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► To cite this version:

R. Bouhroum, A. Boulkamh, L. Asia, S. Lebarillier, Alexandra ter Halle, et al.. Concentrations and fingerprints of PAHs and PCBs adsorbed onto marine plastic debris from the Indonesian Cilacap coast and the North Atlantic gyre. *Regional Studies in Marine Science*, 2019, 29, pp.100611. 10.1016/j.rsma.2019.100611 . hal-02194342

HAL Id: hal-02194342

<https://hal.science/hal-02194342v1>

Submitted on 14 Feb 2020

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Concentrations and fingerprints of PAHs and PCBs adsorbed onto marine plastic debris from the Indonesian Cilacap coast and the North Atlantic gyre

BOUHHROUM R.¹, BOULKAMH A.¹, ASIA L.², LEBARILLIER S.², TER HALLE A.³, SYAKTI A.D.^{4,5}, DOUMENQ P.², MALLERET L.², WONG-WAH-CHUNG P.^{2*}

¹Laboratoire des Techniques Innovantes de Préservation de l'Environnement, Université Frères Mentouri, Constantine 1, Algérie

²Aix Marseille Univ, CNRS, LCE, Marseille, France

³Laboratoire des IMRCP, Université de Toulouse, CNRS UMR 5623, Université Paul Sabatier, 118 route de Narbonne 31062 Toulouse Cedex 9, France

⁴Center for Maritime Biosciences Studies – Institute for Sciences and Community Service, Fisheries and Marine Science Faculty, Jenderal Soedirman University, Kampus Karangwangkal, Jl. dr. Suparno, Purwokerto 53123, Indonesia

⁵Marine Science and Fisheries Faculty - Raja Ali Haji Maritime University, Jl. Politeknik Senggarang-Tanjungpinang, Riau Islands Province, 29100, Indonesia

*corresponding author

E-mail: pascal.wong-wah-chung@univ-amu.fr

Abstract

We investigated the contamination of marine plastic debris from the Indonesian Cilacap coast and the North Atlantic gyre by polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs). The mean concentrations of 15 PAHs in gyre were 58 and 142 ng/g in polyethylene (PE) and polyethylene terephthalate (PET) debris, respectively, while PE and polypropylene (PP) Indonesian debris exhibited an average content of 552 ng/g (mainly naphthalene). The mean concentration of 61 PCBs in open ocean debris was 12.2 ng/g, while coastal debris showed a worryingly high mean value of 1.4×10^4 ng/g, with the notable presence of CB6, 101 and 173. PE gyre debris contained mostly high molecular weight PAHs such as benzo(g,h,i)perylene and indeno(1,2,3-cd)pyrene and low-chlorinated PCBs such as CB52, 77 and 105, whereas PET debris contained low molecular weight PAHs (mainly acenaphthylene and phenanthrene) and high-chlorinated PCBs such as CB204.

Highlights

- First results on the pollution of plastic debris from Indonesian coast and North Atlantic gyre
- The cumulative PAHs and PCBs' concentration is 91 ng/g in North Atlantic gyre debris.
- Indonesian coastal debris are 150 times more polluted than North Atlantic gyre ones
- PAHs' concentrations in PET from the North Atlantic gyre are slightly higher than in PE
- Fingerprints of PAHs and PCBs in marine plastic contrasted according to the microplastic source.

Keywords: Marine plastic debris, PAHs, PCBs, Indonesian Cilacap coast, North Atlantic gyre

1. Introduction

The accumulation of plastic debris in natural aquatic compartments is recognized as a major environmental problem because of their high amounts, their persistence and the limited knowledge available regarding their impact on ecosystems. This form of pollution has been

52 evidenced in oceans as well as in seas, rivers and lakes (Andrady, 2011; Ivar do Sul et al.,
53 2014; Li et al., 2016; Moore, 2008; Syakti et al., 2017; Ter Halle et al., 2017; Van Sebile et al.,
54 2015;).

55 It is widely accepted that plastic debris at the surface of seas and oceans, especially
56 macroplastics, are often responsible for the death of large marine predators by accidental
57 capture or the obstruction of the digestive system (Lavers et al. 2014; Li et al., 2016). More
58 recent studies suggest that this adverse physical effect may also affect smaller marine organisms
59 such as mussels and copepods (Cole et al., 2013; Van Cauwenberghe et al., 2014) or fish and
60 bivalves sold for human consumption (Rochman et al., 2013a; Rochman et al., 2015). These
61 results underline the potential impact of microplastics on tiny animal organisms that could be
62 damaging to marine ecosystems.

63 Moreover, investigation of the potential impact of plastic debris on marine fauna is progressing
64 through the detection in polymer debris of chemical compounds (persistent organic pollutants
65 (POPs), additives) that are often toxic or having a chronic effect (Hirai et al., 2011; Ogata et
66 al., 2009; Rios et al., 2007; Teuten et al., 2009). The presence of various chemicals raises the
67 question of their potential impact on the ecosystem through diffusion in water and transfer in
68 the aquatic species after ingestion. This aspect is of great concern, and recent studies have
69 revealed the presence of hydrophobic organic pollutants traces in marine organisms in regions
70 severely polluted by plastic debris (Gassel et al., 2013; Ryan, 1988; Tanaka et al., 2013;
71 Yamashita et al., 2013). Nevertheless, scientific knowledge regarding the potential
72 environmental impact is still sparse.

73 However, even if many studies have focused on the chemical content of plastic marine debris
74 collected in different aquatic compartment areas, the nature and concentrations of detected
75 compounds show very high variability. For instance, many studies have evidenced a proven
76 presence of PCBs and PAHs in plastics debris (PE and PP) sampled from different locations,
77 with concentrations in a range as broad as 1 to 10,000 ng/g of plastic debris (Hirai et al., 2011;
78 Ivar do Sul et al., 2014; Teuten et al., 2009). But hitherto, no consistent hypothesis has been
79 proposed to explain the considerable heterogeneity of the results.

80 The aim of the present paper is to broaden our knowledge on the distribution pattern and
81 concentration variability of POPs (more specifically PAHs and PCBs) by measuring their
82 concentrations in floating plastic debris, the latter being collected from inshore waters (Cilacap
83 coast, Indonesia) and remote offshore waters (North-Atlantic gyre), respectively, and thus to
84 contribute to fill the gap.

