

# Polycyclic aromatic hydrocarbons (PAHs) in new unexposed and beached expanded polystyrene foams

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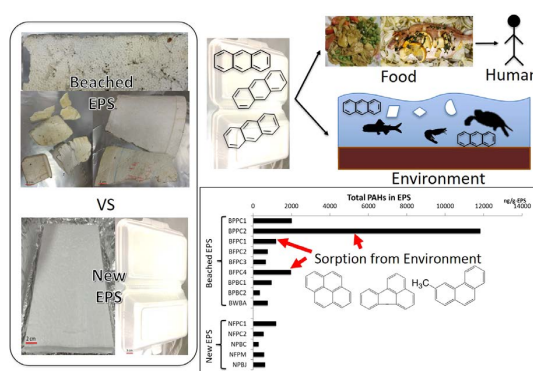
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## ABSTRACT

Among the plastic debris surveyed in the marine environment, a large quantity in terms of volume was found to be expanded polystyrene foam (EPS). This is due to its light weight and popularity as a packaging material. In this study, 28 compounds of polycyclic aromatic hydrocarbon (PAHs) were analyzed in both new and beached EPS collected in California, USA. In beached EPS, a variation in total PAH concentrations of up to 2 orders of magnitude was observed in the 337 to 11,800 ng/g-EPS range. Meanwhile, new EPS showed less variation in total PAH concentrations ranging from 282 to 1,194 ng/g-EPS. Most PAH species were shown to have higher concentrations in beached EPS than in new EPS, suggesting the ability of EPS to sorb PAHs from surrounding seawater. However, with anthracene, there was a significant depletion in beached EPS compared to new EPS. The fact that significant PAH concentrations were found in new unexposed EPS, including food packaging, highlights EPS as an exposure source of toxic chemicals to human food stocks upon migration, as well as to marine wildlife upon disposal. PAH concentrations and composition in new and beached EPS were consistent with those of samples from Japan, Malaysia and Australia, suggesting that the problems associated with EPS is a global issue.



**Key words:** plastic pollution; anthracene; byproducts; food container; styrofoam

## INTRODUCTION

The omnipresence of plastic debris in the marine environment has become a constant problem in recent years, and has attracted the attention and concern of various groups ranging from researchers, non-profit organizations, citizens and students to policy makers and governmental bodies. This is due to the myriad of negative effects that plastic debris has on marine organisms. These include entanglement, asphyxiation (GESAMP, 2010), and the potential for both plastics and associated chemicals to be transferred into marine wildlife and finally to humans through the food web (Teuten et al., 2009; Thompson et al., 2009; Yamashita et al., 2011; Tanaka et al., 2013, 2015, 2018; Ragusa et al., 2021). Among the numerous types of plastic debris found on beaches and in the marine environment, a large quantity, in terms of pieces or volume, has

been found to be expanded polystyrene foam (EPS, Moore et al., 2011). EPS is extremely light, as a result of the heating expansion process, and can support many times its own weight making it a very useful and versatile material. Its non-interconnected cells also allow EPS to be a great heat insulator. Thus, EPS is widely used for packaging especially in the shipping and food industries (Freinkel, 2011). Consequently, EPS is also a major component of urban litter, municipal solid waste (Andrady and Pegram, 1991; Rochman et al., 2013a) and eventually, the open oceans (Moore et al., 2011).

Of the polymers frequently found in the environment, polystyrene (PS) is considered one of the “dirtiest”. Styrene oligomers derived from PS themselves are reasonably suspected of being potentially toxic to marine organisms and mammals (Kitamura et al., 2003; Kwon et al., 2015). Previous studies have also demonstrated the possibility of residual monomers and additives such as plasticizers and antioxidants migrating from plastic packaging to food contents and food simulants (Lickly et al., 1995; Guart et al., 2011; Fasano et al., 2012; Genualdi et al., 2014; Lin et al., 2017). Moreover, EPS has also been shown

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to contain much greater concentrations of polycyclic aromatic hydrocarbons (PAHs) than other polymers found in the marine environment (Van et al., 2012) with virgin PS pellets observed to readily possess up to 200 times more PAHs than those of other polymers, such as polyethylene and polypropylene (Rochman et al., 2013b). Van et al. (2012) also reported new unexposed EPS containing up to 2 orders of magnitude more PAHs than virgin PS pellets, suggesting that the heating and expansion process used to convert PS to EPS generates and infuses PAHs into EPS. This is of concern considering the fact that PAHs have been shown to be toxic, mutagenic and carcinogenic (Kennish, 1997), and that EPS is widely used for food packaging. Thus far, there has been little work focusing on PAHs in EPS with only a few studies such as Van et al. (2012) and Li et al. (2017), touching on the topic. In addition, in these studies, only total PAHs were reported on a mixture of EPS packaging types in Van et al. (2012) and only selected parent PAHs were analyzed in both Van et al. (2012) and Li et al. (2017). By only reporting on selected parent PAHs, the toxicity estimate or PAHs contamination load estimate could be significantly underestimated (Andersson and Achten, 2015). Apart from that, methylated PAHs are also speculated to be potentially mutagenic and/or carcinogenic (Baird et al., 2007). In addition, the relative abundance of methylated PAHs could provide useful information on the sources and generation processes of PAHs (Blumer, 1976; Saha et al., 2012).

