41)

[a] Includes individual and composite samples.

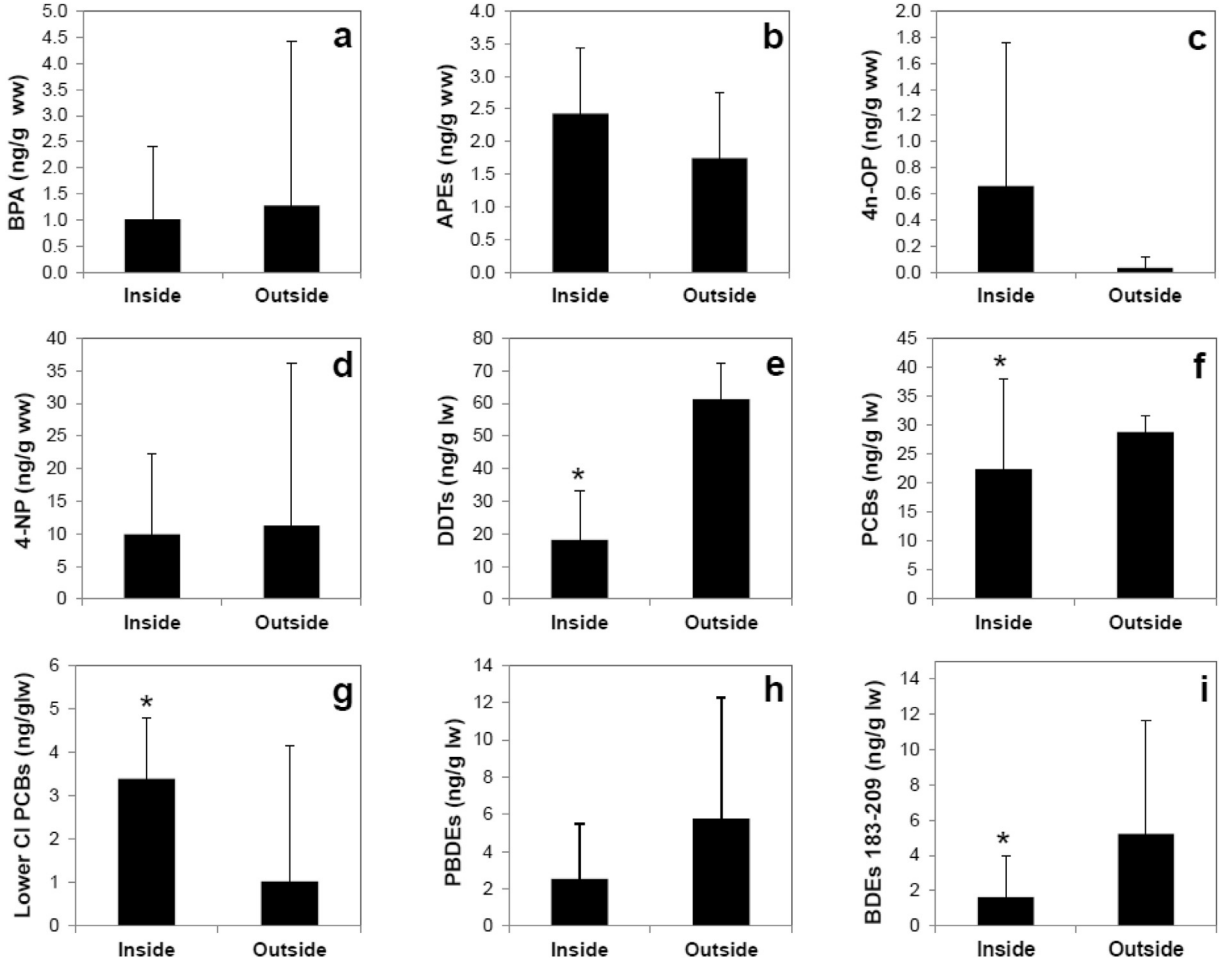

Fig. 2. Comparisons of mean chemical concentrations in lanternfish from inside the gyre (composite and individual fish samples) and outside the gyre for a) BPA, b) APEs, c) 4n-OP, d) 4-NP, e) DDTs, f) PCBs, g) lower chlorinated PCBs, H) PBDEs, and i) BDEs 183–209. Asterisks indicate significant differences in concentrations inside and outside the gyre for DDTs ($p < 0.0001$), PCBs ($p < 0.001$), lower chlorinated PCBs ($p < 0.001$), and higher brominated BDEs ($p < 0.01$) using the Two-sample T-test Assuming Unequal Variances. Note that y-axis scales differ among graphs.

findings of higher brominated BDE congeners in fish from the North Pacific Gyre (Gassel et al., 2013) and from the South Atlantic gyre, where higher brominated BDEs were positively correlated with plastic density (Rochman et al., 2014b).