85

86 **2. Materials and methods**

87 *2.1. Study area and plastic debris sampling*

88 Macroplastics (at least one size greater than 20 mm) were collected by boat in the North Atlantic
89 subtropical gyre (NAG) in June 2015 during the French Expedition 7ème Continent. The plastic
90 debris was collected by the sailing vessel Guyavoile in the accumulation area determined by
91 studying a surface net tow data set (Law et al., 2010). The macroplastics were collected using
92 a telescopic pole. Fifteen macroplastics were collected in June 2015 and the physico-chemical
93 characteristics of some of them were given in a previous publication (Ter Halle et al., 2017). A
94 Manta net of 75 cm × 20 cm with a mesh of 300 µm was used to sample plastic debris, from
95 micro to macro size, along the Indonesian Cilacap Coast (ICC) (Syakti et al., 2017). The Cilacap
96 region is located on the south coast of Java (Indonesia, Indian Ocean) and is surrounded by an
97 area of slough, tributaries, mangrove swamps and intertidal land converted into rice fields. The
98 Manta net was towed by a small boat, moving along 1.8 km of an imaginary transect line on
99 the Cilacap coast, at a speed of 1.9 knots. Five distant marine areas (MT-1 to MT-5) were
100 marked out for sampling. The collected plastic debris were sorted out from the filtered water
101 with tweezers under a binocular microscope and stored in frozen glass sealed vials (-5°C) or at

102 room temperature to avoid any pre-analytical contamination. Site GPS coordinates are given in
103 Table S1 of the Supplementary Material (SM).

104

105 *2.2. Chemicals and reagents*

106 Water and acetonitrile (CH₃CN) for liquid chromatography analysis were Optima® LC-MS
107 grade provided by Fisher Chemical SAS. Hexane and heptane were purchased by VWR (HPLC
108 grade), dichloromethane by Merck (GC grade) and dimethylformamide solution of
109 EMPARTA® for Analysis American Chemical Society with analytical purity of 99.0-99.5 %.
110 A stock PAHs standard mixture was obtained from Supelco (Sigma Aldrich). The stock mixture
111 contained: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene,
112 fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene,
113 benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g,h,i)perylene and
114 indeno(1,2,3-cd)pyrene at 10 µg/mL in CH₃CN.

115 Two stock PCB standard mixtures and standard solutions of individual PCB solutions were
116 used. PCB Mix 20 (CB28, 31, 52, 77, 101, 105, 118, 126, 128, 138, 153, 156, 169, 170 and
117 180), Mix 1 (CB1, 2, 3, 4, 6, 8, 9, 16, 18, 19, 22, 25, 28, 44, 52, 56, 66, 67, 71, 74, 82, 87, 99,
118 110, 138, 146, 147, 153, 173, 174, 177, 179, 180, 187, 194, 195, 199, 203 and 206), individual
119 solutions of CB10, 14, 18, 29, 30, 78, 104, 105, 145, 149, 155, 204 and 209, and Mirex (used
120 as an internal standard for GC-MS analysis), all at 10 ng/µL in cyclohexane, were purchased
121 by Dr. Ehrenstorfer and Accustandard.

122

123 *2.3. Plastic polymer identification*

124 The collected plastic debris were identified by Fourier Transform Infra-Red spectroscopy
125 (FTIR), on a Thermo Electron Nexus spectrophotometer equipped with a diamond crystal Smart
126 Orbit TM accessory. Spectra were recorded in Attenuated Total Reflection (ATR) mode and
127 corrected by ATR correction. Spectra were acquired in the range of 4000-450 cm⁻¹, with 64
128 scans and a resolution of 4 cm⁻¹. After identification using a spectrum database and considering
129 certainty above 90%, debris were classified by type of polymer: PE, PP, PET, polyvinyl
130 chloride (PVC) and polystyrene (PS). Carbonyl index (CI) was also determined for PE and PP
131 plastic debris by FTIR analysis, as described and presented elsewhere (Ter Halle et al., 2017;
132 Syakti et al., 2017).

133

134 *2.4. Extraction of PAHs and PCBs from plastic debris*

135 Extraction from polymers (around 0.1 g) was achieved with a 10 mL portion of a mixture of
136 dichloromethane: heptane 1:1 (v:v). The suspension was maintained under mechanical stirring
137 for 24 hours. This procedure was repeated in the same manner with a fresh 10 mL of the solvent
138 mixture. A third extraction was performed to check that no residual traces of PAHs and PCBs
139 were left as previously proposed by Choi et al., 2013. Extracts were slowly evaporated to
140 dryness under a gentle nitrogen stream and the residue was then dissolved in 250 µL of hexane
141 containing the internal standard (Mirex), before GC-MS analysis. For liquid chromatography
142 analyses, about 0.5 mL was left during the evaporation step and 50 µL of dimethylformamide
143 was added and the solution was evaporated once more. Finally, 150 µL of acetonitrile was added
144 to the residue to reach a total volume of extract of 200 µL, and the resulting solution was filtered
145 on 0.2 µm PTFE filter (Chromacol 4-SF-02(T), Thermo Scientific), before injection. This
146 extraction procedure was carried out for each macroplastic debris collected in NAG and
147 microplastic debris pooled by area in the case of ICC. Further details regarding the extraction
148 method can be found in the SM.

149

150

151

152 2.5. Analysis of PAHs and PCBs

153 Analyses with an Ultra Performance Liquid Chromatography (UPLC Perkin Elmer, Altus 30)
154 equipped with a fluorescence detector were undertaken for determination of PAH
155 concentration. An Agilent Zorbax Eclipse PAH column (1.8 μm , 150 \times 2.1 mm i.d.) placed at
156 40 °C with a flow rate of 0.35 mL/min was used and the injected volume was 2 μL . The gradient
157 used is initially 40% A (CH_3CN) and 60% B (H_2O), after 15 minutes the gradient changes to
158 100% A, and at 23 min it returns to the initial conditions. A four-channel method with different
159 excitation and emission wavelength was developed to detect and quantify all PAHs at low
160 concentration levels (LOQ of 1 to 6.5 ng/g of polymer) except acenaphthylene (Wong-Wah-
161 Chung et al., 2018).