Hence, in this study, 28 individual PAH species including 13 methylated PAHs together with 15 parent PAHs were quantified in both new unexposed EPS and beached EPS from California, USA. EPS was also investigated based on usage types, such as packaging blocks, food packaging containers and packaging peanuts. This paper provides more detailed data on PAHs in EPS by aiming to: 1) identify whether EPS acts as a source or sink for individual PAH species in the marine environment; and 2) identify potential hazards caused by EPS to the environment and to human health through an anal-

ysis of different EPS packaging types. To generalize our findings, we also collected and analyzed EPS from Japan, Malaysia and Australia.

## MATERIALS AND METHODS

The primary samples of this case study were three new unexposed and 8 beached EPS samples from California, USA. A new EPS packaging block (NPBC) was obtained from an electronic store and two food packaging containers (NFPC 1 and 2) were obtained from diners in the Silicon Valley region, California. Beached EPS were collected at Sea View Parkway, Alameda, California, USA (37.74775, -122.24933) by volunteers from Bay Farm Elementary School, Alameda. Based on the shapes and texture of the debris, the samples were sorted into packaging block (BPBC 1 and 2), packaging peanut (BPPC 1 and 2), and food packaging (BFPC 1–4). To address the global spread of EPS problem, 2 additional new EPS samples and 1 additional beached EPS sample were obtained from other countries. These were; 1 new packaging block from an electronic store in Tokyo, Japan (NPBJ), 1 new EPS food packaging container from a restaurant in Johor, Malaysia (NFPM) and beached EPS weather balloon pieces (BWBA) were collected by Tangaroa Blue Foundation from Four Mile Beach, Port Douglas, Queensland, Australia (-16.49811, 145.46677). The collected beached EPS samples were airmailed to the laboratory in Tokyo. Detailed information about the samples can be found in Table 1 and pictures of the samples are available in supporting information (Fig. S1–S7).

Approximately 0.20–0.25 g of EPS was extracted from each sample by dissolving it in 5 mL dichloromethane (DCM). Extracts were then filtered through glass fiber filters (Whatman GF/F, 47 mm) to eliminate aggregates. The filtrates were then subjected to gel permeation chromatography (GPC, 2 cm i.d × 30 cm, CLNpak EV-2000; Showa-denko, Tokyo, Japan) at a flow rate of 4 mL/min to remove polymers. The GPC fraction was collected at retention times of 15 to 25 min and further

Table 1 Expanded polystyrene foam samples detailed information and abbreviations

EPS samples	Abbreviation	Location
<i>New unexposed EPS</i>		
Packaging block	NPBC	Sunnyvale, California, USA
Food packaging -1	NFPC1	Silicon Valley, California, USA
Food packaging -2	NFPC2	Silicon Valley, California, USA
Packaging block	NPBJ	Tokyo, Japan
Food packaging	NFPM	Segamat, Johor, Malaysia
<i>Beached EPS</i>		
Packaging block (small)	BPBC1	Seaview Parkway, Alameda, California, USA
Packaging block (big)	BPBC2	Seaview Parkway, Alameda, California, USA
Food packaging (smooth pieces)	BFPC1	Seaview Parkway, Alameda, California, USA
Food packaging (cup like pieces)	BFPC2	Seaview Parkway, Alameda, California, USA
Food packaging (mix pieces)	BFPC3	Seaview Parkway, Alameda, California, USA
Food packaging (cup)	BFPC4	Seaview Parkway, Alameda, California, USA
Packaging peanut (white)	BPPC1	Seaview Parkway, Alameda, California, USA
Packaging peanut (pink)	BPPC2	Seaview Parkway, Alameda, California, USA
Weather Balloon	BWBA	Four Mile Beach, Port Douglas, Australia

