# Persistent Organic Pollutants (PCBs and PAHs) adsorbed on plastic debris: distribution and concentration versus source

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**Abstract**

Marine plastic debris collected at the surface of Indian Ocean (Indonesian Cilacap coast) and North Atlantic gyre have respectively been studied. The concentration of 15 PAHs and 61 PCBs were evaluated using UPLC-fluorescence detector and GC-MS. PE debris from North Atlantic gyre contain PAHs in a concentration range 0.2-115 ng per gram of polymer while for Indonesian coast debris, concentrations were 17 times higher. PCBs have been detected in 60% of the debris from North Atlantic gyre with concentrations in the range 0.6-59 ng per gram of polymer while coastal debris show concentrations around 300 times higher. Gyre debris appear to contain higher proportion of heavy PAHs (maximum of 95%) and tetra chlorinated PCBs *i.e.* LC-PCBs. Coastal debris mainly contain light PAHs (maximum of 71%) but also HC-PCBs. This results clearly proved the spatial distribution and concentration of PAHs and PCBs dependency of marine area compartments (open ocean versus coast)**.**

**Keywords:** Plastic debris, PAHs, PCBs, spatial distribution, marine coast, gyre

## 1. Introduction

Accumulation of plastic debris in natural aquatic compartments is recognized as a major environmental problem because of high amount of plastic debris, persistence and limited knowledge of their impacts on ecosystems. This pollution has been demonstrated in oceans as well as in seas, rivers and lakes (Ivar do Sul 2014, Li 2016). It is well-admitted that plastic debris at the surface of seas and oceans, especially macroplastics, are often responsible for the death of large marine predators by accidental capture or obstruction of digestive system (Li 2016; Lavers 2014). However, more recent studies suggest that this adverse physical effect may affect smaller marine organisms such as molds and copepods (Cole 2013, Van Cauwenberghe 2014). These results underline the potential impact of microplastics on tiny animal organisms that could be damaging to marine ecosystems. Moreover, the assessment of plastic debris effects on marine fauna is progressing through the detection in polymer debris of chemical compounds (hydrophobic persistent organic pollutants (POPs), additives) often toxic or having chronic effect (Rios 2014, Hirai 2011, Teuten 2009). The presence of various chemicals raises the question of their potential impact by diffusion in water and consequently in biota. This aspect is of great concern and recent studies demonstrated hydrophobic organic pollutants traces in marine organisms in region highly polluted by plastic debris (Gassel 2013, Tanaka 2013, Yamashita 2011). Nevertheless, scientific knowledge about potential environmental consequences are yet largely unknown. However, even if many studies focused on the chemical content of plastic debris collected in different aquatic compartment areas, the nature and concentration of detected compounds showed a very high variability. For instance, many research studies pointed out a proven presence of PCBs and PAHs in plastics debris (polyethylene (PE) and polypropylene (PP)) sampled in different locations with concentrations in the range 1 to 10,000 ng per plastic debris gram (Ivar do Sul 2014, Rios 2014, Hirai 2011, Teuten 2009). Even if many results on plastic debris chemical content are available in the literature, no consistent hypothesis has hitherto been set to explain the disparate results.In this study, we are aiming to gain deeper knowledge on spatial distribution and concentration of POPs in plastic debris based on the determination of PAHs and PCBs concentration in coastal and gyre plastic debris to close these gaps of knowledge.

## 2. Sampling conditions

Manta net of 75 cm × 20 cm and a mesh of 300 μm was used to sample plastic debris from micro to macro size in Indian Ocean (Indonesian Cilacap Coast (ICC)). In North Atlantic gyre (NAG), macro plastic samples were collected using a telescopic pole. Site GPS coordinates are given in Table 1. Manta net samplings are indicated as MT and samples from North Atlantic gyre as NA. Samples were

stored at room temperature or at -10°C in glass flasks to avoid any further contamination.

**Table 1.** Sampling site details

| Sampling | Lat(S) | Long(E) |
|---|---|---|
| MT-1 | 7° 42' 18.25" | 109° 8' 23.93" |
| MT-2 | 7° 44' 32.41" | 109° 5' 50.55" |
| MT-3 | 7° 44' 57.03" | 109° 1' 57.03" |
| MT-4 | 7° 42' 22.39" | 109° 4' 51.94" |
| MT-5 | 7° 44' 54.63" | 108° 59' 48.28" |
| | **Lat(N)** | **Long(W)** |
| NA-1 | 26° 38' 12.70" | 58° 22' 22.87" |
| NA-2 | 26° 28' 46.78" | 58° 30' 1.91" |
| NA-3 | 25° 21' 12.00" | 59° 22' 37.28" |
| NA-4 | 26° 22' 58.91" | 59° 12' 10.30" |
| NA-5 | 26° 33' 30.95" | 59° 00' 11.62" |
| NA-6 | 27° 19' 5.52" | 60° 30' 53.24" |
| NA-7 | | |
| NA-8 | 28° 00' 42.29" | 61° 02' 42.62" |
| NA-9 | 27° 59' 54.23" | 61° 02' 25.98" |

