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Organic micropollutants in marine plastics debris from the open ocean and remote and urban beaches

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ABSTRACT

To understand the spatial variation in concentrations and compositions of organic micropollutants in marine plastic debris and their sources, we analyzed plastic fragments (∼10 mm) from the open ocean and from remote and urban beaches. Polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), dichloro-diphenyl-trichloroethane and its metabolites (DDTs), polybrominated diphenyl ethers (PBDEs), alkylphenols and bisphenol A were detected in the fragments at concentrations from 1 to 10,000 ng/g. Concentrations showed large piece-to-piece variability. Hydrophobic organic compounds such as PCBs and PAHs were sorbed from seawater to the plastic fragments. PCBs are most probably derived from legacy pollution. PAHs showed a petrogenic signature, suggesting the sorption of PAHs from oil slicks. Nonylphenol, bisphenol A, and PBDEs came mainly from additives and were detected at high concentrations in some fragments both from remote and urban beaches and the open ocean.

1. Introduction

The global production of plastic has increased from 1.5 million tonnes in 1950 to 230 million tonnes in 2009, with an average annual growth of around 9% (PlasticsEurope, 2010). Owing to poor management of plastic wastes on land, some plastic wastes reach the sea via various routes, including rivers and urban runoff. The land-based inputs of plastics into the ocean are thought to be increasing (Law et al., 2010). Plastics are also dumped directly to the ocean through fishery and maritime activities. Heavier plastics (e.g., polyvinyl chloride) are accumulated in bottom sediments. Lighter plastics (e.g., polyethylene [PE], polypropylene [PP]) are distributed on the sea surface, although their densities can increase through biofouling (Moret-Ferguson et al., 2010). Some of the light plastics are stranded on beaches and some are accumulated in specific areas in the ocean, such as the North Atlantic Gyre (Moret-Ferguson et al., 2010) and the North Pacific Central Gyre (Moore et al., 2001). Others are physically degraded to microscopic pieces (Thompson et al., 2004; Browne et al., 2008).

The ingestion of plastics by marine organisms has been reported (Gregory, 2009). Recently toxic organic contaminants such as polychlorinated biphenyls (PCBs) and nonylphenols have been detected in marine plastics (Mato et al., 2001). Ingestion of the plastics could be a significant exposure route of the toxicants to marine organisms and their adverse effects on the organisms and marine ecosystems are of concern. A variety of hydrophobic organic compounds (HOCs) were detected in plastics ingested by seabirds (Colabuono et al., 2010). Concentrations and the spatial variations are essential information for the risk assessment of the chemicals associated with marine plastics. Marine plastics contain two types of chemicals: additive-derived chemicals and hydrophobic chemicals sorbed from surrounding seawater (Teuten et al., 2009). We have been globally monitoring HOCs by using beached plastic resin pellets since 2006 (Takada, 2006). The monitoring (International Pellet Watch) revealed concentration ranges and spatial variations of the HOCs such as PCBs and organochlorine pesticides in resin pellets collected from beaches all over the world (Ogata et al., 2009). Resin pellets are significant, but minor components among the marine plastics compared with more abundant fragments of plastic products in marine environments (Law et al., 2010; Moret-Ferguson et al., 2010) and biota (Ryan et al., 2009). However, only limited information is available on the concentra-

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tions of organic micropollutants in marine plastic fragments and their spatial variations. Rios et al. (2007, 2010) demonstrated concentrations of HOCs, including PCBs, from the Central Pacific Gyre and the California coast. In addition to HOCs, plastic fragments may also contain additive-derived chemicals. Some of the chemicals (e.g., nonylphenol) have estrogenic activity and may disrupt the endocrine system when they are ingested by marine biota. In addition to additives, chemicals may accumulate in marine plastics from seawater by sorption. Thus, it is important to distinguish among the multiple sources (e.g., sorption or additives) of the chemicals so as to reduce their potential risk.

Our previous paper (Teuten et al., 2009) presented preliminary data on the concentrations of organic micropollutants in two PE fragment samples. The data were not enough to allow us to discuss the spatial variation of organic micropollutants in marine plastics and their sources. So here we expanded the locations to eight areas: three in the open ocean (Central Pacific Gyre, Pacific Ocean, Caribbean Sea), two remote beaches (Costa Rica, northern Vietnam), and three urban beaches (Tokyo, Kanagawa, Los Angeles). Furthermore, we analyzed PP fragments too. The results allow us to discuss the spatial variation in organic micropollutants in marine plastics and their sources, especially multiple source terms (sorption and additives).

2. Samples

Plastic fragments were collected from open ocean and beaches in remote areas and urbanized areas. The locations and areas are shown in Fig. 1 and the geographic coordinates and details are listed in Table S1. Samples from central pacific gyre (CGPE1–3, CGPE6, CGPP9, CGPP13) were collected in July and August in 2005 on the cruise of ORV *Alguita* of Algalita Marine Research Foundation (AMRF). Further open ocean samples were collected during the cruises of SSV *Robert C. Seamans* and SSV *Corwith Cramer* (both vessels of the Sea Education Association) in the Pacific Ocean (S-217PP) and Caribbean Sea (C-216PE and C-216PP). The detailed information on the cruises is available in supporting information (Table S1). Floating samples were collected using a neuston net. The plastics were sorted out on board and placed in glass bottles or stainless steel containers. The bottles and containers were sealed to minimize the sorption of HOCs from the ambient air and stored in a freezer or at room temperature until analysis. Stranded plastic fragments and resin pellets were collected from remote beaches at Marbella Beach (Costa Rica; CRPE1–5 and CRPP1–5) and Thinh Long, Tonking Bay (Vietnam; TKPE1–2 and TKPP1–3) and urban beaches at Odaiba (Tokyo; ODPE0–3 and ODPP0–2), Kugenuma (Kanagawa; KNPE2–6 and KNPP1–3) and Seal Beach (USA; SBPE1–2 and SBPP1–3). The plastic samples were picked up from stranded materials along the high tide line of the beaches by using solvent-rinsed tweezers. The samples were wrapped in aluminum foil and placed in zip-lock bags. They were sent to the laboratory in Tokyo at room temperature and then stored in a freezer until analysis. No target compounds leached from the zip-lock bags.

