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Persistent organic pollutants in plastic marine debris found on beaches in San Diego, California

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ABSTRACT

Plastic debris were collected from eight beaches around San Diego County, California. Debris collected include: pre-production pellets and post-consumer plastics including fragments, polystyrene (PS) foam, and rubber. A total of $n = 2453$ pieces were collected ranging from <5 mm to 50 mm in size. The plastic pieces were separated by type, location, and appearance and analyzed for polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDT) and its breakdown products, and chlordanes. PAH concentrations ranged from 30 ng g^{-1} to 1900 ng g^{-1} , PCBs from non-detect to 47 ng g^{-1} , chlordanes from 1.8 ng g^{-1} to 60 ng g^{-1} , and DDTs from non-detect to 76 ng g^{-1} . Consistently higher PAH concentrations found in PS foam samples ($300\text{--}1900 \text{ ng g}^{-1}$) led us to examine unexposed PS foam packaging materials and PS virgin pellets. Unexposed PS foam contained higher concentrations of PAHs ($240\text{--}1700 \text{ ng g}^{-1}$) than PS virgin pellets ($12\text{--}15 \text{ ng g}^{-1}$), suggesting that PAHs may be produced during manufacturing. Temporal trends of debris were investigated at one site, Ocean Beach, where storm events and beach maintenance were found to be important variables influencing debris present at a given time.

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1. Introduction

Plastic is a synthetic organic polymer that is manufactured into a wide variety of low-cost, versatile, durable items ranging from consumer to industrial products (Laist, 1987; Hansen, 1990; Derraik, 2002; Ryan et al., 2009). By the end of 2010, the worldwide annual production of plastics was predicted to surpass 300 million tons (Halden, 2010). Today, improper disposal combined with increasing production has led to an increasing accumulation of plastic waste in the marine environment as a result of several sources including run-off, wind-blown litter, and shipping practices (Coe et al., 1997; Derraik, 2002; Ryan et al., 2009).

Plastics have been reported to make up 50–80% of shoreline debris (Barnes et al., 2009) commonly comprised of discarded fishing gear, pre-production pellets, scrubbers and fragments broken down from large debris (Carpenter and Smith, 1972; Colton et al., 1974; Moore et al., 2001; Derraik, 2002; Thompson et al., 2004; Corcoran et al., 2009). Large plastic debris is often removed during beach clean-ups, resulting in large quantities of microscopic and small plastic fragments remaining on beaches and subtidal sediments (McDermid and McMullen, 2004; Thompson et al., 2004;

Rios et al., 2007; Graham and Thompson, 2009). This paper focuses on small shoreline debris found on beaches across San Diego County, CA.

Recent studies have also raised concern over the ability of plastics to adsorb hydrophobic contaminants such as persistent organic pollutants (POPs) to their surfaces at concentrations one million times that in ambient seawater (Mato et al., 2001; Endo et al., 2005; Takada et al., 2006; Rios et al., 2007; Ogata et al., 2009; Teuten et al., 2009). Plastic debris is ingested by a wide range of animals due to mistaken identity or simply by indiscriminate ingestion (Browne et al., 2008; Graham and Thompson, 2009), and thus plastics may serve as a mechanism for POPs to enter the food web (Endo et al., 2005; Teuten et al., 2009).

This study aimed to (1) investigate the quantity and distribution of plastic marine debris present on San Diego County, CA beaches and (2) determine the distribution of hydrophobic contaminants adsorbed to the plastic marine debris across San Diego County. Four groups of contaminants were investigated: sixteen United States Environmental Protection Agency (EPA) priority polycyclic aromatic hydrocarbons (PAHs), seven abundant polychlorinated biphenyls (PCBs: 29, 52, 101, 118, 138, 153, and 180), dichlorodiphenyltrichloroethane and its breakdown products (*p,p'*-DDT, *p,p'*-DDD, and *p,p'*-DDE), and chlordanes (α -, and γ -chlordanes and *trans*-, and *cis*-nonachlor).