In this study, we found no trend related to lanternfish location and BPA, alkylphenols, APEs, and total PBDEs. Fish sampled from inside and outside of the North Pacific Gyre showed no significant differences in contamination by these chemical groups, although we did see a tendency toward higher concentrations of 4n-OP inside the gyre. Although the high variability among samples may have contributed to the overall lack of trend, this result is curious as one might expect greater concentrations of these chemicals outside of the remote gyres. Because these chemicals are typically associated with wastewater treatment plants (de Wit, 2002; Soares et al., 2008; Wright-Walters et al., 2011), higher concentrations near land may be expected. Although long-range transport could account for

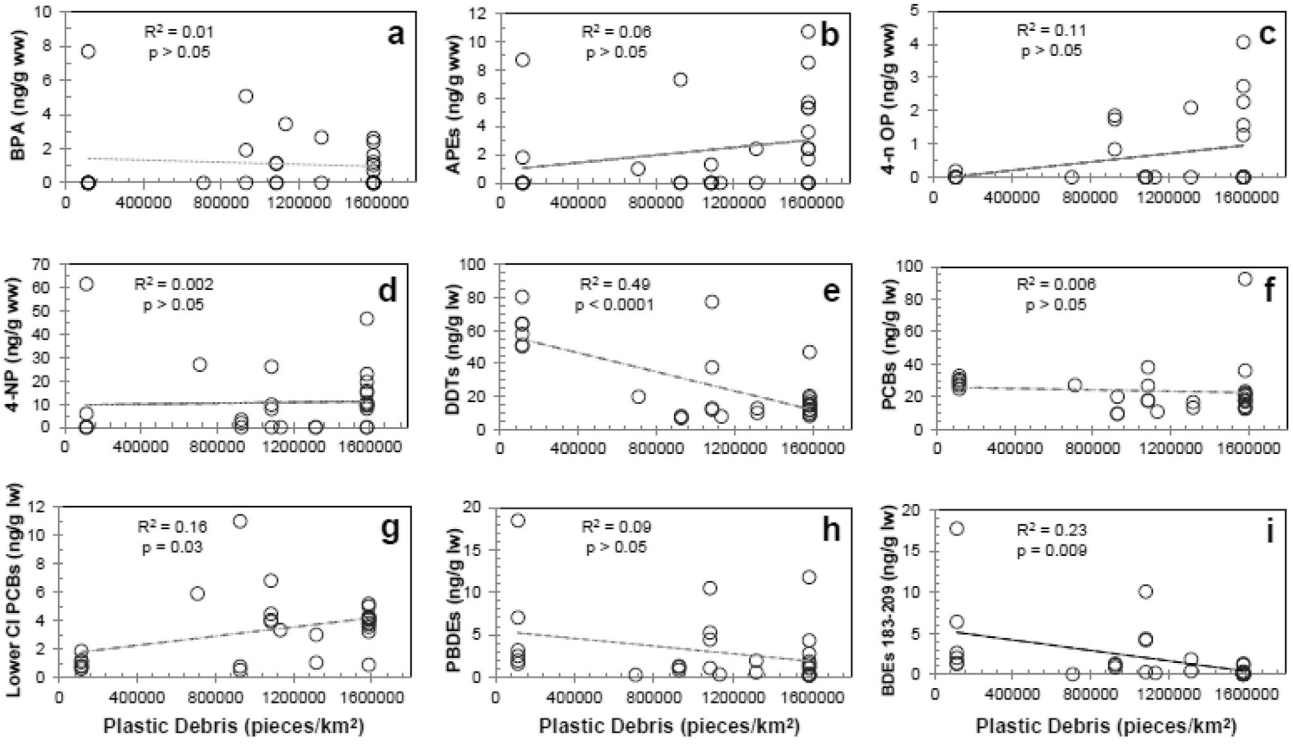

Fig. 3. Correlations between predicted plastic density and chemical concentrations in fish tissue for a) BPA, b) APEs, c) 4n-OP, d) 4-NP, e) DDTs, f) PCBs, g) lower chlorinated PCBs, h) PBDEs, and i) higher brominated BDEs. Note that y-axis scales differ among graphs.

Table 3
Comparison of chemical concentrations in myctophids.