3. Methods and materials

Plastic fragment samples were sorted by near-infrared spectrometer (PlaScan-W OPT Research Inc., Tokyo, Japan) to select polyethylene (PE) and polypropylene (PP) fragments. Any wet samples were first dried in a glass desiccator with silica gel before sorting. PE and PP fragments varying in size from a few mm to a few cm were analyzed. The first two letters of sample codes (e.g., CG) indicate the specific locations (e.g., Pacific Central Gyre) and the following two letters indicate the polymer types (i.e., PE and PP). Photos of all the plastic fragment samples are shown in Figure S1. Between three and five single plastic fragments were analyzed for individual locations to determine piece-to-piece variation. However, for some samples (i.e., KEPE3, KEPE4, KEPE6), several fragments were combined and analyzed together due to smaller fragment sizes. These fragments appeared to have the same color, type, and product. ODPE00, ODPP00, and SBPP3 were composite samples of plastic fragments with different colors and shapes. For plastic fragments from the Central Pacific Gyre, plastic fragments of the same appearance were combined and analyzed together. Samples from SEA cruises (i.e., S-217PP, C-216PE, C-216PP) were composites of fragments of different color and most likely different products.

Plastic fragment samples were Soxhlet-extracted with 150 mL of dichloromethane (DCM) at a rate of 3–4 cycles per hour for 18 h. The extracts were spiked with isotopically-labeled surrogate standards for PCBs, PAHs, PBDEs, nonylphenol (NP), octylphenol (OP), bisphenol A (BPA), as listed in Table S2, and then rotary-evaporated just to dryness. The residue was dissolved in 1 mL of 25% hexane/75% DCM (v/v) and transferred onto a 5%-H_2O-deactivated silica gel column (1 cm i.d. × 9 cm). Five fractions (I, II, II, IV, and V) were obtained by successive elution of 20 mL of 25% hexane/75% DCM, 20 mL of 40% hexane/60% DCM, 30 mL of 60% hexane/40% DCM, 30 mL of DCM, and 45 mL of 50% DCM/50% acetone, respectively. Eluent in Fraction I was roto-evaporated just to dryness, redissolved into 0.4 mL of hexane, and transferred onto fully activated silica gel column (0.45 cm i.d. × 18 cm). Fraction Ia containing alkanes and Fraction Ib containing PCBs, DDTs, PBDEs, and PAHs were eluted with 4 mL of hexane and 10 mL of 25% hexane/75% DCM, successively. Eluent in Fraction Ib was rotary-evaporated to ~0.5 mL and transferred to a 1.5-mL glass vial. The solvent in the vial was evaporated just to dryness under gentle nitrogen stream and the residue was re-dissolved into appropriate volume (50–1000 μL) of *iso*-octane containing internal injection standards (IIS) listed in Table S2. One microlitre aliquots were analyzed by gas chromatography. PCBs, DDTs, and PBDEs were determined by gas chromatography-ion-trap mass spectrometers (GC-ITMS; Thermo Fisher ITS) on MS/MS mode. PAHs were analyzed by GC interfaced with a quadrupole MS (GC–MS; Agilent 5973 MSD with

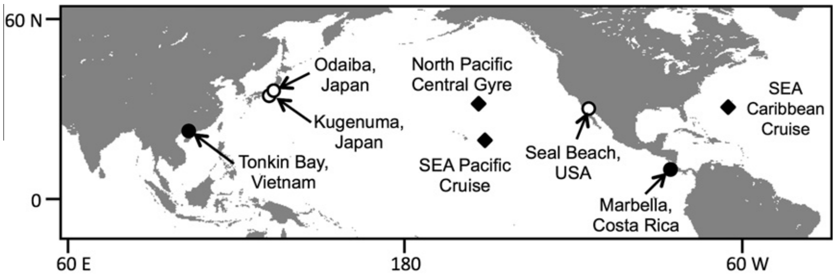

Fig. 1. Sampling locations. Closed diamond: open ocean sample; closed circle: remote beach sample; open circle: urban beach sample.

6890 GC). The volume of solution IIS was adjusted so that the concentrations of the target compounds were within the range of the calibration lines drawn from the results of injecting a range of standard solutions. Detailed operation conditions are listed in Table S3.

Fractions III and V from the 5% deactivated silica gel elution were reduced in volume to ~0.5 mL by rotary evaporator and transferred to 1.5 mL clear vials, respectively. The solvent in the vials were evaporated just to dryness under gentle nitrogen stream. To the vial 50 μL each of pyridine and acetic anhydride was added and mixed. After keeping the mixture at room temperature overnight, the reaction was stopped with addition of 200 μL of 4 N HCl. Acetylates, including acrylates of the target analytes alkylphenols and bisphenol A, were extracted with *n*-hexane. The hexane extracts were transferred to 1.5 mL clear vials. Hexane in the vials was evaporated just to dryness under gentle nitrogen stream. The acetylates were re-dissolved into an appropriate volume (50–1000 μL) isooctane containing IIS which is listed in Table S2. One-microliter aliquots were analyzed by GC–MS (Agilent 5973 MSD with 6890 GC) operated in SIM mode. On every sample run, a range of standard solution concentrations was derivatized to draw calibration lines for quantification. Detailed operation conditions are listed in Table S3.

The reproducibility of the analyses was confirmed by four replicate analyses of aliquots of extracts of plastic fragments from Kugenuma beach. The relative standard deviations of the concentrations of the all the target compounds were less than 20%. Recovery was tested by spiking with native standards of target compounds (PCBs, DDTs, PBDEs, PAHs, NP, BPA) to the aliquots of the fragment extracts. The recoveries of all the target compounds were more than 74% except for OP (23–41%). Each sample set normally contained 5 samples and a procedural blank. As the blank, a baked clean glass thimble was Soxhlet-extracted with the same amounts of solvent, and the extract was subjected to the same procedure as the sample extract. The amounts detected in the procedural blank were subtracted from the detected amounts in the samples in the same set. Normally procedural blank was as follows. $\sum$PCBs and $\sum$DDTs: ~1 ng/g; $\sum$PBDEs: ~0.3 ng/g; BDE#209: ~0.1 ng/g; $\sum$PAHs: 100 ng/g; NP and OP: ~1 ng/g, BPA: ~3 ng/g.

4. Results and discussion

4.1. Validity of DCM extraction

In the present study, moderately polar organic solvent, i.e., DCM, was used to extract various compounds with wide range of polarities from PCBs to BPA. Non-polar solvents such as hexane might be efficient to extract apolar compounds such as PCBs. To confirm the efficiency of DCM extraction, sequential extractions were made for several plastic samples. The plastic samples were extracted with DCM and the extracted plastic fragments were further Soxhlet-extracted with hexane. As shown in Table 1, less than 10% of CB congeners were detected in the hexane extracts, indicating that the DCM extraction is more ideal for PCBs. For the other target compounds, minor proportions (<25%) of the compounds were extracted with hexane, except for BDE209. The amount of BDE209 was six times greater when extracted with hexane than with DCM. This indicates that BDE209 concentrations reported in the present study are most likely underestimates. Regarding polar solvent, our previous study demonstrated that methanol (MeOH) was not efficient for the extraction of PCBs, DDE and nonylphenol from marine polypropylene pellets (Mato et al., 2001). For one polyethylene sample, MeOH extraction was applied prior to DCM extraction. No significant amount of target compounds were detected in the MeOH extracts. Based on these results, DCM was selected in the present study as appropriate solvent to extract wide range of chemicals in marine plastic.