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2. Methods

2.1. Plastic debris sampling methods

Sampling locations were selected across San Diego County, California from north to south and included: Torrey Pines State Beach (TP), La Jolla Shores (LJS), Pacific Beach (PB), Mission Beach (MBe), Mission Bay (MBay), Ocean Beach (OB), Coronado Beach (CB), and Imperial Beach (IB; Fig. 1). At each location initial survey sampling was conducted whereby small plastic debris (<50 mm) were indiscriminately collected from the sand's surface with stainless steel tweezers, quantified, and stored in baked amber glass vials. To account for temporal trends of debris we chose one site, OB, to apply a standardized sampling method adapted from the National Oceanographic and Atmospheric Association's (NOAA) Marine Debris Program (Arthur et al., 2009). This site was chosen due to the relatively higher amount of plastic debris found during initial survey sampling. On 16 separate sampling days over a 9-week period all types of plastic debris were quantified using plastic sifters with <2 mm mesh size and 1 m² metal quadrats. The area of beach sampled was approximately 26 × 13 m separated into two regions, north and south. Quadrats were placed 2 m apart from a stationary reference point close to the water line and 5 m apart along the beach in both the northern and southern sampling area (Fig. 2). Stationary reference points were used in order to ensure frames were placed in the same location each sampling day. Plastic debris was separated from the sand's surface with tweezers, then the top one-inch of sand was collected, sieved and remaining debris was sampled (Fig. 2). Plastic debris was quantified and separated by color, size and type (pellets, fragments, PS foam, and rubber) and stored in a -20 °C freezer until chemical analysis.

2.2. Chemical analysis

Plastic samples were grouped into four types: pre-production pellets, plastic fragments, PS foam, and rubber. All plastics were

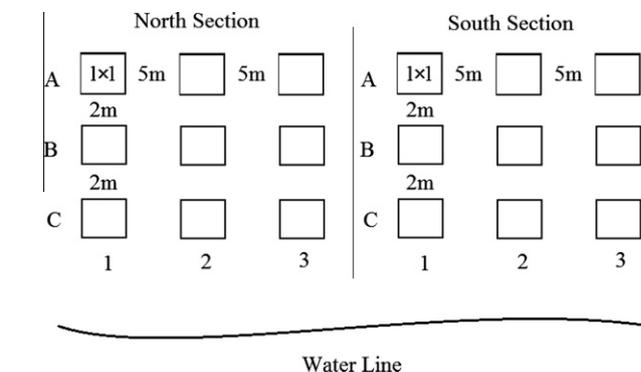


Fig. 2. Standardized sampling method conducted on Ocean Beach for plastic debris sample using 1 m² quadrats conducted in a 26 × 13 m area divided into 2 sections, North and South.

cleaned and extracted for determination of contents of PAHs, PCBs, DDTs, and chlordanes. Approximately one gram of pre-production pellets and varying weights of other types were pooled for extraction based on physical appearance. Pellets were pooled based on weathering, fouling, and yellowing; fragments and PS foam were pooled based on size and color. Additional samples of PS were analyzed to compare concentrations of PAHs between environmental samples and non-environmental samples and were prepared using the same methods as beach samples.

Information regarding all chemical standards, analytes, internal and recovery standards for analyses in this study are detailed in the Supporting Information (SI). Samples were extracted twice by sonication in 1:1 acetone:hexane solution and concentrated under nitrogen flow. For sample clean-up, we used solid phase extraction (SPE) with silica based cartridges eluted with 100% hexane and 4:1 dichloromethane:hexane. Final extract was spiked with known amounts of recovery standard prior to GC/MS analysis. Detailed extraction methods and instrumental parameters including QA/QC procedures for chemical analysis can be found in the SI.