purified through 2 step silica gel column chromatography; 5% H<sub>2</sub>O-deactivated silica gel column (1 cm i.d × 9 cm) and separated through fully activated silica gel column (0.47 cm i.d × 18 cm) into 3 fractions: fraction I (n-alkanes and hopanes), fraction II (PCBs and DDE), and fraction III (DDT, DDD, 4 HCH isomers [ $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ ] and PAHs). The PAHs in fraction III were determined via gas chromatography/mass spectrometry (GC-MS, Agilent Technology, 5,973 series). The sum of 28 quantified PAH species ranging from 3-ring to 7-ring species consisted of phenanthrene (Phe), anthracene (Anth), 4H-cyclopenta[*def*]phenanthrene (CPP), 3-methylphenanthrene (3-MP), 2-methylphenanthrene (2-MP), 9-methylphenanthrene (9-MP), 1-methylphenanthrene (1-MP), fluoranthene (Fluo), pyrene (Py), benzo[*b*]fluorene (a-MPy), 4-methylpyrene (b-MPy), 2-methylpyrene (c-MPy), 1-methylpyrene (d-MPy), benzo[*a*]anthracene (BaA), chrysene (Chry), 5 methylated isomers of chrysene or benz[*a*]anthracene, benzo[*b*]fluoranthene (BbF), benzo[*j*]fluoranthene (BjF), benzo[*k*]fluoranthene (BkF), benzo[*e*]pyrene (BeP), benzo[*a*]pyrene (BaP), perylene (Pery), indeno[1,2,3-*cd*]pyrene (IndPy), benzo[*ghi*]perylene (BgHiP), and coronene (Cor) is expressed as  $\Sigma$ 28 PAHs. Details of peak identification and instrumental conditions are available in Saha et al. (2009). PAH concentrations in the samples were recovery-corrected using recoveries of surrogates (i.e., deuterated PAHs) spiked before analysis, as described in Saha et al. (2009). The reproducibility of this analytical procedure (i.e., GPC, 2 step column chromatography and instrumental determination) was confirmed through the analysis of 4 aliquots from a single extract of weather balloon EPS from Four Mile Beach, Port Douglas, Australia. The relative standard deviation of the concentrations was less than 10% for all PAH species. Recovery was tested by spiking the extracts with authentic standards; the recovery was >96% for all PAHs except perylene with a recovery of 74–79%. A procedural blank using only solvent was run with every set analyzed (4 samples). Analytical values <3 × the corresponding blanks were considered below the limit of quantification (LOQ). The LOQs were normally ~50 ng/g-EPS for  $\Sigma$ 28 PAHs.

## RESULTS AND DISCUSSION

PAH concentrations of all samples can be found in Table 2.  $\Sigma$ 28 PAHs observed in beached EPS ranged from 337 ng/g-EPS to 11,821 ng/g-EPS with a variation of up to 2 orders of magnitude. Meanwhile,  $\Sigma$ 28 PAHs concentrations in new EPS varied less with a range of 282 ng/g-EPS to 1,194 ng/g-EPS. Overall, higher concentrations of PAHs were detected in beached EPS samples than in new EPS samples. This can be ascribed to the sorption of hydrophobic organic compounds (HOCs) including PAHs to PS. Rochman et al. (2013b) demonstrated the sorption of PAHs to PS pellets through in-situ sorption experiments. The trend of higher PAH concentrations in beached EPS was more pronounced for packaging blocks with beached EPS (337–949 ng/g for BPBC1 and 2) compared with new EPS (282 ng/g for NPBC). However, the trend was not so distinct for food packaging. Beached EPS food packaging (BFPC1–4) had higher range of  $\Sigma$ 28 PAHs with 657–1,962 ng/g, than new samples (NFPC1 and 2) with a range of 539–1,194 ng/g, although the results overlapped. Overall, EPS could be seen to sorb PAHs from the surrounding seawater

more than being a source of PAHs in the environment. However, the significant moderate to high PAHs found in new EPS samples, especially in NFPC 1, are of concern.

The results of the present study are consistent with, and support the fact that sorption of HOCs to plastics is controlled by various factors, including the size of plastics and their surface porosity (Endo and Koelmans, 2016). The highest concentrations of PAHs were found in beached packaging peanuts BPPC2 and BPPC1 with 11,821 and 2,005 ng/g-EPS respectively (Fig. 1). This was followed by beached food packaging BFPC 4 and BFPC 1 with PAH concentrations of 1,962 and 1,188 ng/g-EPS respectively. As shown in Table 1, smaller, lighter, and more porous pieces of EPS were shown to have higher concentrations of PAHs, i.e. BPBC1 > BPBC2 and BPBC2 > BPPC1. The porosity and smaller size could have provided a greater surface area for PAHs sorption. Individual EPS sample conditions can be observed in supporting information (Fig. S1–S7). The BPPC2 pieces were not only porous but also highly weathered with cracks and sand-like aggregates embedded within the cracks of the sample, which possibly caused the extremely high PAH concentrations.