4.2. Concentrations and sources of organic micropollutants in marine plastic fragments

Analytical data on all the target compounds for polyethylene and polypropylene fragments are shown in Fig. 2 and Table 2. PCBs were detected in all the plastic fragments with wide range of concentrations from 1 to 436 ng/g (Table S4). This is consistent with studies that have shown that PCBs are sorbed to plastics. For example, the sorption of PCBs to plastics was demonstrated by laboratory (Adams et al., 2007) and *in situ* (Mato et al., 2001) adsorption experiments. For individual locations, there was an observed tendency that PE fragments had higher concentrations of PCBs than PP fragments. Among samples collected from a Japanese beach, PE pellets had higher concentrations of PCBs than PP pellets (Endo et al., 2005), on account of the more hydrophobic nature of PE than PP. An *in situ* adsorption experiment demonstrated greater sorption of PCBs to PE pellets than PP pellets (Mato et al., 2002). Laboratory adsorption experiments showed higher distribution coefficients of PAHs for PE than for PP (Karapanagioti and Klontza, 2007, 2008; Teuten et al., 2007; Karapanagioti et al., 2010). Large piece-to-piece variations (up to 2 orders of magnitude) in PCB concentration were observed for individual locations and for each polymer type (i.e., among PP and among PE). Large variations were observed also for pellets and ascribed to the combination of slow sorption, different residence times in water, and different pathways among plastics (Endo et al., 2005). Fragments are more heterogeneous in terms of shape and weathering than the pellets and these could also contribute to the large variation in PCB concentration. Different residence times and pathways of movement through the environment for both plastics and PCBs are consistent with variation in PCB compositions as shown in Fig. 3.

PCBs concentrations in plastic fragments were higher in urban beaches than those in remote beaches and the open ocean (Fig. 2). This could be due to different magnitudes of PCB inputs, i.e., industrialized countries used larger amounts of PCBs resulting in larger releases of PCBs to the coastal environments during periods of their economic growth in 1960s and early 1970s. Because of the persistent and hydrophobic nature of PCBs they are legacy pollutants in coastal sediments. They can become resuspended and desorbed to contaminate overlying waters and the water column. Higher concentrations of PCBs in the plastic fragments most likely reflect the legacy of PCB discharge in the USA and Japan. PCB concentrations in the fragments from the Pacific Central Gyre (1–78 ng/g) were similar to those reported by Rios et al. (2010). Penta- to hepta-chlorinated congeners were predominant in the beached fragment samples from Kugenuma, Seal Beach, Tonking Bay and Costa Rica, while tetra-chlorinated congeners had greater relative abundance in Odaiba Beach (Fig. 3). In the open ocean, fragment samples had more tri and tetra chlorinated congeners relative to the higher chlorinated congeners. This latter congener pattern is similar to those observed for water samples in the open ocean (Sobek and Gustafsson, 2004; Gioia et al., 2008). PCBs are atmospherically transported to open ocean and, therefore, volatile (lower chlorinated) congeners are selectively transported to the open ocean (Gioia et al., 2008). Rios et al. (2010) reported the predominance of lower-chlorinated congeners in the plastic fragment samples from the Central Pacific Gyre. They estimated that lower chlorinated congeners were atmospherically transported. In spite of this general pattern in the congener relative abundance profiles in open ocean samples, large fragment-to-fragment differences were observed in the concentrations. In some oceanic samples (e.g., CGPE3), middle chlorinate congeners (i.e., pnet- and hexa-chlorinated) were dominant as is frequently observed in urban

Table 1
Efficiency of DCM extraction of marine plastics for organic micropollutants.

% Extraction with hexane after DCM extraction*									
	PCB	DDTs	PBDEs		PAHs		AP		BPA
	CB#138	DDD	BDE#47	BDE#209	Phe	Chry	NP	OP	BPA
Polyethylenen									
KNPE**	<0.6	n.a.	7.1	n.a.	6.9	6.2	0.7	0.6	n.a.
Polypropylene									
KNPP1	1.6	24	2.9	n.a.	<30	<19	4.2	2.2	35
KNPP2	<0.9	8.4	3.3	n.a.	<19	<13	0.4	13	n.a.
KNPP3	4.6	<27	3.3	n.a.	n.a.	n.a.	20	n.a.	n.a.
CGPP9	<10	<20	0.3	n.a.	n.a.	n.a.	<5.0	<50	n.a.
CGPP13	9.3	<23	0.0	600	n.a.	n.a.	<4.2	<24	n.a.

*% Of amounts of target compound extracted with hexane to sum of extracted with DCM.
** Amounts of target compounds in DCM extraction are sum of Soaking in DCM in 72 h and Soxhlet extraction.

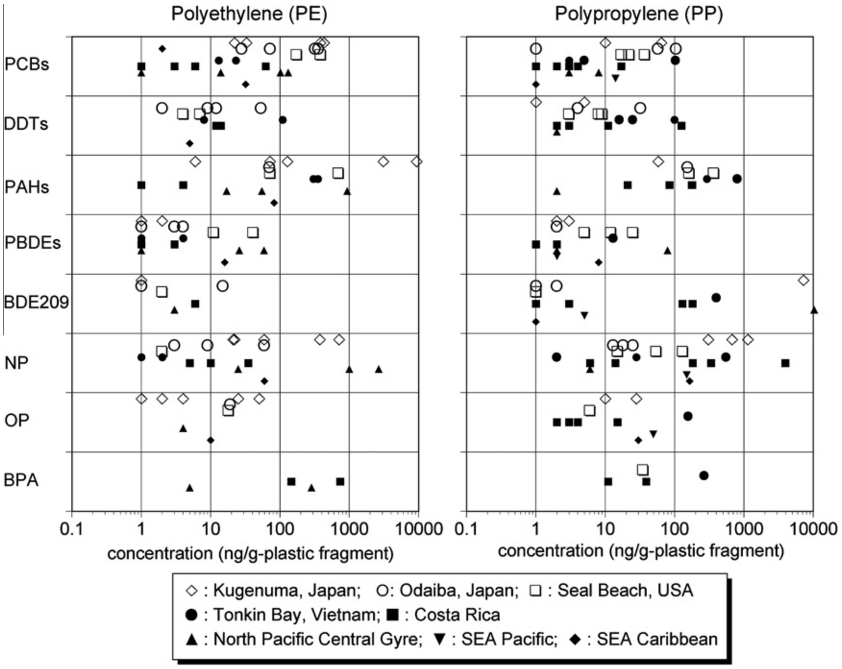

Fig. 2. Concentrations of organic micropollutants in marine plastic fragments.

coastal samples. This suggests that marine fragments can act as vectors of PCBs to open ocean areas.