162 A gas chromatography mass spectrometer (GC-MS, Perkin Elmer, Clarus 600 / 600C) equipped
163 with a programmable temperature vaporizing (PTV) injector (programmed from 50 to 250°C
164 at 200°C min^{-1}) enabled the identification and quantification of the above-listed 61 PCBs,
165 except PCB 28 and 31 which were co-eluted. A DB-5MS column (30 m \times 0.25 mm \times 0.25 μm)
166 was used and 1 μL was injected in splitless mode. A specific temperature program from 60 to
167 280°C was developed to separate all PCBs: 60°C for 1 min, heated to 150°C at 40°C/min, then
168 to 280°C at 6°C/min and held 3 min. Source temperature was fixed at 250°C and compounds
169 ionization was performed by electron ionization (70 eV). The quadrupole mass spectrometer
170 operated in simultaneous collection of full scan and selected ion recording (Selected Ion Full
171 Ion mode or SIFI mode). Two GC-MS methods were developed with specific SIFI detection
172 program to analyse 39 PCBs (using Mix 1 as reference) and other 22 PCBs (using Mix 20 and
173 individual PCBs as reference). LOQ for PCB range from 0.08 to 4.3 ng/g of polymer. QA/QC
174 procedures for chemical analysis can be found in the SM.

175

176 3. Results and discussion

177 3.1. Plastic debris characterization

178 Indonesian samples are micro-debris sizing a few millimeters in length and with a total mass of
179 1.3 g, as described previously (Syakti et al., 2017). Infrared results showed that the 146
180 Indonesian pieces are made up mainly of PE (71% w/w), PP (27% w/w) and PS (two pieces,
181 i.e. a little less than 2% w/w).

182 North Atlantic gyre samples contained 15 macroplastics each of them measuring more than a
183 few centimeters. Different types of polymer were identified: PE (sample NA-1 to NA-9), PET
184 (sample N-10 to N-12), PS and PVC. Samples N-10 and N-12 correspond to floating closed
185 bottles and the PVC sample is a yellow foam float. Herein are exclusively presented the results
186 obtained on the most abundant plastic debris, i.e. PE and PET for gyre debris (individually
187 analysed), and PE and PP for ICC debris (pooled from Manta net samples before analysis).

188

189 3.2. Determination of PAHs

190 Concentrations of individual PAH and average concentrations of 15 PAHs in marine plastic
191 samples collected in the North Atlantic Gyre and along the Indonesian coast are presented in
192 Tables 1 and 2, respectively. Only quantifiable values are shown in the Tables. For NAG
193 samples, the average concentration (\bar{C}) of a congener was calculated as the total concentration
194 of this congener divided by the number of plastic debris (n) of the same chemical nature,
195 considering null any concentration below its quantifiable limit. For ICC samples, a mean, which
196 takes into account the mass of debris collected at each site, was calculated for any single
197 congener. Thus, the weight-average of a given congener was calculated by dividing the total
198 mass of this congener by the total mass of the polyolefin debris collected throughout the five
199 sites (MT-1 to MT-5). The overall mean concentration was calculated in a similar way. It is
200 equal to the number-average concentration of 15 PAHs contained in NAG-PE or NAG-PET
201 and the mass-average concentration of 15 PAHs contained in pooled ICC-samples.

Table 1
Concentrations of PAHs in ng per gram in North Atlantic gyre plastic debris.

PAH (in ng/g)	PE									\bar{C} (n = 9)	PET			\bar{C} (n = 3)
	NA-1	NA-2	NA-3	NA-4	NA-5	NA-6	NA-7	NA-8	NA-9		NA-10	NA-11	NA-12	
Naphthalene	-	-	2.19	-	-	4.73	-	-	-	0.77	15.4	5.98	1.70	7.71
Acenaphthylene	0.33	0.21	0.32	-	-	0.44	-	0.25	0.2	0.19	4.30	103	138	82.0
Fluorene	0.89	0.63	1.57	1.09	-	2.84	-	1.41	-	0.94	9.78	0.67	0.61	3.69
Phenanthrene	4.87	5.02	4.12	4.61	5.9	7.15	1.86	5.53	6.8	5.10	75.5	1.48	0.92	26.0
Anthracene	1.37	-	-	-	-	-	-	-	-	0.15	-	-	-	-
Fluoranthene	1.17	0.76	0.54	5.17	2.62	0.93	-	0.78	-	1.33	32.6	-	-	10.9
Pyrene	1.82	1.74	1.07	15.0	7.16	1.07	3.16	1.4	2.4	3.87	21.4	-	-	7.15
Benzo(a)anthracene	0.75	-	-	33.1	1.38	-	-	-	-	3.91	4.59	-	-	1.53
Chrysene	-	-	-	20.8	-	1.52	-	-	-	2.48	6.64	-	-	2.21
Benzo(b)fluoranthene	0.63	0.49	0.49	14.0	3.15	10.7	2.17	0.52	-	3.57	1.96	-	-	0.65
Benzo(k)fluoranthene	0.47	0.43	0.42	2.74	2.61	2.43	1.82	0.43	-	1.26	-	-	-	-
Benzo(a)pyrene	2.98	2.74	2.71	7.05	16.8	6.17	11.9	2.76	-	5.90	-	-	-	-
Dibenzo(a,h)anthracene	-	-	-	2.11	-	4.34	-	-	-	0.72	-	-	-	-
Benzo(g,h,i)perylene	1.4	1.27	1.26	10.6	7.82	115	5.45	1.28	-	16.0	-	-	-	-
Indeno(1,2,3-cd)pyrene	0.86	0.48	2.19	-	-	98.0	-	0.94	1.8	11.6	-	-	-	-
Σ PAHs	17.5	13.8	16.9	116	47.4	255	26.3	15.3	11.2		172	111	142	
\bar{C} (n = 9 or 3)		58.0									142			
Global Mean Concentration:	78.7													

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3.2.1. PAHs in North Atlantic gyre (NAG) plastic debris