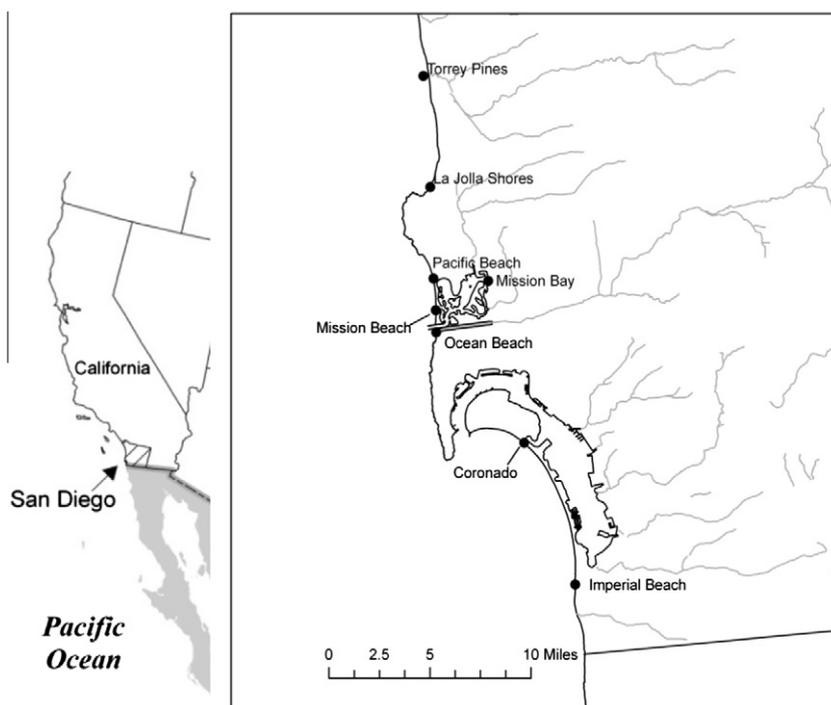


Fig. 1. Locations of sampling sites located in San Diego County. Figure generated utilizing ArcGIS version 9.3, Environmental Systems Research Institute, Inc. (ESRI) Redlands, California.

3. Results and discussion

3.1. Size distribution of small plastic debris collected

A total of $n = 2453$ individual plastic debris was collected during this study. $N = 1778$ pieces were prepared for chemical analysis. The size distributions of all plastics collected are summarized in Appendix Table B2 in the SI, 68% of which were <5 mm in size which is consistent with Browne et al., 2010 who found a greater abundance of microplastic compared to macroplastic along estuarine shorelines of the United Kingdom.

3.2. Standardized sampling and variables affecting the occurrence of plastic debris

All plastic was found on the surface of the sand with a greater abundance of debris observed in row A sampling areas. Fragments and pellets were consistently found in and around washed up algae and seagrass, suggesting that plastic is carried onto shore by natural debris. Initial survey sampling observations revealed that beaches with narrow sand areas and barriers (i.e., manmade walls) dividing beach area from streets contained less debris overall (LJS and TP). This suggests that high tide reaches these barriers and carries debris back into the marine environment. Beach grooming and storm events also influenced the presence of plastic debris. Sampling occurring days following a storm event revealed a large increase in the amount of plastic pellets collected (Fig. 3) consistent with findings by Lattin et al. (2004) and Storrier et al. (2007) who suggest that storm events can increase plastic debris discharged into the environment. The amount of algae present also increased, covering much of the beach. Shortly after storm events, beach grooming occurred resulting in removal of natural debris, which may have influenced fluctuations in the amount of plastic debris. Variability of debris present may also be affected by current patterns, wind direction, and amount of beach use (Storrier et al., 2007).

3.3. POPs in plastic marine debris

Samples from each site, grouped based on similarities, were combined to yield a total of $n = 39$ samples. Table 1 summarizes the total summed concentrations of sixteen PAHs, seven PCBs, DDTs, and chlordanes (see Table B1 in the SI for complete list of congeners) for each sample analyzed.