PCBs were measured also in pellet samples collected from the same beaches. For the pellet samples, median concentrations indicate the pollution status of the coastal waters (Ogata et al., 2009). Median of PCB concentrations in the pellets showed good correlation with median concentrations of PCBs in plastic fragments (Fig. 4). This correlation indicates that PCB concentrations in the fragments can be estimated from the results of International Pellet Watch.

PAHs were also detected in all the plastic fragments. Fragment-to-fragment variability was observed for PAHs concentrations. PAHs concentrations ranged from 1 to 9300 ng/g. Again, fragments from urban beaches had higher PAHs concentrations, probably due to the proximity to anthropogenic activities. Anthropogenic sources of PAHs are categorized into two groups, i.e., petrogenic and pyrogenic (Lima et al., 2005). The two sources can be distinguished by molecular profiles. Petrogenic PAHs are often rich in lower molecular weight compounds such as naphthalene and alkyl naphthalenes, phenanthrenes and alkyl phenanthrenes, whereas pyrogenic PAHs are rich in higher molecular weight compounds. Figure S2 shows the molecular profiles of PAHs in the plastic fragments. As with the PCBs, there was fragment-to-fragment variability in molecular profiles among the same polymer types in the same locations. Generally PAHs in the fragments were of lower molecular weight, suggesting petrogenic sources of PAHs in the plastic fragments. The ratio of higher-molecular-weight PAHs (benzo[a]anthracene – coronene) to lower-molecular-weight PAHs (phenanthrene – pyrene), H/L ratio, was 0.32 ± 0.49 ($n = 40$; Table 2) in the fragments. These ratios are more indicative of petrogenic PAHs than pyrogenic PAHs. Crude oils ($n = 7$) had an H/L of 0.19 ± 0.07, whereas wood combustion products and coal combustion products had an H/L of 2.17 ± 1.34 and 6.99 ± 9.92, respectively, in Saha (2008). Also H/L ratios in the plastic fragments are lower than those observed in coastal sediments, e.g., Tokyo Bay sediment = 0.91 ± 0.37 ($n = 24$), and Vietnamese rural sediments = 0.87 ± 0.15 ($n = 6$), both indicative of pyrogenic sources (Saha et al., 2009). It is most plausible that plastic debris on the sea surface have much more opportunity to be exposed to oil slicks or floating tar particles from oil cargo tanker ballast washings or natural oil seeps. A petrogenic signature for the plastic debris was confirmed by the abundance of alkylated PAHs, which is

Table 2
Concentrations and compositional parameters of organic micropollutants in marine plastic fragments.

	PCBs	PAHs				PBDEs			DDTs					Alkylphenols			BPA
	∑PCBs (ng/g)	∑PAHs (ng/g)	MP/P	Pyr/Fluo	H/L	BDE-209 (ng/g)	∑PBDEs (ng/g)	%BDE-209	DDE (ng/g)	DDD (ng/g)	DDT (ng/g)	∑DDTs (ng/g)	DDT/∑DDTs	OP (ng/g)	Total NP (ng/g)	NP/OP	BPA (ng/g)
KNPE2	436	3028	2.39	0.71	0.13	0.0	0.8	0	138	n.d.	n.d.	138	n.a.	25.1	372	14.8	0.2
KNPE3	379	9297	2.66	0.94	0.15	0.0	1.6	1	198	n.d.	n.d.	198	n.a.	49.2	706	14.3	0.5
KNPE4	22	69	0.90	0.21	0.20	0.5	1.7	31	1.4	n.d.	n.d.	1.4	n.a.	0.5	17.8	33.0	0.5
KNPE5	2	0	n.a.	n.a.	n.a.	0.0	0.02	0	0.2	n.d.	n.d.	0.2	n.a.	0.0	1.3	>1.3	0.0
KNPE6	22	17	1.63	n.a.	0.01	0.1	1.6	7	0.5	n.d.	n.d.	0.5	n.a.	1.8	55	30.0	0.2
KNPP1	37	151	0.71	0.63	0.25	228	230	99	0.6	0.7	0.1	1.4	0.08	27.6	670	24.3	3.4
KNPP2	6	172	0.49	0.75	0.48	0.0	3.9	0	1.5	0.7	5.5	7.6	0.72	8.0	1244	156	0.0
KNPP3	5	137	0.63	0.68	0.10	135	136	99	0.2	0.4	0.4	1.0	0.43	0.8	305	378	0.0
ODPE00	159	269	1.74	2.01	0.64	0.7	4.2	16	22	3	27	52	0.51	0.0	4.4	>4.4	0.0
ODPE01	285	521	0.43	1.06	0.11	11.9	15.5	76	16	2	9	27	0.33	2.7	13.4	5.0	0.0
ODPE02	26	83	0.44	0.83	0.08	0.7	1.1	70	0.7	0.4	1.4	2.4	0.57	1.8	3.7	2.1	1.4
ODPE03	66	58	0.61	1.06	0.17	0.5	2.0	26	0.9	0.5	8.1	9.4	0.85	17.4	0.3	0.01	0.0
ODPP00	43	106	1.53	0.76	0.66	0.9	1.7	54	5.9	3.8	24	33	0.71	0.2	22.4	117	0.0
ODPP01	71	270	1.62	0.82	0.01	0.9	2.7	33	2.0	1.1	3.1	6.2	0.50	0.5	10.7	20.9	0.0
ODPP02	2	261	0.57	0.97	0.05	1.3	1.4	95	0.0	0.0	0.2	0.2	0.86	2.6	8.5	3.3	2.8
SBPE1	399	656	0.95	0.59	0.16	1.5	12.6	12	2.4	0.2	1.6	4.1	0.38	16.8	0.0	0.0	6.8
SBPE2	178	79	0.50	0.71	0.51	0.2	41.4	1	2.1	1.3	3.5	7.0	0.51	0.0	1.1	>1.1	0.0
SBPP1	20	378	1.18	0.56	0.50	0.2	12.6	1	7.6	0.2	0.7	8.4	0.08	0.7	7.2	10.3	0.0
SBPP2	15	39	0.32	0.92	0.28	0.1	5.3	3	1.2	0.2	0.8	2.2	0.36	0.1	48.3	461	26.2
SBPP3	119	144	1.37	1.63	0.35	0.6	25.5	2	5.8	1.1	1.3	8.2	0.16	4.9	130	26.7	0.0
TKPE1	26	359	1.91	0.90	0.67	2.4	6.5	37	42	12	53	108	0.50	0.8	0.7	0.9	0.0
TKPE2	9	389	2.86	0.42	0.26	1.4	2.1	67	3.9	2.4	4.4	11	0.41	1.5	0.0	0.0	0.0
TKPP1	6	73	0.68	0.37	0.09	0.2	0.3	62	3.2	8.8	2.0	14	0.14	0.9	0.0	0.0	0.0
TKPP2	102	2024	3.79	1.09	0.20	398	412	97	4.7	5.3	14	24	0.59	154	551	3.6	263.6
TKPP3	3	307	1.24	0.49	0.18	0.1	0.4	19	4.5	2.4	92	99	0.93	0.0	26.2	4002	0.4
CRPE1	1	3	n.a.	n.a.	0.03	0.4	0.5	81	0	0	0	0.6	0.78	1.3	3.8	3.0	0.0
CRPE2	6	25	1.09	n.a.	0.00	0.6	1.1	54	0	14	9	23.2	0.39	1.6	33.4	20.9	0.0
CRPE3	3	3	n.a.	n.a.	0.39	5.6	5.8	98	0	0	1	1.2	0.95	0.0	2.4	>2.4	0.0
CRPE4	61	1	n.a.	n.a.	0.18	0.8	3.4	24	6	1	5	11.3	0.43	1.2	8.9	7.7	729.7
CRPE5	1	4	n.a.	n.a.	0.03	0.4	1.6	26	0	0	1	0.6	0.87	0.7	3.8	5.3	139.6
CRPP1	5	98	4.95	1.02	0.04	0.8	2.5	33	1	0	0	1.6	0.27	1.9	13.9	7.1	0.4
CRPP2	17	179	8.45	1.43	1.23	180	180	100	n.a.	n.a.	n.a.	n.a.	n.a.	14.4	335	23.2	35.4
CRPP3	3	284	2.55	2.70	0.02	0.2	2.1	12	1	0	2	3.3	0.51	0.2	5.3	25.8	0.8
CRPP4	1	33	0.67	1.25	0.47	2.3	2.4	96	3	1	8	11.2	0.68	3.3	3936	1196	8.6
CRPP5	3	147	0.26	1.08	0.23	128	129	100	10	16	98	124.4	0.79	2.4	181	76.9	2.0
CGPE1	3	129	0.76	1.21	0.04	0.2	25.8	1	0	n.a.	n.a.	0.0	n.a.	4.0	997	248	4.1
CGPE2	50	868	1.98	1.12	0.00	2.6	59.1	4	2	n.a.	n.a.	2.0	n.a.	n.a.	n.a.	-	283.4
CGPE3	7	65	2.14	2.63	2.49	0.0	1.0	3	1	n.a.	n.a.	0.5	n.a.	0.2	23.8	96.7	0.2
CGPE6	78	12			1.35	0.0	0.3	1	0	n.a.	n.a.	0.0	n.a.	0.2	132	652	0.0
CGPP9	1	16	n.a.	1.28	0.69	9907	9909	100	0	0	0	0.5	0.27	0.1	5.9	44.9	0.0
CGPP13	4	24	n.a.	1.08	1.54	162	242	67	0	0	0	0.6	0.58	1.8	5.8	3.3	0.0
S-217 PP	14	112	3.61	1.10	0.22	4.3	6.2	69	0	0	1	0.8	0.77	40.4	135	3.3	0.0
C-216 PE	29	105	2.78	1.38	0.25	0.3	15.7	2	2	0	3	4.8	0.64	8.5	58.1	6.9	1.0
C-216 PP	1	88	1.66	1.74	0.18	0.7	9.1	7	0	0	0	0.4	0.42	26.4	159	6.0	3.3