## 3. Material and Methods

### 3.1 Chemicals

Water and acetonitrile ($CH_3CN$) for liquid chromatography analysis were Optima® LC-MS grade provided by Fisher Chemical SAS. Hexane and heptane were purchased by VWR (LC grade) and dichloromethane by Merck (GC grade). Stock PAHs standard mixture was obtained from Supelco (Sigma Aldrich): naphthalene (N), acenaphthylene (Acy), acenaphthene (Ace), fluorene (F), phenanthrene (P), anthracene (Ant), fluoranthene (Fl), pyrene (Pyr), benzo(a)anthracene (BaAnt), chrysene (Chr), benzo(b)fluoranthene (BbFl), benzo(k)fluoran-thene (BkFl), benzo(a)pyrene (BaPyr), dibenzo(a,h)an-thracene (DBahAnt), benzo(g,h,i)perylene (BghiPer) and indeno(1,2,3-cd)pyrene (Ipyr) at 10 $\mu g.mL^{-1}$ in $CH_3CN$. Stock PCBs standard mixtures were used and individual standard PCB solutions. PCB Mix 20 (PCB28, 31, 52, 77, 101, 105, 118, 126, 128, 138, 153, 156, 169, 170 and 180), individual solutions of PCB10, 14, 18, 29, 30, 78, 104, 105, 145, 149, 155, 156, 169, 204 and 209, Mirex, used as internal standard for GC-MS analysis, and Mix 1 (PCB1, 2, 3, 4, 6, 8, 9, 16, 18, 19, 22, 25, 28, 44, 52, 56, 66, 67, 71, 74, 82, 87, 99, 110, 138, 146, 147, 153, 173, 174, 177, 179, 180, 187, 194, 195, 199, 203 and 206) at 10 $ng.\mu L^{-1}$ in $CH_3CN$ were purchased by Dr. Ehrenstorfer and Accustandard.

### 3.2 Plastic polymer identification

Collected plastic debris have been identified using FTIR spectroscopy a Thermo Electron Nexus spectrometer equipped with a diamond crystal Smart Orbit™. Spectra were recorded in ATR mode and corrected. Spectra were acquired in the range 4000-450 $cm^{-1}$, with 64 scans, and a resolution of 4 $cm^{-1}$.

### 3.3 Extraction of Polymer

Solid liquid extraction method consisted in automatic stirring during 24 hours (two times), and the solvent was a mixture of 10 mL solutions of dichloromethane: heptane 1:1 (v:v). A third extraction was realized to control any traces of residual POPs. This has been developed for extraction from PE and PP polymers. Extract is slowly evaporated to dryness under gentle nitrogen stream and the residue dissolved in 250 μL of hexane containing internal standard (Mirex) before GC-MS analysis. For LC analysis, about 0.5 mL is left and 50 μL of dimethylformamide is added and the solution is evaporated once more. Finally, 150 μL of acetonitrile is adding and filtrating on 0.2 μm PTFE filter (Chromacol 4-SF-02(T), Thermo Scientific) before injection.

### 3.4 Analysis of PAHs and PCBs

Analyses with an Ultra Performance Liquid Chromatography (UPLC Perkin Elmer, Altus 30) equipped with a fluorescence detector were undertaken for PAHs concentration determination. A Agilent Zorbax Eclipse PAH column (1.8 μm, 150 × 2.1 mm i.d.), a flow rate of 0.35 $mL.min^{-1}$ were used and the injected volume was 2 μL. A four channels method with different excitation and emission wavelength was developed to detect and quantify all PAHs at low concentration levels (0.5 to 10 $\mu g.L^{-1}$) except Acy. A gas chromatography-mass spectrometer (GC-MS, Perkin Elmer, Clarus 600/600C) equipped with a split/splitless PTV injector (from 50 to 250°C at 200°C.$min^{-1}$) allowed 61 PBCs identification and quantification. A DB-5MS column (30 m × 0.25 mm × 0.25 μm) was used and a specific temperature program from 60 to 280°C was developed to separate all PCBs. Electronic impact (70 eV) allowed ionization and quadrupole mass spectrometer, the detection of PCB congeners in Single Ion Monitoring mode.