The composition of PAHs in both new and beached EPS samples was carefully examined in order to understand the difference in the distribution of individual PAHs (Fig. S8). Anthracene was observed to be predominant in NPBC, followed by phenanthrene, while small amounts of methylchrysenes and methylphenanthrenes were detected (Fig. 2a). The presence of alkylated PAHs in new EPS could be derived from raw materials as plastics are basically made out of hydrocarbons in the form of oil and gas as raw materials (Hocking, 1991; Freinkel, 2011; Rochman et al., 2013b). 1-MP and 2-MP have also been previously reported in virgin PS pellets (Rochman et al., 2013b). Although alkylated PAHs were present at significant but trace concentrations, most new unexposed EPS exhibited lower ratios of methyl PAHs to parent PAHs (Table 2). The depletion of methylated PAH species is evidence of pyrogenic origin of PAHs in unexposed EPS samples (Fig. S9), i.e., pyrogenic PAHs could be generated during the heating processes during EPS production (Harper, 2000).

Meanwhile, in both beached samples BPBC1 and BPBC2, a significant depletion of anthracene and a slight depletion of methylchrysenes were observed as shown in Figs. 2b and 2c. On the other hand, a substantial amount of phenanthrene was observed in both beached samples. Fluoranthene and pyrene showed only a slight increase in BPBC2, but a significant increase in pyrene could be seen in BPBC1. Similar trends in the shift of phenanthrene and anthracene were also observed in food packaging EPS between new food packaging (NFPC1 and NFPC2) and beached food packaging (BFPC1, BFPC2, BFPC3 and BFPC4), as shown in Fig. 3. This unique aspect in the PAHs composition of anthracene predominance in unexposed EPS, which was also reported by Rochman et al. (2013b), was not observed in any other pyrogenic sources examined by Saha et al. (2009, 2012). Thus, this could be related to the polymerization of styrene with the benzene ring. Anthracene in food packaging EPS was found at concentrations of hundreds of ng/g. Anthracene is considered toxic and its leaching and migration into food are of concern (Li et al., 2017).