n.d.: not determined.

n.a.: not available.

ΣPCBs: sum of concentrations of CB-8, CB-18, CB-28, CB-52, CB-49, CB-44, CB-74, CB-66, CB-101, CB-99, CB-87, CB-110, CB-118, CB-105, CB-151, CB-149, CB-146, CB-153, CB-138, CB-158, CB-128, CB-167, CB-156, CB-157, CB-178, CB-187, CB-183, CB-177, CB-172, CB-180, CB-170, CB-189, CB-199, CB-196, CB-206, CB-195, CB-194, CB-206, CB-209.

ΣPAHs: sum of concentrations of anthracene, phenanthrene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzofluoranthene, benzo[e]pyrene, benzo[a]pyrene, benzo[a]pyrene, perylene, indeno [1,2,3-cd] pyrene, benzo[ghi]perylene, and coronene.

MP/P: a ratio of the sum of 1-methylphenanthrene, 2-methylphenanthrene, 3-methylphenanthrene, 9-methylphenanthrene to phenanthrene, Pyr/Fluo: A ratio of pyrene to fluoranthene.

H/L ratio: a ratio of the sum of benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzofluoranthene, benzo[e]pyrene, benzo[a]pyrene, benzo[a]pyrene, perylene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene and coronene to sum of anthracene, phenanthrene, methylphenanthres, fluoranthene, pyrene.

ΣPBDEs: sum of concentrations of BDE-3, BDE-7, BDE-15, BDE-17, BDE-28, BDE-49, BDE-71, BDE-47, BDE-66, BDE-77, BDE-100, BDE-119, BDE-99, BDE-85, BDE-126, BDE-154, BDE-153, BDE-138, BDE-183, BDE-209.

ΣDDTs: sum of DDT, DDE, and DDD.

Total NP: sum of concentrations of all the 11 peaks identified as 4-nonylphenol.

dominant in petrogenic sources. A ratio of sum of methylphenanthrenes to phenanthrene (MP/P ratio) is calculated as an index of the abundance of alkylated PAHs. The MP/P ratio in the plastic fragments ranged from 0.26 to 8.45, with an average of 1.72 ± 1.59 (Table 2). According to Saha et al. (2009), the MP/P ratios indicative of exclusively petrogenic and exclusively pyrogenic sources of PAHs are 0.4 and 2.0, respectively. MP/P ratios also indicate that PAHs in the plastic fragments are heavily influenced by petrogenic sources. In particular, samples with higher PAHs concentrations than 1000 ng/g (KNPE2, KNPE3, TKPP2) had MP/P ratio of exclusive petrogenic origin (>2). However, Rios et al. (2010) reported a pyrogenic signature in the samples from Central Pacific Gyre. In future efforts, we should investigate the cause of the difference.