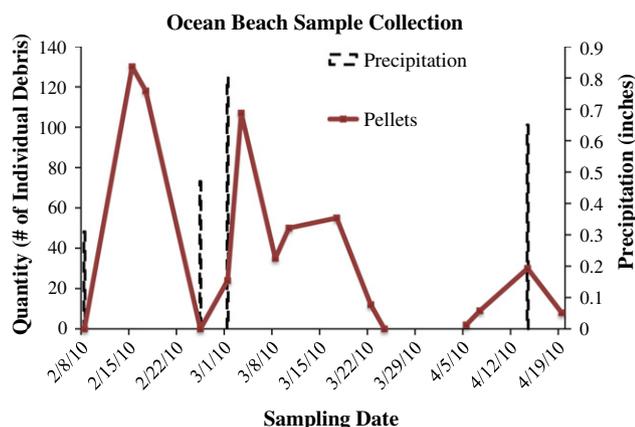


Fig. 3. Total amounts of plastic pellets found in area of 26×13 m² at Ocean Beach along the sampling dates. Missing data reflects missed sampling dates due to weather. Precipitation data was obtained from the National Weather Service. Source: National Atmospheric and Oceanic Administration (NOAA). National weather forecast office. Retrieved May 13, 2010, from <http://www.wrh.noaa.gov/sqx/hydro/qpe/qpe.php?wfo=sgx>.

PAH concentrations were greater than other contaminants across all plastic types. The median PAH concentrations were 92 ng g^{-1} for fragments, 42 ng g^{-1} for pellets, and 340 ng g^{-1} for PS foam. For all samples, PAH concentrations ranged from 18 ng g^{-1} to 1900 ng g^{-1} (Table 1). The greatest concentration (1900 ng g^{-1}) was found in PS foam collected at MBe. PAH concentrations were consistently high in all PS foams and relatively greater than in plastic fragments and pellets. But, other POP concentrations were not greater in PS. This result suggests that there may be additional sources of PAHs to PS foams than environmental exposure.

With the exception of two samples, PCBs, DDTs, and chlordanes were detected in all samples confirming that these POPs persist and accumulate in the environment despite their ban in the United States. Concentrations of PCBs, DDTs, and chlordanes were relatively similar regardless of plastic types ranging from 0.56 ng g^{-1} to 170 ng g^{-1} . Overall, yellowed or weathered pellets contained concentrations of POPs twice the concentration of pellets that were not yellowed, consistent with previous studies (Endo et al., 2005; Rios et al., 2007). Yellow or eroded pellets have increased surface area due to polymer weathering that increases the effective diffusivity (Mato et al., 2001; Endo et al., 2005; Karapanagioti and Klontza, 2008).

Spearman's rho nonparametric correlation was utilized to determine correlations among the subgroups of POPs (Table 2). There was no significant correlation between PAHs and the remaining POPs, but there were significant positive correlations among PCBs, DDTs, and chlordanes. This result suggests that sources and pathways of PAHs to plastics are different than the other POPs examined in this study. For example, contamination of PCBs, DDTs, and chlordanes on plastics may occur exclusively via exposure to the environment, but contamination of PAHs may also be a byproduct of the manufacturing process of PS foam.

3.4. PAH concentrations in non-environmental polystyrene and polystyrene foam

Due to the unexpected, yet consistently greater PAH concentrations in the PS foam samples, we hypothesized that non-littered PS foam might contain PAHs. To test the hypothesis, three packaging materials made with PS foam were chosen for PAH analysis. The samples were never exposed to the marine or coastal environment. The unexposed PS foam samples had comparable concentrations of PAHs as the PS foam samples collected from the beaches ($240\text{--}1700 \text{ ng g}^{-1}$ vs. $300\text{--}1900 \text{ ng g}^{-1}$ respectively). Additionally, we analyzed virgin pre-production PS pellets to speculate whether these PAH concentrations could be a product of the manufacturing of PS to PS foam. Three PS pellet samples, two grams each, had total PAH concentrations at $12\text{--}15 \text{ ng g}^{-1}$. These PS pellets were obtained directly from industry and were never exposed to the environment. Prior to analysis, the PS pellets were cleaned by pure water and air dried.