204 As shown in Table 1, several PAHs were detected in all PE samples collected in NAG (NA-1
205 to NA-9). From four to thirteen PAHs were identified in each piece of PE. The presence of
206 phenanthrene and pyrene was always confirmed at concentrations of about 1 to 15 ng/g.
207 Indeno(1,2,3-cd)pyrene and benzo(g,h,i)perylene were both detected in samples NA-1 to NA-
208 3, NA-6 and NA-8 at concentrations of around 0.5 to 98 ng/g and 1.4 to 115 ng/g, respectively.
209 A remarkably high level of the two previous PAHs can be noted in sample NA-6.
210 Concentrations of each PAH differ by up to three orders of magnitude, from 0.2 to 115 ng/g,
211 while total PAHs (Σ 15PAHs) ranged from 11.2 to 255 ng/g, with an average concentration of
212 $\bar{C} = 58$ ng/g. Such levels of concentration in total PAHs agree with previous studies carried
213 out on PE and PP plastic debris collected in the North Pacific central gyre (Hirai et al., 2011).
214 PAHs were also detected in PET debris (NA-10 to NA-12), with the systematic presence of
215 naphthalene, acenaphthylene, fluorene and phenanthrene. Five more PAHs were identified but
216 only in NA-10, namely fluoranthene, pyrene, benzo(a)anthracene, chrysene and
217 benzo(b)fluoranthene. NA-10 is clearly different from the other two samples in terms of PAHs
218 levels and pattern. The concentrations of individual PAH were also distributed over a wide
219 range, i.e. 0.6 to 138 ng/g. Total PAHs extracted from PET debris amounted from 111 ng/g for
220 NA-11 to 172 ng/g for NA-10, which is similar to the highest concentrations measured in PE
221 NAG samples. The PAHs concentrations in the three PET samples averaged 142 ng/g.
222 The comparison of the amounts of each PAH in one or the other gyre polymers shows that
223 individual PAHs are unequally distributed between PE and PET. The mean concentrations are
224 notably greater in PET than in PE for naphthalene (7.7 vs 0.8 ng/g), acenaphthene (82 vs 0.2
225 ng/g), fluorene (3.7 vs 0.9 ng/g), phenanthrene (26 vs 5.1 ng/g), fluoranthene (10.9 vs 1.3 ng/g)
226 and pyrene (7.2 vs 3.9 ng/g).
227 When comparing the overall concentrations of PAHs in the two types of polymer, it is found
228 that the average amount of PAHs adsorbed onto PET debris is 2.5 times larger than PE (142 vs
229 58 ng/g, respectively).
230

231

3.2.2. PAHs in Indonesian Cilacap coastal (ICC) plastic debris

232 From Table 2, it can be seen that PAHs were detected in all five Manta net samples collected
233 from the coast. On the one hand, ten to thirteen PAHs were identified in each sample, nine of
234 which were systematically evidenced (naphthalene, fluorene, pyrene, fluoranthene, pyrene,
235 benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene and benzo(g,h,i)perylene). On
236

237 the other hand, anthracene and benzo(a)anthracene were not detected in any of the Indonesian
 238 samples. The concentrations of each PAH showed a very wide disparity, rising from 0.6 ng/g
 239 for the lowest one (chrysene in MT2), to 1.3×10^3 ng/g for the highest (naphthalene in MT-4).
 240 The mean concentrations of $\Sigma 15$ PAHs were 153 ng/g in MT-5 (the least contaminated sample)
 241 and 2.0×10^3 ng/g in MT-4 (the most contaminated one). This is in accordance with
 242 concentrations determined in plastic debris collected in the Japanese coast area and urban
 243 beaches (Hirai et al., 2011; Rios et al., 2014; Teuten et al., 2009). This behaviour can be
 244 attributed to the proximity of anthropogenic activities. This is especially patent for MT-4, which
 245 is close to boat transport and fishing. It is noteworthy that naphthalene was the most abundant
 246 PAH, its mean concentration (around 315 ng/g) counting for more than half of the overall mean
 247 PAH concentration (around 552 ng/g).
 248

Table 2
 Concentrations of PAHs in ng per gram in Indonesian Cilacap coast plastic debris.

PAH (in ng/g)	MT-1	MT-2	MT-3	MT-4	MT-5	\bar{C} (n=5)
Naphthalene	421	162	83.4	1.26×10^3	72.6	315
Acenaphthylene	-	3.86	-	-	-	0.26
Fluorene	43.6	3.75	2.23	23.2	4.07	11.2
Phenanthrene	45.4	32.3	10.6	147	27.4	47.6
Anthracene	-	-	-	-	-	-
Fluoranthene	45.1	12.7	16.7	163	11.9	41.2
Pyrene	62.6	11.2	17.6	148	10.9	40.0
Benzo(a)anthracene	-	-	-	-	-	-
Chrysene	22.6	0.60	8.69	-	-	3.41
Benzo(b)fluoranthene	-	5.05	-	-	4.90	3.02
Benzo(k)fluoranthene	20.7	4.64	3.31	17.9	2.48	7.21
Benzo(a)pyrene	52.8	12.5	9.25	50.2	3.86	17.9
Dibenzo(a,h)anthracene	44.8	8.89	4.81	69.2	11.5	23.7
Benzo(g,h,i)perylene	72.1	19.9	9.26	58.8	3.90	21.9
Indeno(1,2,3-cd)pyrene	57.7	30.2	8.22	62.1	-	19.4
Σ PAHs	888	308	174	2×10^3	153	552

Global Mean Concentration: **552**

Note: Average concentrations (\bar{C}) are weighted arithmetic means, taking into account the mass of debris collected at each site.

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251 3.2.3.- Fingerprint of PAHs

252 Attention was paid to the distribution between heavy molecular weight (more than three fused
 253 aromatic rings) and low molecular weight (2-3 fused aromatic rings) PAHs (H- and L-PAHs,
 254 respectively), as presented in Figs. 1 and 2. Percentages of H-PAHs (or L-PAHs) were
 255 calculated as the ratio between H-PAH (or L-PAH) concentration in ng/g divided by total PAH
 256 concentration in ng/g, multiplied by 100.
 257

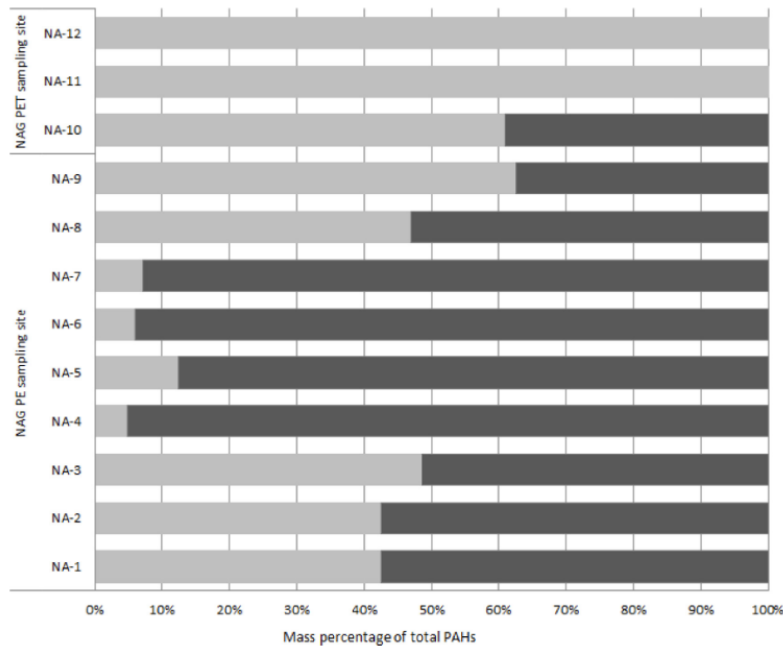


Fig. 1. Distribution of PAHs in percentage in North Atlantic gyre plastic debris. L-PAHs (grey), H-PAHs (black).