DDTs concentrations (i.e., sum of DDT, DDD, and DDE) in the fragment samples ranged from 0.2 to 124 ng/g (Table 2). Their spatial distributions were different from those for PCBs and PAHs. This is probably due to DDT being introduced into the environment as a pesticide that was applied for agriculture fields in different geographic locations than the PCBs and PAHs. The agricultural application was banned in most areas and now it is mostly the environmental redistribution of legacy pollution that is occurring. In addition, limited amounts of DDT are currently used in tropical countries for anti-Malaria control. Detection of high concentrations of DDTs around 100 ng/g in the plastic samples from Vietnam (TKPE1, TKPP3) and Costa Rica (CRPP5) can be explained by the current application of DDT pesticide for the anti-Malaria control. DDT is predominant over its degradation products (DDD and DDE) in the samples (TKPP3 and CRPP5), which is expressed as higher ratios of DDT to the sum of DDT, DDD, and DDE (i.e., 0.93 and 0.79, respectively). The dominance of DDT indicates the presence of DDT family compounds recently released to the environment. Legacy pollution of DDTs was also detected in some locations in Japan (e.g., ODPE00 and ODPE01) where degradation products of DDT were dominant. Among the degradation products, DDE was generally dominant over DDD. This is reasonable because the plastic fragments are floating on the sea surface where conditions favor aerobic conversion of DDT to DDE.

PBDEs were detected in all the fragment samples. Concentrations of total PBDEs including all the congeners ranging from tri-BDE to BDE209 were from 0.02 to 9900 ng/g. Higher concentrations (over 100 ng/g) of PBDEs were detected in PP fragments at several locations including remote beaches (e.g., TKPP2, CRPP2, CRPP5) and the open ocean (CGPP9, CGPP13) as well as urban beaches (KNPP1, KNPP3) as shown in Fig. 5a. BDE209 represented over 97% of measured congeners in these samples except for CGPP13 which was accounted for 67% of the total PBDEs (Fig. 5a). BDE209 is used as flame retardant, often referred to as deca-BDE, in Asian countries and USA. These high concentrations of PBDEs are likely to be derived from deca-BDE in the polypropylene plastics. Normally deca-BDE is added to plastic products at much higher concentration. For example in waste electrical and electronic equipment, tens to thousands μg/g of BDE209 have been detected (e.g., Morf et al., 2005). Yet, concentrations of BDE209 detected in the plastic fragments in the present (up to 10 μg/g) study were lower than those normally added to plastics. Lower concentrations can be explained by photodegradation of BDE209 (Soderstrom et al., 2003) during floating on the sea surface.

In most of samples that had lower concentrations of PBDEs than 100 ng/g, BDE47, BDE99, and BDE28 were the dominant congeners in the samples (Fig. 5b). Because these congeners are commonly detected in seawater (Mizukawa et al., 2009) and are hydrophobic, these lower brominated congeners in plastics suggests sorption from seawater. Significant concentrations of BDE183 were detected in plastic fragments from Kugenuma, Odaiba, Tonkin Bay, Costa Rica, and Pacific Central Gyre. BDE183 accounted for ∼1% to ∼ 60% of all the congeners in the samples. Especially, samples from Pacific Central Gyre had much higher concentrations of BDE183, ∼60% of all the congeners. BDE183 is generated through photodegradation of BDE209 (Soderstrom et al., 2003). BDE183 is one of major congeners in commercial products octa-BDE (La Guardia et al., 2006). These products are currently not used in Japan.

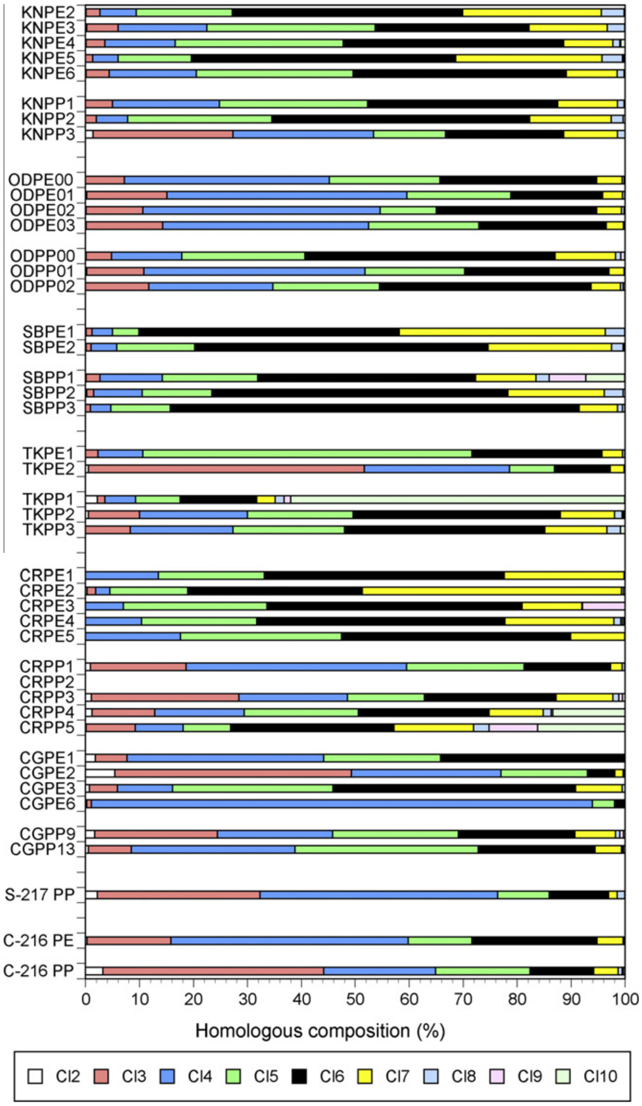

Fig. 3. Homologous compositions of PCBs in Marine plastic fragments.

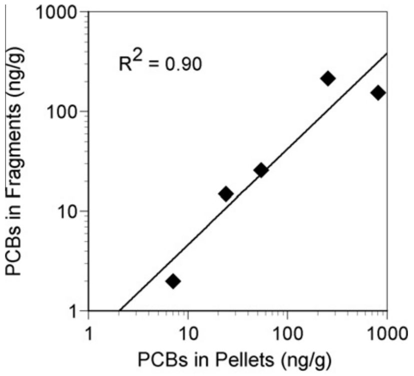

Fig. 4. Relationship of PCB concentrations in plastic fragments and plastic resin pellets.