Phenanthrene and anthracene are similar in terms of mo-

Table 2 PAH concentrations and ratios in expanded polystyrene foam samples

	Unexposed EPS					Beached EPS								
	NPBC	NFPC1	NFPC2	NPBJ	NFPM	BPBC1	BPBC2	BFPC1	BFPC2	BFPC3	BFPC4	BPPC1	BPPC2	BWBA
Phe	83.4	<i>115.2</i>	27.6	176.8	137.5	241.9	242.1	751.9	524.9	27.2	1,197.4	709.2	3,077.9	485.8
Anth	165.2	881.4	397.0	273.6	259.2	32.7	40.6	38.1	10.8	3.2	188.4	2.2	119.5	18.5
CPP	<i>0.8</i>	23.5	7.5	2.3	11.4	3.1	2.1	17.5	11.6	24.0	25.3	8.9	192.1	6.2
3-MP	2.6	163.3	66.3	27.5	25.0	34.4	3.6	18.6	17.6	30.7	53.3	50.0	246.3	42.4
2-MP	2.5	<i>16.2</i>	4.9	36.3	10.9	42.6	5.0	26.3	25.0	26.4	84.4	66.1	421.2	54.9
9-MP	2.6	<i>16.1</i>	8.0	24.0	20.3	25.7	9.1	70.8	34.0	42.4	20.0	58.5	118.6	39.0
1-MP	<i>1.3</i>	<i>3.0</i>	<i>0.7</i>	16.4	4.8	15.4	2.6	14.5	12.6	11.5	25.1	30.6	184.5	28.6
Fluo	9.4	7.7	3.3	17.7	13.3	41.6	16.5	78.8	52.2	171.5	246.2	121.0	3,262.3	25.8
Pyr	2.4	1.5	<i>1.2</i>	18.0	8.4	275.8	5.0	26.7	21.3	39.6	50.3	89.7	1,232.3	21.8
aMPy	<i>0.4</i>	<i>4.6</i>	<i>1.5</i>	<i>1.6</i>	17.3	3.0	<i>0.7</i>	5.2	3.1	12.4	7.6	7.0	55.5	1.9
bMPy	0.2	2.7	0.5	3.6	9.2	101.6	0.3	7.4	2.0	8.7	4.2	24.0	59.7	3.1
cMPy	0.4	2.7	0.8	3.7	8.8	74.9	0.3	5.5	1.8	8.6	2.1	21.4	37.7	3.4
dMPy	<i>0.3</i>	<i>3.1</i>	<i>0.9</i>	2.3	6.7	25.5	<i>0.2</i>	1.9	0.8	2.9	1.2	7.5	18.2	1.9
BaA	<i>0.1</i>	<i>12.4</i>	<i>2.4</i>	3.5	4.5	<i>1.2</i>	<i>1.5</i>	<i>1.1</i>	<i>1.4</i>	4.2	<i>1.5</i>	<i>1.5</i>	122.9	1.0
Chry	<i>0.4</i>	<i>5.1</i>	<i>8.1</i>	6.0	6.8	8.4	4.5	16.4	9.4	74.2	21.3	14.3	1,025.2	5.9
eMC	4.2	16.1	2.1	2.2	1.3	0.2	1.5	0.5	0.3	1.0	0.5	6.5	5.7	1.5
fMC	6.4	22.4	3.2	0.8	1.1	0.3	2.1	2.2	1.4	1.4	9.7	5.0	82.6	0.3
gMC	1.5	6.4	2.3	0.3	3.0	3.1	0.4	4.9	3.2	2.0	2.6	14.0	64.2	0.6
hMC	2.5	<i>1.4</i>	3.3	<i>0.1</i>	3.9	1.4	<i>0.3</i>	2.5	1.2	12.6	<i>0.6</i>	7.4	7.9	<i>0.0</i>
iMC	<i>0.4</i>	<i>6.0</i>	7.4	<i>0.4</i>	<i>4.0</i>	<i>0.4</i>	<i>1.0</i>	1.2	<i>0.6</i>	6.3	<i>0.9</i>	3.1	51.8	<i>0.4</i>
BbF	<i>0.6</i>	8.9	<i>6.1</i>	1.7	<i>1.1</i>	3.5	<i>1.3</i>	54.3	5.3	60.6	6.5	46.5	319.9	<i>1.0</i>
BF	1.5	11.4	0.9	1.4	5.8	3.8	0.6	41.0	3.9	36.4	4.0	632.1	338.2	2.2
BeP	<i>0.3</i>	<i>7.1</i>	<i>1.9</i>	1.5	3.8	3.2	<i>0.9</i>	<i>0.6</i>	2.2	29.2	4.2	23.2	230.3	<i>0.7</i>
BaP	<i>0.5</i>	<i>6.0</i>	<i>8.2</i>	<i>1.0</i>	3.2	1.8	<i>1.6</i>	<i>1.3</i>	<i>1.5</i>	15.0	<i>1.9</i>	15.9	151.9	<i>0.3</i>
Pery	<i>0.2</i>	3.9	<i>2.0</i>	<i>0.3</i>	<i>0.6</i>	<i>0.2</i>	<i>0.7</i>	<i>1.3</i>	<i>0.2</i>	<i>1.4</i>	<i>1.5</i>	3.4	18.6	<i>0.4</i>
IndPy	0.2	46.1	7.3	0.7	2.0	1.6	0.2	1.7	0.8	15.0	1.6	18.5	174.2	3.5
BghiP	<i>0.3</i>	<i>11.3</i>	2.7	1.4	4.0	3.6	<i>0.9</i>	<i>1.8</i>	3.0	16.3	3.6	2.8	188.6	<i>0.7</i>
Cor	<i>0.7</i>	<i>7.1</i>	<i>1.3</i>	<i>0.6</i>	3.5	<i>0.2</i>	<i>0.6</i>	<i>0.5</i>	<i>1.4</i>	3.7	2.2	16.0	13.8	<i>0.4</i>
Σ28 PAHs	282	1,194	539	622	569	949	337	1,188	749	657	1,962	2,005	11,821	748
Σ4 PAHs	<LOQ	<LOQ	<LOQ	11	11	14	5	71	15	154	28	77	1,620	7
Σ10 PAHs	11	65	8	34	36	68	22	192	77	422	287	874	5,814	38
Σ16 PAHs	262	972	440	505	456	621	312	1,026	646	490	1,751	1,704	10,468	571
MP/P	0.1	1.7	2.9	0.6	0.4	0.5	0.1	0.2	0.2	4.1	0.2	0.3	0.3	0.3
Pyr/Fluo	0.3	0.2	0.4	1.0	0.6	6.6	0.3	0.3	0.4	0.2	0.2	0.7	0.4	0.8
MC/C	31.2	3.0	1.7	0.4	1.2	0.6	0.9	0.6	0.6	0.3	0.6	2.3	0.2	0.4
Mpy/Py	0.1	1.4	0.8	0.3	1.9	0.6	0.1	0.2	0.1	0.2	0.1	0.3	0.0	0.2
MPAH/PAH	0.1	0.2	0.2	0.2	0.2	0.5	0.1	0.2	0.2	0.3	0.1	0.2	0.1	0.3
Anth/Phe	1.98	7.65	14.38	1.55	1.89	0.14	0.17	0.05	0.02	0.12	0.16	0.00	0.04	0.04

\*Figures in *italic* are below the limit of quantification (LOQ)