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This enabled us to evidence some singularities between all plastic debris. On one hand, it appeared that NAG PE samples mainly contain H-PAHs with concentration percentages in the range of 37 (NA-9) to 95% (NA-4), linked to the presence of markedly higher amounts of benzo(g,h,i)perylene and indeno(1,2,3-cd)pyrene (see Table 1). On the other hand, PET NAG debris were mainly polluted by L-PAHs (from 61 (NA-10) to 100% (NA-11 and NA-12)) as well as ICC plastic debris (from 55 (MT-3) to 72 % (MT-4)).

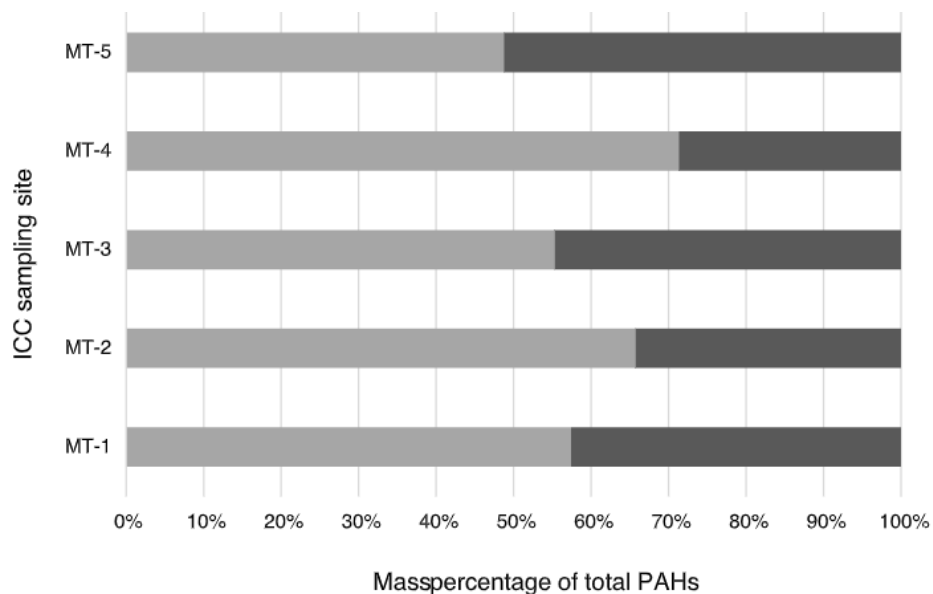


Fig. 2. Distribution of PAHs in percentage in Indonesian Cilacap coast plastic debris. L-PAHs (grey), H-PAHs (black).

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Acenaphthylene and phenanthrene account for the highest concentrations in NAG PET debris (Table 1), and naphthalene and phenanthrene in coastal debris (Table 2).

274 *3.3. Determination of PCBs*

275 In this part, the research focused on 61 PCBs congeners, among which 17 were detected in
 276 NAG samples and 31 in ICC samples (Tables 3 and 4). Only PCBs that were present in a
 277 quantifiable amount are reported in these tables.

278
 279 *3.3.1. PCBs in North Atlantic gyre debris*

280 According to Table 3, PCBs were quantified in six of the nine PE samples (i.e. NA-1 to NA-3,
 281 NA-6, NA-8 and NA-9), while the presence of PCBs was confirmed in all three PET samples.
 282

Table 3
 Concentrations of PCBs in ng per gram in North Atlantic gyre plastic debris.

PCB (in ng/g)	PE							PET			
	NA-1	NA-2	NA-3	NA-6	NA-8	NA-9	\bar{C} (n=9)	NA-10	NA-11	NA-12	\bar{C} (n=3)
CB1	-	-	-	-	-	14.9	1.66	-	-	-	-
CB2	0.59	-	-	-	-	-	0.07	-	-	-	-
CB10	-	-	3.47	-	-	-	0.38	0.72	1.31	-	0.68
CB18	-	-	-	-	-	-	-	-	0.13	-	0.04
CB19	-	-	-	-	-	4.3	0.48	-	-	-	-
CB22	-	-	-	-	-	8.5	0.94	-	1.38	-	0.46
CB28+31	-	-	-	-	-	-	-	0.54	0.94	1.08	0.85
CB30	-	1.73	-	-	0.77	-	0.27	-	-	-	-
CB52	-	-	11.0	50.4	-	-	6.82	-	-	-	-
CB77	-	-	-	-	-	10.2	1.13	-	-	-	-
CB78	-	-	-	-	2.52	-	0.28	-	-	-	-
CB87	-	-	-	-	-	12.9	1.43	1.44	2.42	-	1.29
CB105	-	-	1.24	1.43	-	7.9	1.17	-	-	-	-
CB156	-	-	-	-	-	-	-	-	2.59	-	0.86
CB174	-	4.68	-	-	-	-	0.52	-	-	-	-
CB204	-	-	-	-	-	-	-	-	15.1	-	5.02
\sum PCBs	0.59	6.41	15.7	51.8	3.29	58.7		2.7	23.8	1.08	
\bar{C} (n = 9 or 3)			15.2				9.21				

Global Mean Concentration: 12.2

283
 284
 285 1 to a maximum of 6 congeners were quantified in PE samples, and 2 to 3 congeners on average
 286 per sample. CB 105 was the most frequently detected (it was found in three samples). The
 287 concentrations of each PCB ranged from 0.6 to 50 ng/g, while concentrations of Σ PCBs ranged
 288 from 0.6 (NA-1, the least polluted sample) to 59 ng/g (NA-9, the most polluted sample). Similar
 289 concentrations were determined in plastic debris collected in the North Central Pacific gyre
 290 (Hirai et al., 2011; Rios et al., 2010), revealing some uniformity of PCB content in the plastics
 291 drifting in open oceans.