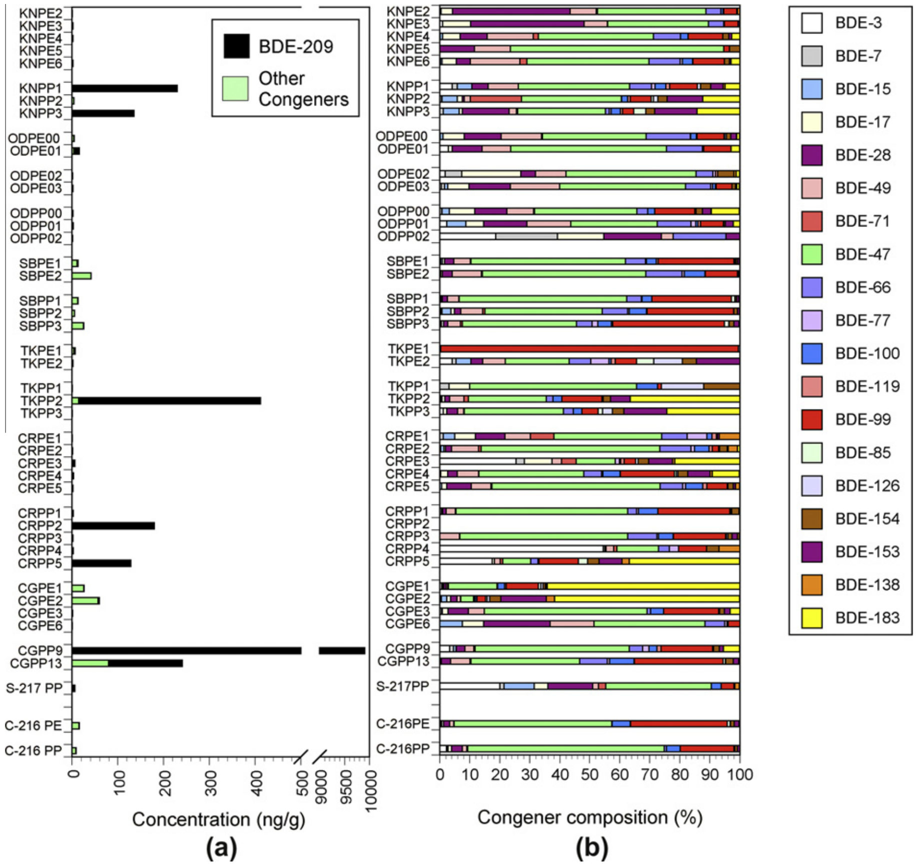

Fig. 5. Concentrations (a) and Compositions (b) of PBDEs in marine plastic fragments.

However, they are still used in the other Asian countries and the USA (Hites, 2004). Thus, BDE183 in the plastic samples has two potential sources: degradation of BDE209 originally in the plastic and/or sorption of octa-BDE-derived BDE183 in seawater. For the sample from the Pacific Central Gyre, where there is lower direct input from land anthropogenic sources and therefore trace PBDE concentrations are expected in seawater, BDE183 is more likely to be from photodegradation of BDE209. For the other locations, source identification of BDE 183 cannot be ascertained from present data.

Alkylphenols (NP and OP) were detected in almost all fragment samples including pieces from the open ocean and remote beaches (Fig. 6a, Table 2). The concentration of NP was 0.3–3940 ng/g, and 0.1–153 ng/g for OP (Table 2). Again, these chemicals can be derived from additives and/or sorption from surrounding seawater. Alkylphenol polyethoxylates (APEO) surfactants are used mainly for industrial cleaning agents. APs are generated during degradation of APEO in sewage treatment plants and receiving waters. APs are supplied to the ocean via rivers and sewage effluents. Significant concentrations of nonylphenols (e.g., ~10 ng/L) were observed in surface waters of Tokyo Bay, while much higher concentrations of nonylphenol (~500 ng/L) were detected in waters from adjacent rivers (Nakada, 2003). Only trace concentrations (0.002–0.15 ng/L) of NP were detected in open ocean waters (Kannan et al., 1998). Therefore, sporadically high concentrations of nonylphenols in the plastic fragments from open ocean (CGPE1) and remote beaches (CRPP4) cannot be ascribed to sorption from seawater but rather additive-derived nonylphenol. Trisnonylphenol phosphates (TNP) are compounded to some plastic products as anti-oxidant agents (Gilbert et al., 1986). The TNP product itself contains NP as unreacted raw material. Furthermore, TNP can be hydrolyzed and oxidized to NPs. Also APEOs are used for emulsion polymerization in plastic production (Talmage, 1994). Detection of nonylphenols in plastic products has been reported (Isobe et al., 2002; Loyo-Rosales et al., 2004). These additive-derived NP are a source of NP detected in oceanic plastic fragments. This was supported by the predominance of NP over OP in those samples. OP-based additives are rarely used and only NP is detected from commercial plastic products (Isobe et al., 2002). Some amounts of octylphenolpolyethoxylates, which generate OP, are used as surfactant and discharged to urban aquatic environments. The ratio of NP to OP (NP/OP ratio) in sewage effluents and Tokyo Bay water was reported as 7.0 ± 1.5 ($n = 12$) and 8.4 ± 2.2 ($n = 15$), respectively, (Nakada, 2003). Due to the more hydrophobic nature of NP (log Kow: 4.48) than OP (log Kow: 4.12), NP/OP ratios in Tokyo Bay sediments were higher (19.3 ± 9.1) than the water samples (Nakada, 2003). The plastic fragments that had sporadically high concentrations of nonylphenols (e.g., CGPE1, CRPP4) exhibited very high NP/OP ratio ranging from 156 to 1200 (Fig. 6b). This would indicate that the NP in these samples is derived from plastic additives and not from sorption from surrounding seawaters. Lower NP/OP ratios were observed in the other fragment samples that generally had lower NP concentrations (Fig. 6). Lower concentrations of NP in these fragment samples are most likely from sorption of surfactants-derived NP.

In most locations including urban coasts, only trace concentrations (<1 ng/g) of BPA were detected. Again sporadically high concentrations of BPA were detected in some plastic fragment samples from remote beaches (CRPE4, CRPE5) and the open ocean (CGPE2) (Table 2). BPA is a constituent monomer of polycarbonate plastic and epoxy resin. Unreacted monomers in the plastics and resin and degradation products from the polymers can be leached to the environment. BPA is also used as additives to some plastics. Leaching of BPA from commercial plastic products (Casajuana and Lacorte, 2003) and dumped plastics (Teuten et al., 2009) has been reported. A combination of these sources contribute BPA to sewage, sewage effluents, river water, and coastal waters. Concentrations of BPA in coastal water are the same order of magnitude as nonylphenol (e.g., low ng/L range in Tokyo Bay water, Nakada, 2003). Due to its lower hydrophobicity (log $K_{ow} = 3.40$), sorption of significant concentrations of BPA to marine plastics is unlikely. Sporadic high concentrations of BPA were detected in plastic fragments from remote coasts (730 ng/g at CRPE4) and open ocean fragments (283 ng/g at CGPE2; Table 2, Fig. 2). These higher BPA concentrations are most likely a result of BPA being a constituent of the plastic products.