\*Σ4 PAHs are the sum of BaA, Chry, BbF, BaP

\*Σ8 PAHs are the sum of Phe, Anth, CPP, 1-MP, Fluo, Pyr, BaA, Chry

\*Σ16 PAHs are the sum of Phe, Anth, CPP, 1-MP, Fluo, Pyr, BaA, Chry, BbF, BF, BeP, BaP, Pery, IndPy, BghiP, Cor

molecular weight and hydrophobicity, hence, the selective significant depletion of anthracene seen between new and beached EPS could be due to its high photoreactivity with an estimated half-life of an hour compared with phenanthrene with a half-life of 20.4 hours (Bertilsson and Widenfalk, 2002). This coupled with the fact that plastic floats on water before being beached, and thus experiencing constant exposure to sunlight, could cause anthracene to be selectively depleted and lost instead of leaching out into the marine environment. EPS can also sorb

PAHs similar to PS pellets as previously reported by Rochman et al. (2013b), especially lower molecular weight PAHs as they tend to reach saturation faster. However, significant higher molecular weight PAHs were observed in BFPC3, more than in any other food packaging samples. They were also detected in BPPC1 and BPPC2 suggesting that EPS can act as a temporary sink for most PAHs. The high porosity of EPS causes it to break down physically relatively quicker especially in the marine environment (Andrady and Pegram, 1991) creating larger

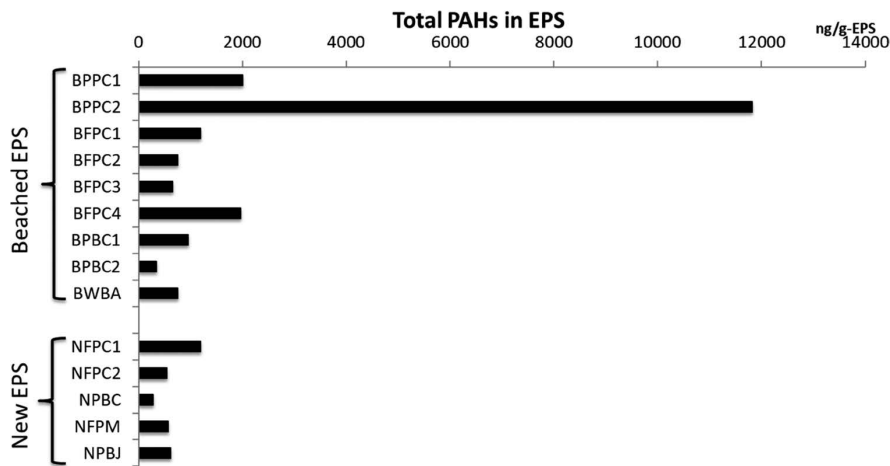
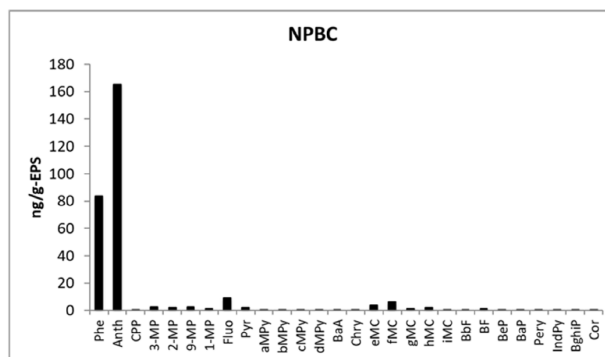
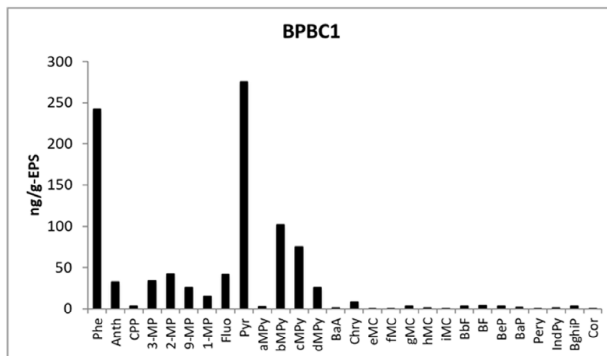


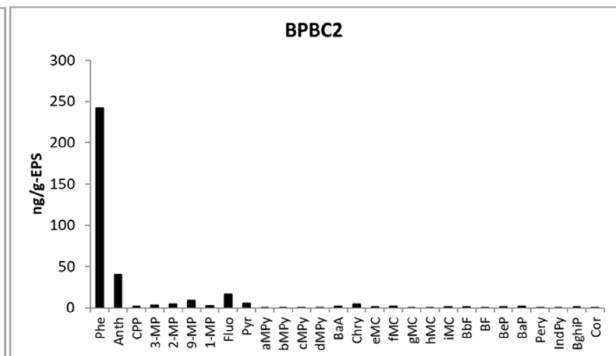
Fig. 1 Total PAH concentrations in new unexposed and beached EPS samples



(a) New unexposed packaging block (large block)



(b) Beached packaging block (small pieces)



(c) Beached packaging block (large block)

Fig. 2 PAHs composition of new unexposed and beached EPS packaging block from California, USA

quantities of smaller pieces with large surface areas for pollutant sorption. EPS has also been reported to contribute the most in terms of total PAH concentrations despite its minimal contribution in mass compared with plastic fragments and pellets according to a survey conducted in San Diego, California (Van et al. 2012).