292 PCBs were also detected in PET debris (NA-10 to NA-12), with the systematic presence of CB
 293 28+31. Other PCBs were mainly identified in NA11, namely CB 10, 18, 22, 87, 156 and 204,
 294 which clearly distinguished this sample from the two others. In PET debris, PCB concentrations
 295 were within a very narrow range of concentrations from 0.5 to 2.6 ng/g, except for two values,
 296 i.e. 0.1 to 15.1 ng/g. Concentrations of Σ PCBs were of the same order of magnitude (from 1.1
 297 to 24 ng/g) as in PE debris.

298
 299 *3.3.2. PCBs in Indonesian coastal plastic debris*

300 PCBs were detected in all ICC samples, as shown in Table 4. CB173 was the most abundant
 301 PCB, its average concentration, i.e. 8.84×10^3 ng/g, counting for more than 63% of the average

302 concentration of total-PCBs. Other PCBs often detected in these samples were CB 6, 10, 29,
 303 77, 147 and particularly CB101, with a significantly high mean concentration (3.08×10^3 ng/g,
 304 representing about 22% of total-PCBs mean concentration). Individual CBs concentrations
 305 were in the range 0.9×10^3 to 1.6×10^4 ng/g, while total-PCBs concentrations extended from
 306 1.3×10^3 for MT-3 to 1.9×10^4 ng/g for MT-1. The mean concentration of Σ PCBs was 1.4×10^4
 307 ng/g. These worrying concentrations are many times higher than those cited in the literature for
 308 the coastal area and urban beaches (Frias et al., 2010; Hirai et al., 2011; Rios et al., 2014; Teuten
 309 et al., 2009; Zhang et al., 2015).
 310

Table 4
 Concentrations of PCBs in ng per gram in Indonesian Cilacap coast plastic debris.

PCB (in ng/g)	MT-1	MT-2	MT-3	MT-4	MT-5	\bar{C} (n=5)
CB1	–	–	42.0	208	2.18	40.6
CB3	7.45	–	–	–	–	0.79
CB4	–	–	–	1.87×10^3	364	511
CB6	10.6	–	55.2	261	214	1.68×10^3
CB10	–	–	–	1.02×10^3	–	171
CB16	–	2.29	44.3	–	6.37	8.69
CB18	–	48.6	0.94	72.3	74.9	56.4
CB22	–	–	36.9	404	–	71.6
CB25	–	–	58.5	–	–	6.67
CB28+31	–	8.97	–	–	82.8	45.9
CB29	1.61×10^3	–	28.6	–	45.1	198
CB44	–	1.83	–	–	–	0.12
CB52	–	3.02×10^3	–	–	–	201
CB56	–	12.6	–	–	16.6	9.92
CB66	–	16.0	–	37.4	–	7.30
CB67	21.7	–	–	113	–	21.2
CB74	–	2.79	–	167	–	28.1
CB77	928	–	126	492	100	249
CB82	256	–	–	–	–	27.0
CB101	1.55×10^4	131	508	2.59×10^3	1.72×10^3	3.08×10^3
CB104	256	–	–	–	–	27.1
CB138	–	14.4	–	–	–	0.96
CB146	2.13	–	–	–	–	0.22
CB147	–	3.66	305	929	106	248
CB149	–	–	19.3	–	–	2.20
CB153	14.6	–	32.9	–	28.5	20.9
CB173	42.2	56.9	–	1.57×10^3	1.56×10^4	8.84×10^3
CB177	19.7	–	–	–	–	2.08
CB179	–	17.5	18.2	–	–	3.24
CB180	–	38.6	–	–	–	2.57
Σ PCB	1.8×10^4	3.37×10^3	1.27×10^3	9.74×10^3	1.84×10^4	1.40×10^4

Global Mean Concentration: 1.40×10^4

Note: Average concentrations (\bar{C}) are weighted arithmetic means, taking into account the mass of debris collected at each site.

311
 312
 313
 314

315 3.3.3. Fingerprint of PCBs

316 The PCB distribution between high-chlorinated (≥ 5 chlorine atoms i.e. from CB82 to CB209)
 317 and low-chlorinated (≤ 4 chlorine atoms i.e. from CB1 to CB81) congeners (HC- and LC-PCBs,
 318 respectively) was then examined (Rodriguez et al., 2016). The distributions of PCB homologs
 319 are presented in Figs. 3 and 4. Fig. 3 evidences that PE NAG samples overwhelmingly contain
 320 LC-PCBs (from 64 (NA-9) to 100% (NA-1, NA-2 and NA-8)), more specifically tri- and tetra-
 321 chlorinated congeners for all samples, except for sample NA-1 which contains monochlorinated
 322 PCBs exclusively. For PET samples, it is hard to clearly identify a particular trend for the
 323 distribution of PCB homologs, even if HC-PCBs were predominantly detected with
 324 concentration percentages ranging from 53 (NA10) to 84% (NA-11).
 325

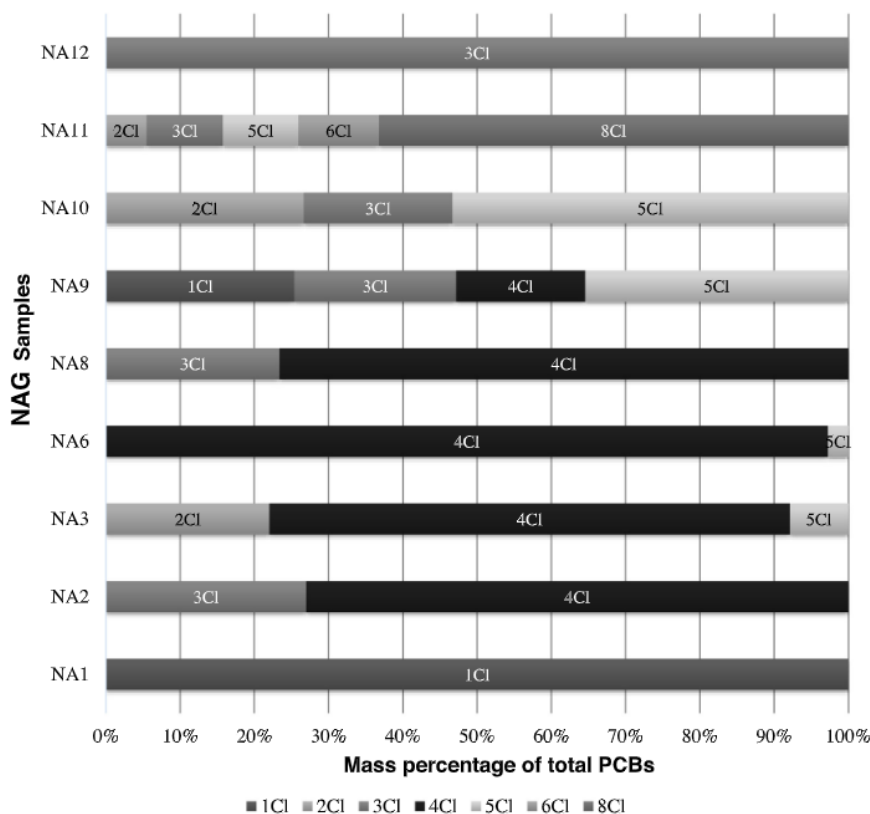


Fig. 3. Distribution of PCB homologs in percentage in North Atlantic gyre plastic debris. NA-1 to NA-9 (PE), NA-10 to NA-12 (PET).