5. Ecotoxicological implication

Regarding PCBs and PAHs, higher concentrations were observed in plastic fragments from urban beaches. Hence, risks associated with these hydrophobic pollutants are higher in urban beaches than remote beaches and the open ocean. However, the presence of sporadically high concentrations of PCBs and PAHs should be continually assessed for their toxicological effects on marine life such as sea birds. Conventionally sediment particles, colloids, and soot have been considered as carrier of HOCs. Those carriers are basically heavier than water and can be eventually deposited to sediments in coastal zones. Plastics are considered as another important carrier of pollutants in marine environments. Because PP and PE are lighter than water and persistent, they do not sink to coastal sediments unless entwined with other more dense debris and can be transported to the open ocean. Thus, plastic fragments can laterally deliver HOCs to more remote area and open ocean. Due to partitioning processes, PCB concentrations in plastic fragments from remote beaches and the open ocean could be approaching equilibrium (i.e., trace) concentrations through desorption during offshore transport. Lower concentrations of the HOCs were observed in the offshore and remote locations which is in accord with the expectation of desorption. However, the present study also demonstrated that some plastic fragments from remote beaches and the open ocean had sporadically high concentrations of HOCs. If they were in equilibrium with surrounding water (i.e., open ocean or remote waters), their concentrations would be uniform and trace. However, we observed high variability in the concentrations and compositions of HOCs in the plastic fragments from open ocean and remote beaches. This indicates that it is likely that these plastic fragments did not have a long enough residence time to reach equilibrium, pointing to the presence of "fresh" plastic fragments in the open ocean.

Absorption of HOCs to plastic fragments is kinetically controlled by diffusive transfer within the plastic matrix. In the case of plastic sheeting 20 μm thick, it takes a few days to reach equilibrium (Adams et al., 2007). Pellets of a few mm take ~200 days to reach equilibrium (Karapanagioti et al., 2010). Plastic fragments were similar to or larger than resin pellets and, therefore, would take more than 200 days to reach equilibrium. If plastic fragments in urban waters are transported rapidly by ocean currents to open ocean or remote areas; and then marine organisms could take up such "freshly contaminated" plastics before they reach equilibrium, the organisms would be exposed to greater amounts of HOCs. Gouin et al. (2011) assessed the importance of marine plastics as vectors of HOCs to marine organisms as insignificant. However, their calculations were based largely on equilibrium portioning. In future efforts, the non-equilibrium phenomena and sporadic high concentrations of HOCs in the open ocean that we and Rios et al. (2010) observed

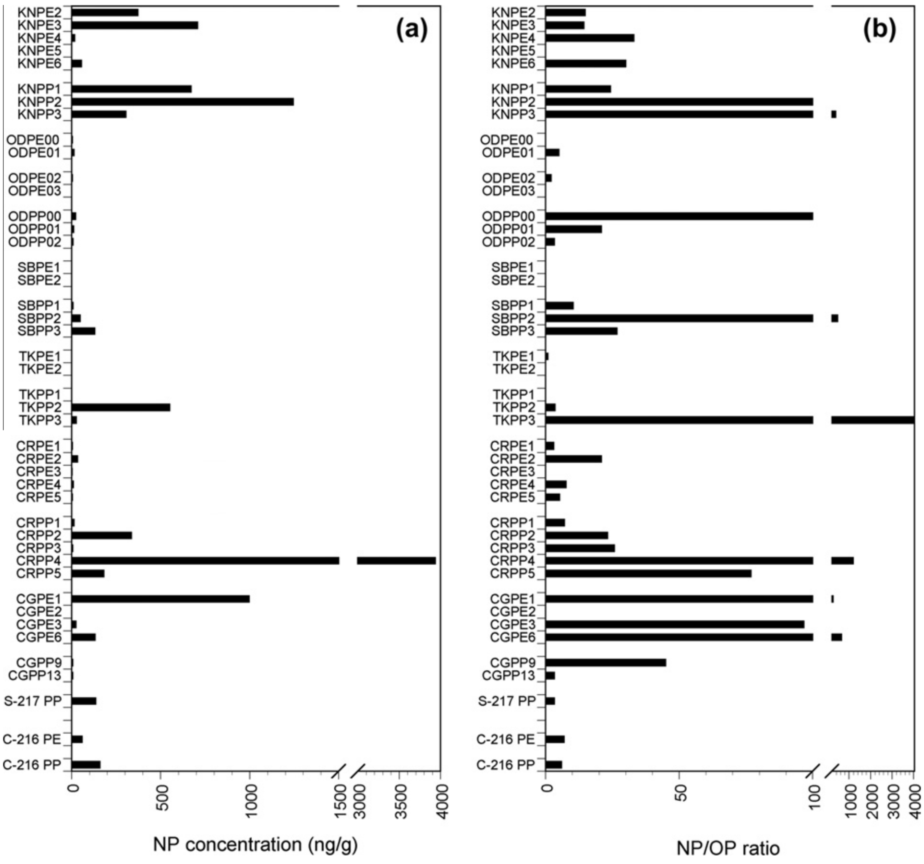

Fig. 6. NP concentrations and NP/OP ratio in marine plastic fragments.

should be incorporated into the assessment. Zarfl and Matthies (2010) evaluated the potential of marine plastics to transport PCBs and PBDEs to the Arctic by using the concentrations of PCBs and PBDEs reported in our previous paper (Teuten et al., 2009). They also estimated an insignificant role of marine plastics. Future calculations should use more data and include the sporadically high concentrations of PCBs and PBDEs.

Furthermore, in several plastic fragment samples from remote beaches and the open ocean, relatively high concentrations of additive-derived chemicals, i.e., NP, BPA, PBDEs were detected. Due to remoteness from the urban areas, concentrations of these anthropogenic compounds in the open ocean and remote coastal waters are low. Due to the metabolizable nature of NP (Takeuchi et al., 2009) and BPA, they are not biomagnified up the food web to higher trophic levels. Thus, NP and BPA in the plastic fragments could be significant exposure pathways of these chemicals to the higher-trophic-level animals. Also, because PBDEs are less biomagnified than PCBs in marine ecosystem (Mizukawa et al., 2009), exposure of PBDEs from ingested plastic could be more important than those through the food web. To assess the actual exposure of the chemicals to marine biota, bioavailability of these chemicals in plastics should be considered. Chemicals in plastics are thought to be less bioavailable due to diffused presence in plastic polymer matrix. However, chemical components in digestive tracts such as surfactants can enhance the bioavailability (i.e., leaching) of these chemical and uptake by organisms. Further studies are needed to address these issues of bioavailability and bioaccumulation for the chemicals associated with plastics as reported in the preceding.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.marpolbul.2011.06.004.

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