Likewise, the unexposed PS foam samples had PAH concentrations one to two orders of magnitude greater than the virgin pre-production PS pellets. This result strengthens our argument that the manufacturing of PS to PS foam produces PAHs. PAH formation during PS foam production has not been reported in the literature. However, PAH formation was reported during incineration of polystyrene (Hawley-Fedder et al., 1984). A common method to produce PS foam (also called expanded polystyrene) includes heating; a volatile blowing agent is impregnated into polystyrene beads or pellets through the use of temperature and applied pressure (US EPA, 1993). Therefore, in this heating process PAH formation may occur. In addition, PAH concentrations in the virgin pre-production PS pellets ($12\text{--}15 \text{ ng g}^{-1}$) were greater than those in other types of pre-production pellets (HDPE: $3\text{--}6 \text{ ng g}^{-1}$, PP: 2--

Table 1
Summary of location, color, size, quantity, mass, and concentrations of PAHs, PCBs, chlordanes, and DDTs with samples organized by plastic type, sampling location, and chronologic order based on extraction date. N.D. indicates non-detectable concentrations and N.A. indicates data that is not available.

Type	Location	Color	Size (mm)	Count	Mass (g)	PAHs (ng g ⁻¹)	PCBs (ng g ⁻¹)	Chlordanes (ng g ⁻¹)	DDTs (ng g ⁻¹)	
Fragments	Coronado	Mix	<5 to 30	13	3.28	37	4.5	2.1	3	
	Imperial Beach	White	N.A.	N.A.	1.24	100	16	12	37	
		White	<5 to 15	51	1.4	250	10	4.4	33	
		White	<5 to 15	80	1.56	180	47	9.6	8	
	Mission Bay	Mix	30 to <35	4	1.63	93	2.5	5.2	17	
		Mix	10 to <30	14	0.55	90	3.5	8	2	
	Mission Beach	Mix	30 to <35	6	2.62	49	3.2	4.2	2	
		Mix	10 to <30	8	0.94	64	5.8	5.2	8	
	Ocean Beach	Mix	<5 to <25	N.A.	2.72	77	3.1	2	N.D.	
		White	10 to <30	18	1.2	1200	9.8	2.6	2	
		Mix	5 to <25	9	1.01	170	9.9	5.9	5	
		Mix	N.A.	17	1.88	140	12	15	72	
		White	<5 to <20	52	1.61	37	5.5	4.5	7	
		Mix	15 to <30	10	1.62	27	24	8.7	12	
		White	15 to <35	14	0.8	110	5.3	6.9	3	
	Torrey Pines	Mix	15 to <35	9	1.76	61	13	21	10	
	Median			14	2	92	8	6	8	
	Range			4–52	.55–3.28	27–1200	2.5–47	1.90–21	1.5–72	
	Pellets	Imperial Beach	Yellowed	<5	66	1.03	130	9.6	32	9
			Yellowed	<5	75	1.4	68	15	27	41
Yellowed			<5	54	1	210	42	17	8	
La Jolla Shores		Yellowed	<5	55	1.01	42	4.9	12	3	
		Yellowed	<5	43	1.01	18	5.3	9	3	
		Yellowed	<5	40	1.01	55	4	36	7	
		Yellowed	<5	54	1.38	52	10	21	6	
		Yellowed	<5	N.A.	1.06	26	3.8	1.8	1	
Ocean Beach		Yellowed	<5	40	1.01	28	9.3	6.2	2	
		Yellowed	<5	44	1.01	120	6.3	7.8	10	
		Yellowed	<5	44	1	35	8.9	11	4	
		Yellowed	<5	50	1.01	22	12	17	10	
		Yellowed	<5	40	1	36	9.2	20	29	
		Yellowed	<5	47	1	18	8.5	5.1	2	
		Yellowed	<5	42	1.02	39	4.2	4.5	49	
		Yellowed	<5	42	1.01	38	5.9	10	3	
		Yellowed	<5	48	1	120	37	34	26	
		Yellowed	<5	N.A.	1.02	85	32	170	64	
		Yellowed	<5	43	1.01	48	19	15	16	
		Median			44	1	42	9.2	15	8.4
		Range			40–75	1.00–1.40	18–210	3.8–42	1.8–170	0.56–64
		Polystyrene Foam	Coronado	Mix	25 to <35	5	0.65	300	N.D.	20
La Jolla Shores			Mix	15 to <30	N.A.	0.23	330	9.4	4.7	7.5
Mission Beach	Mix		<5 to <25	8	0.44	350	6.6	6.8	75	
	Mix		15 to <30	7	0.43	1900	14	60	36	
Median				7	0.44	340	9.4	13	21.85	
Range			5–8	0.23–0.65	300–1900	6.6–14	4.7–60	7.5 to 75		
Polystyrene Foam Packaging		Soft White			0.11	1100				
		Hard White			0.11	240				
		Soft Yellow			0.15	1700				
Median				0.11	1100					
Range				0.11–0.15	240–1700					
Polystyrene Pellets	Blanks	Sample 1		2	12					
		Sample 2		2	15					
		Sample 3		2	14					
	Median			2	14					
	Range			2	12–15					