The patterns and concentrations of PAHs found both in new and beached EPS collected in California, USA, were confirmed by those observed in Japan, Malaysia and Australia. 28 PAHs in new EPS packaging block from Japan (NPBJ) and food packaging from Malaysia (NFPM) were within the range found in new EPS from California, with 622 and 569 ng/g-EPS

respectively (Fig. 1). Although NPBJ recorded more than double the concentration of NPBC, NFPM had a very similar concentration to that of NFPC2. Beached weather balloon EPS (BWBA) from Australia also showed PAH concentrations within the range of other beached EPS, with values of 748 ng/g-EPS. Particularly, PAH concentrations and composition in weather balloon BWBA were very similar to those found in food packaging BFPC 2 with PAH concentrations of 749 ng/g-EPS (Figs. 1 & S8). This could be due to the fact that, both these samples have similar porosity and density. Weather balloons provide essential weather observations and data. Since weather balloons needed to move freely according to wind

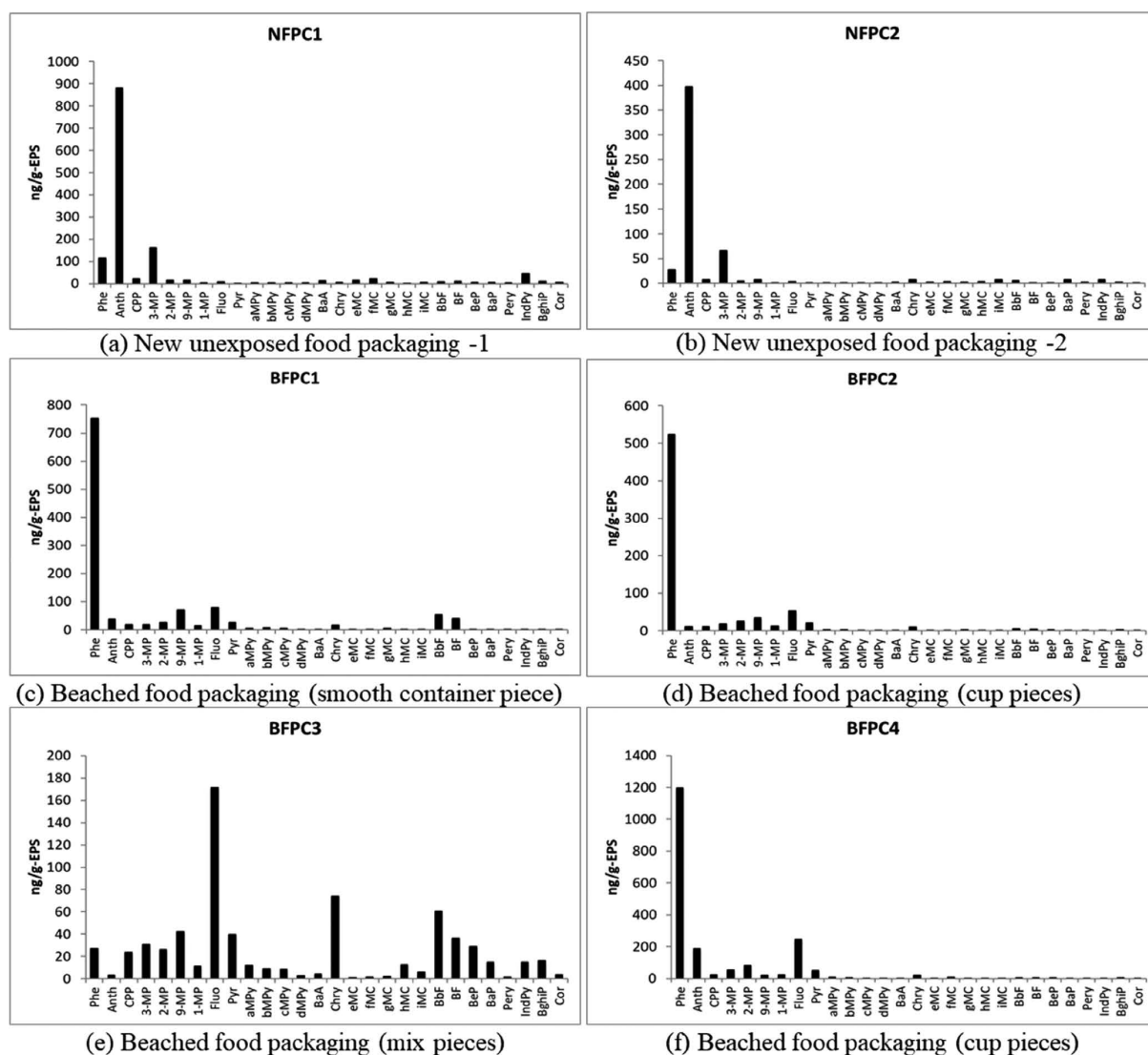


Fig. 3 PAHs composition of new unexposed and beached EPS food packaging from California, USA

speeds and directions, extremely light material, (i.e., EPS), was used as the target base. However, concerns emerged in regards to the possible adverse effects of these released EPS (O'Shea et al., 2014). Thus, the Australian Bureau of Meteorology (BoM) started to replace the EPS weather balloon bases with cardboard at the end of 2014 (Australian Marine Debris Initiative, 2018). As the transition happened, the number of weather balloon EPS pieces surveyed on beaches were observed to have significantly decreased from 2016 onwards as shown in Fig. S10. The present analytical results i.e., the accumulation of PAHs in weather balloon EPS, as well as in other EPS samples, validate previously expressed concerns regarding the utilization of EPS in weather balloons and its necessary replacement. The differences between the PAH compositions of new and beached EPS in these additional samples were also consistent with those observed in the Californian samples (Fig. S8), with the dominance of anthracene in new samples, and phenanthrene in beached samples.

In order to compare the results obtained in this study with those reported in existing literature,  $\Sigma 16$  PAHs and  $\Sigma 8$  PAHs were also calculated. In terms of  $\Sigma 16$  PAHs (Table 2), the con-