326 However, Fig. 4 clearly shows that tetra-, penta-, hexa- and hepta-chlorinated congeners are the
 327 most abundant PCBs in all ICC samples. This trend can be correlated to the homologs pattern
 328 of Arochlor 1254 and 1260, Monsanto technical mixtures extensively marketed and used
 329 (Frame et al., 1996) with the predominant presence of HC-PCBs (52% (MT-4) to 95% (MT-
 330 5)).
 331
 332

333 **4. Discussion**

334 All marine plastic debris studied contained PAHs and PCBs. According to the sampling location
 335 of debris and to its polymeric nature, PAH and PCB content varied widely regarding: (i) the
 336 pattern, (ii) the individual concentrations, and (iii) the sum concentrations.

337 **1-** The difference between the relatively
 338 high PAH and PCB concentrations observed in coastal debris and the much lower
 339 concentrations observed in gyre debris may be explained by three parameters, the residential
 340 time in the marine environment, which is closely associated with the diffusion rate and the
 341 equilibrium sorption coefficients of POPs, the polymer size and the polymer structure (Choi et
 342

343 al., 2013; Frias et al., 2010; Hirai et al., 2010; Hüffer et al., 2018; Rios et al., 2007; Rios et al.,
 344 2010; Rochman et al., 2013b; Rusina et al., 2010; Zhang et al., 2015).
 345

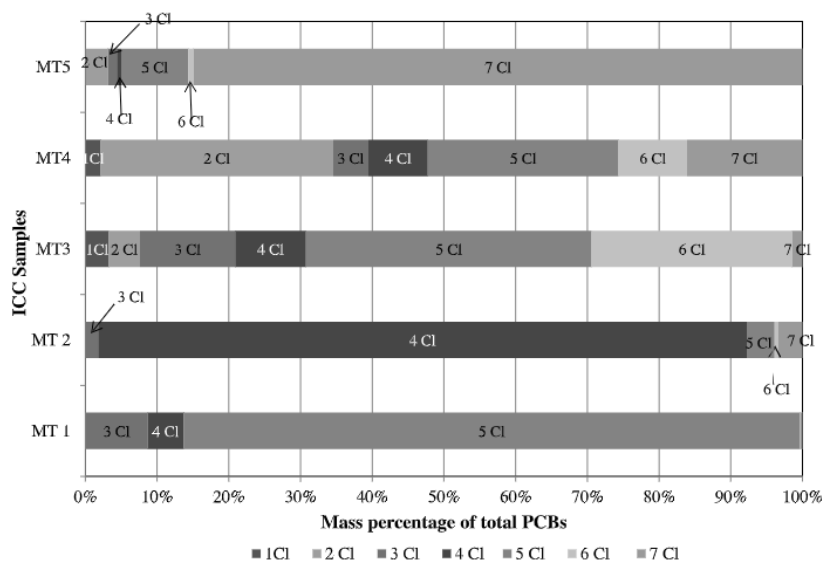


Fig. 4. Distribution of PCB homologs in percentage in Indonesian Cilacap coast plastic debris.

346
 347
 348 (i) As the plastic debris drift away towards remote oceans, plastics may undergo two processes
 349 yielding a decrease of contaminants adsorbed. On the one hand, this is actually expected to
 350 happen because plastic debris are much less exposed to contaminants, since the water of more
 351 remote locations gets gradually cleaner and much less affected by natural or anthropogenic
 352 pollution. Such desorption process is controlled by the diffusion. On the other hand, PAHs and
 353 PCBs are tied to plastics by physical hydrophobic interactions, which decrease due to the well-
 354 known photo oxidation of the polymeric matrix such as polyolefins (Gardette, 1995; Gardette
 355 et al., 2013). It is worth recalling that solar light exposure could be responsible for major
 356 structural changes in polyolefin polymers: formation of polar groups on chains, chain scission,
 357 crosslinking, cracking, erosion and fragmentation (Gardette, 1995; Gardette et al., 2013). Such
 358 changes could significantly alter the sorption properties of the polymer leading to the decrease
 359 in concentrations of PAHs and PCBs in the debris, as recently reported for PS microplastic
 360 particles (Hüffer et al., 2018). This hypothesis is supported by CI values of plastic debris that
 361 are often considered as representative of the photo oxidative state of the polymer: the higher
 362 the values are, the higher the polymer is photooxidized. CI values that were determined for
 363 NAG PE samples (around 0.7) are indeed ten times higher than those determined for ICC PE
 364 and PP samples (around 0.08), according to previous works (Syakti et al., 2017; Ter Halle et
 365 al., 2017). This assumption needs further experiments to confirm the sorption alterations of
 366 plastic debris for POPs under solar light exposure. Secondly, it is possible that some chemical,
 367 photochemical or microbial degradation of POPs takes place, decreasing their content in these
 368 plastics, as described in point 3.
 369 (ii) The microscopic size of coastal debris provides a surface area and an external shell volume
 370 that are, per unit of mass, much larger than that of gyre's macroscopic debris, greatly enhancing
 371 their adsorption capacity. Indeed, in a forthcoming work, we show that an external shell, which
 372 is less than 200 micrometers thick, constitutes the only volume that is effective for the uptake
 373 of POPs.
 374 (iii) The erosion process through sea transport also plays a significant role in the much lower
 375 pollution of NAG debris as compared to ICC debris because of the loss of the surface layer
 376 heavily contaminated with pollutants favoured by photochemical and mechanical processes.
 377