5 ng g⁻¹). This may be related to the aromaticity of PS due to the use of a styrene monomer. Further research is necessary for investigation of the relationship between PAH formation and PS and PS foam.

3.5. Mass, quantity, and POP concentration comparisons

Pellets made up the majority of analyzed samples. We analyzed approximately 827 pellets, 305 fragments, and 20 PS foam samples

(Fig. 4). Fragments held the greatest overall mass at 26 g, followed by pellets at 20 g, and PS foam at 1.8 g. This discrepancy between quantity and mass is most likely due to the uniform size of pellets <5 mm while fragments ranged from <5 mm to 35 mm in size. These trends, however, are not consistent with the distribution of POP concentrations (Fig. 4). The most striking observation is that PS foam contributed the greatest concentration of total PAHs although its contribution in mass and quantity were minimal compared to that of pellets and fragments. PS foam also contributed

Table 2Spearman's *Rho* Correlation between PAHs, PCBs, Chlordanes, and DDTs. N represents sample size.

Correlations			PAHs	PCBs	Chlordanes	DDTs
Spearman's <i>Rho</i>	PAH	Correlation coefficient	1.000	.006	-.002	.145
		N	39	38	39	38
	PCB	Correlation Coefficient	.006	1.000	.589**	.523**
		N	38	38	38	37
	Chlordane	Correlation coefficient	-.002	.589**	1.000	.436**
		N	39	38	39	38
	DDT	Correlation coefficient	.145	.523**	.436**	1.000
		N	38	37	38	38

** Correlation is significant ($p < 0.05$).

largely to total chlordanes and DDTs compared to its contribution in total mass. This result suggests that environmental loadings of these pollutants cannot be estimated only from the mass and quantity of plastic debris in the marine environment without the concentrations.

3.6. Contaminant differences across San Diego beaches

To reduce variability, only plastic pellets were used to compare differences in contaminant concentrations across beaches. Due to the limited and variable number of samples, statistical tests were not appropriate and only trends were considered. Concentrations of each group of POPs are described within a dot plot in logarithm scale by subgroups of the three locations: IB, OB, and LJS (Appendix Fig. C2 in the SI). The majority of plastic pellet samples were collected from OB (12 out of 19 samples) and had variable concentrations of POPs. Three pellet samples from OB contained high concentrations of PAHs and chlordanes. However, this was not observed for DDTs and PCBs. There were also no noticeable trends for the samples from other sites. Samples collected from IB revealed the greatest concentrations of POPs overall. This may be attributed to its close proximity to the Tijuana River, with a watershed of 1731 km² (Nordby and Zedler, 1991), that carries waste from the Tijuana region into San Diego County and flows out to the Pacific Ocean via Imperial Beach (Fairey et al., 1998). Further studies of plastic marine debris in IB are recommended.