centrations in this study ranged 262–972 ng/g-EPS for new EPS and 312–10,468 ng/g-EPS for beached EPS. Apart from BPPC2, the range of  $\Sigma 16$  PAH concentrations in this study was similar to that reported by Van et al. (2012) with values ranging 240–1,700 ng/g-EPS in new EPS and 300–1,900 ng/g-EPS for beached EPS. Shi et al. (2020) also reported a similar range of  $\Sigma 16$  PAHs with beached EPS collected from Shantou Bay ranging 81–7,714 ng/g. Li et al. (2017) reported 8 PAHs with the lowest molecular weight among the 16 US EPA priority PAHs with a range of 311–534 ng/g and an average of 425 ng/g for PS food contact materials. Both NFPM and NFPC2 exhibited a similar range of concentrations with  $\Sigma 8$  PAHs of 446 and 448 ng/g-EPS respectively. Meanwhile NFPC1 was observed to have roughly double that concentration. It should be noted that there were minor differences between the selective PAH compounds analyzed in this study and those in the other studies referenced here. The PAH concentrations and compositions observed in the present study were consistent with those reported globally.

## CONCLUSION

New unexposed EPS contains pyrogenic PAHs, especially anthracene, which are generated during the heating and manufacturing processes with concentrations reaching hundreds of ng/g-EPS. In addition, EPS sorbs PAHs from the surrounding environment, resulting in  $\Sigma$ 28 PAHs in beached EPS with values in the high hundreds up to 1,188 ng/g-EPS. This discovery is alarming as EPS can pose an immediate risk upon disposal into the marine environment and has the potential for chemical transfer into food products. Although this study focused on EPS from California, USA, the data were consistent with additional samples from Japan, Malaysia and Australia, as well as with the results of previous studies. This demonstrated that the problems associated with EPS constitute a global issue.

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## SUPPLEMENTARY MATERIAL

Fig. S1, New unexposed EPS packaging block—NPBC (left) and NPBJ (right); Fig. S2, New unexposed EPS food packaging—NFPC (left) and NFPM (right); Fig. S3, Beached EPS packaging block—BPBC1 (left) and BPBC2 (right); Fig. S4, Beached EPS food packaging—BFPC1 (left) and BFPC2 (right); Fig. S5, Beached EPS food packaging—BFPC3 (left) and BFPC4 (right); Fig. S6, Beached EPS packaging peanut—BPPC1 (left) and BPPC2 (right); Fig. S7, Beached EPS weather balloon—BWBA; Fig. S8, PAHs composition in new unexposed and beached EPS samples; Fig. S9, Alkylated PAHs to parent PAHs ratios for source determination in EPS samples; Fig. S10, Survey of EPS weather balloon pieces collected on beaches along the coast of Australia. Data obtained from the Australian Marine Debris Initiative Database courtesy of Tangaroa Blue Foundation.

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