Mean concentrations (ng g^{-1} lipid weight)[a]							
Location	Depth	Species	% Lipid	PCBs	DDTs	PBDEs	Reference
N. Pacific Gyre	surface	*M. nitidulum*	4.9	22	18	2.5	This study
CA Current	surface	*T. crenularis*	3.4	29	61	5.8	
Western N. Pacific Off- Tohoku, Japan	bathypelagic 480 m	*Lampanyctus jordani*	11	34	47	1.3	Takahashi et al. (2010)
Western N. Pacific Off-Tohoku, Japan	upper bathypelagic/micronektonic 1000 m	*L. jordani*	9.8	37	104.5		de Brito et al. (2002)

Range (ng g^{-1} wet weight)							
Location	Species	BPA	APs	APEs	PCBs	PBDEs[b]	Reference
N. Pacific Gyre	*M. nitidulum*	nd-5.1	nd-47	nd-11	0.55–3.4	0.007–0.05	This study
CA Current	*T. crenularis*	nd-7.7	nd-62	nd-8.7	0.84–1.3	0.05–0.59	
S. Atlantic Gyre	*M. phengodes, Symbolophorus* sp.	nd-6.2	nd-10.8	nd-5.4	0.018–5.97	0.002–11.2	Rochman et al. (2014b)
Antarctica	*G. nicholsi*					0.089 ± 0.015	Borghesi et al. (2009)

[a] The numbers of congeners/isomers analyzed for PCBs, DDTs, and PBDEs in this study were 41, 6, and 13, respectively, and 62, 3, and 14, respectively, in Takahashi et al. (2010). de Brito et al. (2002) reported 117 PCB congeners and 3 DDT isomers.
[b] The number of PBDE congeners reported was 13 in this study and Rochman et al. (2014b) and 23 in Borghesi et al. (2009).

the presence of these chemicals that we observed in offshore fish (GESAMP, 2010), it is unlikely that concentrations would be comparable to levels near major population centers and industrial sources. Alternatively, because BPA, APEs, and PBDEs are chemical additives in plastic (Rochman et al., 2014b), and can be transported long distances while sorbed on floating plastic debris (Engler, 2012), it is possible that exposure to contaminated plastic in locations with higher plastic density contributed to the concentrations found in gyre fish.

While total PCBs were higher in lanternfish from the California Current, lower chlorinated PCBs were the only chemical group with significantly higher concentrations in fish from the gyre and positively correlated with plastic density. Hirai et al. (2011) noted that plastic fragments sampled from the North Pacific Gyre had more tri-and tetra-chlorinated congeners compared to beached samples that had more penta-to hepta-chlorinated congeners. Rios et al. (2010) also reported the predominance of lower chlorinated congeners in plastic samples from the North Pacific Gyre. This suggests that in this case study, plastics may have been a measurable source of lower-chlorinated PCBs. Lower chlorinated PCB congeners are more like than higher chlorinated PCBs to associate with plastic due to their lower hydrophobicity and lower molecular weight

(Rochman et al., 2013). Lower chlorinated PCB congeners may also be more easily metabolized and eliminated, as they pass through trophic levels, and therefore less abundant in fish tissue as a function of eating prey (Teuten et al., 2009). Thus, the higher concentrations of lower chlorinated PCBs in gyre fish may be coming from non-prey items such as microplastics in this case.

For lower chlorinated PCBs, the following congeners comprised the largest percentage of the sum of lower chlorinated PCBs in fish from the gyre and the California Current: PCBs 52, 61, 66, 20, 49, and 31. These six congeners combined represented 50%, 62%, and 67% of lower chlorinated PCBs in gyre individuals, gyre composite samples, and California Current fish, respectively. However, PCB congener 11 constituted ten and three percent of lower chlorinated PCBs in individuals and composite samples from the gyre, respectively, but less than 0.1% in fish from the California Current. Similarly, PCB-44 made up eight percent in gyre individuals, was not present in composite samples from the gyre, and comprised less than one percent in fish from outside the gyre (See Supplementary Table S5). Thus, in general, the profiles of lower PCB congeners were similar in fish from inside and outside the gyre. However, one notable exception, PCB-11, represented up to ten percent of lower chlorinated congeners in the gyre fish but was trivial in the California Current lanternfish. This congener is not a dechlorination product of Aroclors, but is a by-product of pigment manufacturing (specifically, diarylide yellow), and has been detected in air and wastewater (Hu and Hornbuckle, 2010). PCB-11 is one of many "non-legacy PCBs" originating from pigments used mostly in the coloration of paints, printing inks, and plastics, but also in paper, textiles, cosmetics, crayons, and building materials (Rodenburg et al., 2015).