378 **2-** Another interesting difference, which was observed between polyolefin samples collected
379 from the coast and polyolefin samples collected from the open ocean, is the contrasting
380 distribution of the PCB content (LC and HC-PCBs) as well as PAH fractions (L- and H-PAHs).
381 HC-PCBs and L-PAHs predominate in the coastal samples, while LC-PCBs and H-PAHs are
382 prominent in the gyre samples. Even if plastic debris were collected from different ocean basins,
383 that could justify such disparity, PAH distribution suggests a petrogenic signature in coastal
384 debris and a pyrogenic one in gyre debris, as previously mentioned respectively for Japanese
385 coastal debris and Central Pacific Gyre samples (Hirai et al., 2011; Rios et al., 2012). In addition,
386 the predominance of HC-PCBs in coastal samples and of LC-PCBs in open ocean samples is
387 consistent with results reported by the same authors (Hirai et al., 2011; Rios et al., 2012).
388 The difference in distribution for PCBs may be explained by some sorption and
389 photodegradation processes. In fact, it has been reported that highly chlorinated congeners have
390 great affinity for PE-like polymers. The equilibrium sorption values (Choi et al., 2013) and the
391 diffusion coefficients of PCBs (Rusina et al., 2010) should favored HC-PCBs in open ocean
392 samples and LC-PCBs in coastal ones. Nevertheless, assuming that PCBs may undergo
393 sunlight-induced chemical degradation, some HC-PCBs could be converted to lower
394 chlorinated ones (Miao et al., 1999). Consequently, the proportion between LC-PCBs and HC-
395 PCBs is gradually inverted.

396 The higher percentage of H-PAHs in NAG PE samples could be justified by the higher affinity
397 between H-PAHs and PE (high equilibrium sorption coefficient) and the more efficient
398 diffusion of L-HAPs in PE (high diffusion coefficients): indeed, the PE-water partitioning
399 coefficient increases with PAH molar mass, as reported by Choi et al., 2013 while their
400 diffusion coefficient decreases (Fries et al., 2011; Rusina et al., 2010). Such a relationship,
401 associated with a long residence time in seawater (as is suggested by relatively high CI values),
402 could favour the accumulation of H-PAHs to the detriment of L-PAHs. This hypothesis is
403 consistent with the field experiments of Rochman and co-authors that highlighted the release
404 of L-PAHs and the continuous accumulation of H-PAHs (Rochman et al., 2013b). In line with
405 this interpretation is the low content of H-PAHs found in ICC plastic debris. The involvement
406 of any photochemical process allowing the transformation of L-PAHs to H-PAHs, must be
407 discarded since it has been shown that the photodegradation of PAHs on soil leads to oxidized
408 compounds (Marquès et al., 2017). However, the concentration decrease of L-PAHs in open
409 ocean debris could be ascribed to their more efficient phototransformation in comparison to H-
410 PAHs, as described previously on soils (Marquès et al., 2016). Such an assumption needs
411 further experiments on the behaviour of POPs on plastic debris for it to be confirmed.

412 **3-** Although of the same polymeric nature and from the same location, some samples displayed
413 drastically different contents. This cannot be justified by a probable difference in their
414 residential time (because of similar IC values), since it must be equivalent and hence should not
415 have such an impact. However, what happens to plastics before being sampled is completely
416 unknown. For instance, some NAG debris can be either transported from the American coasts
417 or from European ones, having thus initially different composition (additives, flame retardants,
418 etc.), and having then be in contact with different aquatic environments affected differently by
419 anthropogenic activities.

420 **4-** The significant qualitative and quantitative (i.e. congener compositions and concentrations)
421 differences between PE and PET samples, both issued from the gyre, can be attributed to the
422 different tendencies of these polymers to adsorb PAHs and PCBs, in addition to other processes
423 mentioned above. Rochman and coworkers (Rochman et al., 2013b) pointed out that PS
424 adsorbed low molecular weight PAHs more efficiently than PP. These results supported the
425 property of aromatic polymers, PET being one of them, to adsorb at their surface more
426 efficiently L-PAHs than do polyolefins, such as PE, through strong hydrophobic interactions
427 generated by the presence of π bonds in both compounds. Concerning the content of PCBs, it

428 seems reasonable to assume that similar interactions could also explain the difference between
429 PE and PET gyre samples.

430

431 **6. Conclusion**

432 The study of PAHs and PCBs contained in plastic debris collected in different areas clearly
433 showed differences in concentration and distribution. Coastal debris was revealed to contain
434 high concentrations of PAHs and PCBs, with a predominance of low molecular weight PAHs
435 and high-chlorinated-PCBs, while gyre plastic debris are less polluted samples but contain
436 mainly high molecular weight PAHs and low-chlorinated-PCBs. This suggests the degradation
437 or release of chemicals during transport by ocean currents and/or atmospheric air, to which can
438 add the chemical and physical phototransformation of the plastic debris. It is worth recalling
439 that microplastics, and even more so, nanoplastics (particles smaller than 100 nm in size), may
440 be transported in air. Thus, the presence of highly toxic hydrophobic compounds was confirmed
441 in marine plastic debris from the Indonesian Cilacap coast, in huge concentrations, and the
442 North Atlantic gyre, in quite low concentrations. Acting as a sink of PAHs and PCBs, plastic
443 debris may pose threats to the marine ecosystem, by their potential to release these POPs into
444 either remote seawater or the organisms of aquatic animals, after their ingestion. This pathway
445 adds up to other routes such as ventilation of water and food ingestion. In particular, the
446 presence of high molecular weight PAHs in plastic debris collected in remote oceans, such as
447 the carcinogenic benzo(a)pyrene, merits serious consideration from scientists and political
448 authorities. The high PCB concentrations in Indonesian plastic debris, especially CB52, 101
449 and 153, which are on the list of the European six indicators, is a matter of major concern
450 regarding the pollution levels caused by plastics in this part of the world (Jambeck et al., 2015).
451 The detection of CB77 and CB105 in plastic debris collected in the gyre is also of great concern,
452 because both are listed in the European Water Framework Directive (2013/39/EU) as priority
453 substances and are considered as dioxin-like compounds. In addition, the presence of LC-PCBs,
454 such as CB52 and 105, in NAG debris is worrying because of their possible accumulation in
455 marine organisms, as demonstrated by Yamashita et al., 2011.

456

457

458 **Acknowledgements**

459 We would like to express our gratitude to the Algerian government for Exceptional National
460 Program grant (PNE 198/2016-2017), Total Corporate Foundation, the Indonesia Program
461 DIPA UNSOED (DIPA/023.04.2.189899/2014), the French 7th Continent Expedition
462 association for debris sampling and PerkinElmer for providing their HPLC system.

463

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