3.7. Regional POP concentration comparisons from recent literature

There are several recent global studies regarding POP concentrations found on plastic debris (Mato et al., 2001; Endo et al., 2005; Takada et al., 2006; Rios et al., 2007; Ogata et al., 2009). The previous studies examined primarily plastic pellets. The levels of PAHs and PCBs in the pellets in this study are comparable to previous studies (Table 3). The pellet concentrations of DDE in San Diego beaches are similar to those found on beaches in Japan (Mato

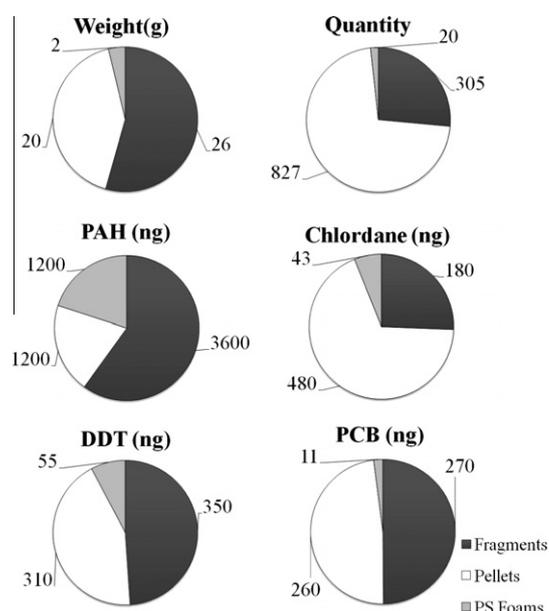


Fig. 4. Proportion of plastic make up of this study by quantity and mass and proportion totals of POP concentrations found in extracted samples by type. In the pie charts, white, light gray, and dark gray colors indicate pellets, polystyrene foams, and fragments.

et al., 2001) but less than those reported by Rios et al. (2007) in the North Pacific Gyre, Hawaii, California, and the Guadalupe Islands in Mexico.

In a study conducted by the International Pellet Watch, plastic pellets were examined from 30 beaches from 17 countries (Ogata et al., 2009). Results indicated greater concentrations of PCBs from San Francisco (SF), Los Angeles (LA), and Boston ranging from 300 ng g⁻¹ to 600 ng g⁻¹. SF and LA are located on the California

Table 3

Comparisons of, PAH, PCB, and DDE concentrations found in pellets from previous studies.

Pellets study	Sampling location	PAH range (ng g ⁻¹)	PCB range (ng g ⁻¹)	DDE range (ng g ⁻¹)
This study	8 beaches in San Diego, CA, USA	18–210	3.8–42	0.33–8.2
Mato et al., 2001	4 Locations: Kasi Seaside Park, Keihin Canal, Tokyo Bay, Shioda Beach, Japan		3.97–117	0.16 to 3.1
Endo et al., 2005	47 Beaches, Japan		<28–2300	
Takada et al., 2006	Beaches, Japan		5–892	
Rios et al. (2007)	10 Locations: North Pacific Gyre, selected sites from California, Hawaii, Guadalupe Island, Mexico	39–1200	27–980	22 to 5600
Ogata et al., 2009	8 locations in California		32–605	.65–128
	Tokyo Bay, Sgami bay, and Osaka Bay		169–453	1.35

coast about 502 and 102 miles respectively from San Diego County. The greatest concentration of DDT was also found in SF and LA as well as Vietnam ranging from 110 ng g⁻¹ to 300 ng g⁻¹. Concentrations found in our study were found to be dramatically smaller with concentrations of DDT found in pellets ranging from 0.56 ng g⁻¹ to 64 ng g⁻¹ and 3.8 ng g⁻¹ to 42 ng g⁻¹ for PCBs. Ogata et al. (2009) found that POP concentrations on plastic pellets were consistent with local mussel monitoring programs, suggesting that data regarding POP concentrations on plastic pellets could serve as sentinel or a first screening for global pollution.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2011.09.039.

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