Laboratory investigations have demonstrated the possibility of transfer of contaminants from plastic, e.g., PBDEs in amphipods (Chua et al., 2014) and fish (Rochman et al., 2013); nonylphenol, phenanthrene, and Triclosan in lugworms (Browne et al., 2013); and PCBs in lugworms (Besseling et al., 2013). Field studies have also provided evidence for plastic as a source of certain chemicals in nature. Phthalates, common plasticizers, were found in high concentrations in euphausiid prey and large filter feeders (fin whale and basking shark) in the Mediterranean (Fossi et al., 2014) and were correlated with the number of plastic pieces ingested by seabirds (Hardesty et al., 2015). Chemicals added to plastic as flame retardants have been associated with biota exposed to plastic. Jang et al. (2016) identified a relationship between hexabromocyclododecane (HBCD) flame retardants and expanded polystyrene (EPS) buoys in mussels living on the EPS substrate. Their findings were supported both by concentrations and isomeric profiles of HBCD. Gassel et al. (2013) found greater concentrations of BDEs 183–209 in juvenile yellowtail from the North Pacific Gyre, and Rochman et al. (2014b) observed fish tissue concentrations of higher brominated PBDE congeners that were positively correlated with plastic density. Tanaka et al. (2013) detected higher-brominated PBDE congeners in seabirds; the same congeners were not present in the natural prey of the seabirds but were measured in plastic from the stomachs of the birds.

Still, modelers and other researchers have argued that the proportion of hydrophobic organic chemicals sorbed by plastics is small compared to other media in the ocean and that other exposure pathways, such as dermal absorption, suspended particulate matter, and contaminated prey, are more important routes (Besseling et al., 2017; Gouin et al., 2011; Koelmans et al., 2016; Lohmann, 2017). Koelmans et al. (2016) recognized various carbon-based media such as dissolved organic carbon, organic colloids, black carbon, and biota in the marine environment that also sorb and transfer chemicals and calculated the fraction of hydrophobic organic chemicals sorbed by plastics to be small compared to that sorbed by other media in the ocean. They estimated that plastic concentrates chemicals by factors up to 10^7 from the water but concluded that plastic is irrelevant for the transport of chemicals because the mass of water is about a factor of 10^{13} larger than that of plastic (Koelmans et al., 2016). Koelmans et al. (2016) also asserted that for most habitats, bioaccumulation of chemicals from natural prey would contribute far more than from ingested microplastic.

However, in a recent study from a similar system, Chen et al. (2018) calculated plastic mass to biomass ratios and determined that microplastics outweigh prey in the same size range (0.5–5 mm) by 40 times (and by 180 times if all buoyant plastic and biota >0.5 mm are considered). They thereby showed that microplastics at the surface may make up a significant dietary contribution. Furthermore, using fugacity ratios calculated between predators and plastic samples captured within the North Pacific Gyre, they provided support for the role of plastics in transferring chemicals to marine organisms in the North Pacific accumulation zone (Chen et al., 2018). Differentiating between exposure to contaminants via plastic and prey is challenging. Even so, the most common prey of mesopelagic fish—copepods, euphausiids, amphipods, larvae, and decapods—have all been reported to ingest microplastics (Wieczorek et al., 2018) such that some of the contamination of prey could be attributed to ingestion of microplastics, and trophic transfer may also contribute to body burden. Given the abundance of myctophid fish in oceans and their crucial role as a prey species, bioaccumulation of chemicals from plastics has a potentially important role in open oceanic gyres.

In conclusion, we present three lines of evidence that microplastics can be a source of chemical exposure for lower-chlorinated PCBs in this system. Lower chlorinated PCB congeners (mono to tetra) were significantly higher in lanternfish in the gyre and were positively correlated with plastic density. The concentration of lower chlorinated congeners was also greater in fish M-143, in which three pieces of plastic were identified in the stomach, compared to the other fish whose stomachs were examined. Teuten et al. (2009) initially demonstrated the transfer of PCBs, especially lower chlorinated congeners, from ingested plastics to the tissue of seabird chicks and showed that lower chlorinated congeners can be an indicator of the contribution of PCBs from plastic to body burden. Similarly, the mass of plastic ingested by adult seabirds was positively correlated with concentrations of lower-chlorinated congeners (with up to 4 chlorines), but not with higher chlorinated congeners or total PCBs (Yamashita et al., 2011). Our results concur with these other studies suggesting that plastic can be a source of exposure to lower chlorinated PCBs.

Results from studies assessing the role of plastic as a transport mechanism for contaminants relative to other media have been mixed, reflecting the complexity of factors involved (Rochman, 2016). In nature, the role of plastic debris appears to be complicated, with some studies suggesting that for some plastics, in some locations, with some species and for some chemicals, plastics may be a source of chemicals to wildlife. Continued studies could help delineate these components, as well as probe questions regarding plastics as a source of additive chemicals to wildlife and any mixture effects from the physical particles and their chemical components.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.03.002.

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