## 4. Results and discussion

### 4.1 Plastic debris characterization

IR spectroscopy analyses showed that ICC plastic samples are mainly PE, and PP and in a lesser extent PS. On the other hand, PE is the polymer mostly present among NAG macroplastic collected samples (9/15). Herein are exclusively presented the results obtained on PE gyre debris, and PE and PP ICC debris were grouped by manta net before analysis.
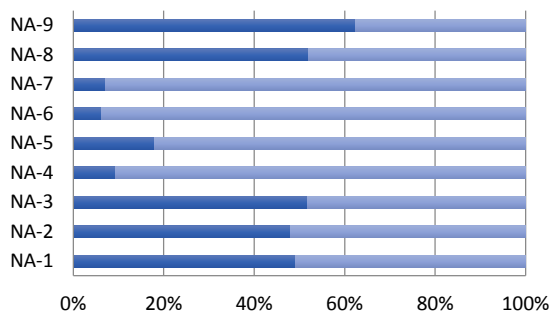
### 4.2 Determination of PAHs

Individual PAH and PCB concentrations in collected samples are presented in Table 2 and 3. As shown in Table 2, PAHs were detected in all PE samples collected in NAG and 4 to 13 PAHs were identified in each sample. The presence of P and Pyr was always confirmed while InP and BghiPer were detected in 3 and 8 samples respectively. Individual PAHs concentration was in the range 0.2 to 115 $ng.g^{-1}$ while total PAHs concentrations revealed to be in the range 11.2 to 255 $ng.g^{-1}$ in accordance with similar concentration determined in plastic debris collected in North Pacific central gyre (Hirai 2011). Moreover, the distribution between heavy (more than four fused aromatic rings) and light (up to than four fused aromatic rings) PAHs (H-PAHs and L-PAHs, respectively) put in evidence that NAG samples mainly contains H-PAHs with mass percentages in the range 37 to 95% (Figure 1). PAHs were detected in all manta net samples collected in ICC as shown in Table 3 and 10 to 13 PAHs were identified in these samples. However, Ant and BaAnt were undetected in Indonesian samples (not shown) while many PAHs were

systematically evidenced (N, F, P, Fl, Pyr, BkFl, BaPyr, DBahAnt and BghiPer). Individual PAH concentration was in the range 0.6 to 1,262.4 ng.g$^{-1}$, naphthalene being the most abundant PAH. Total PAHs concentration revealed to be in the range 153.5 to 2,002.8 ng.g$^{-1}$ in accordance with similar concentration determined in plastic debris collected in Japanese coast area and urban beaches (Rios 2014, Hirai 2011, Teuten 2009). Figure 2 shows that ICC samples contains more L-PAHs than H-PAHs ( > 62%).

**Table 2.** PAHs concentration in ng per NAG plastic debris gram

| NA | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 |
|---|---|---|---|---|---|---|---|---|---|
| N | - | - | 2.29 | - | - | 4.73 | - | - | - |
| Ace | 0.33 | 0.21 | 0.32 | - | - | 0.44 | - | 0.25 | 0.2 |
| F | 0.89 | 0.63 | 1.57 | 1.09 | - | 2.84 | - | 1.41 | - |
| P | 4.87 | 5.02 | 4.12 | 4.61 | 5.9 | 7.15 | 1.86 | 5.53 | 6.8 |
| Ant | 1.37 | - | - | - | - | - | - | - | - |
| Fl | 1.17 | 0.76 | 0.54 | 5.17 | 2.62 | 0.93 | - | 0.78 | - |
| Pyr | 1.82 | 1.74 | 1.07 | 15.02 | 7.16 | 1.07 | 3.16 | 1.4 | 2.4 |
| BaAnt | 0.75 | - | - | 33.08 | 1.38 | - | - | - | - |
| Chr | - | - | - | 20.79 | - | 1,52 | - | - | - |
| BbFl | 0.63 | 0.49 | 0.49 | 14.0 | 3.15 | 10.67 | 2.17 | 0.52 | - |
| BkFl | 0.47 | 0.43 | 0.42 | 2.74 | 2.61 | 2.43 | 1.82 | 0.43 | - |
| BaPyr | 2.98 | 2.74 | 2.71 | 7.05 | 16.79 | 6.17 | 11.86 | 2.76 | - |
| DBahAnt | - | - | - | 2.11 | - | 4.34 | - | - | - |
| BghiPer | 1.4 | 1,27 | 1.26 | 10,61 | 7.82 | 114.69 | 5.45 | 1.28 | - |
| Ipyr | 0.86 | 0.48 | 2.19 | - | - | 98.04 | - | 0.94 | 1.8 |
| **ΣPAHs** | **17.5** | **13.8** | **16.9** | **116.3** | **47.4** | **255.0** | **26.3** | **15.3** | **11.2** |



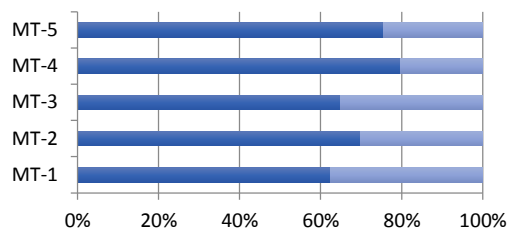**Figure 1.** PAHs distribution in mass percentage in NAG plastic debris: light (blue), heavy (light blue).

*4.3 Determination of PCBs*

In this part of the study, 61 PCBs were sought and 12 different PCBs were detected in NAG samples and 28 PCBs in ICC (Figure 3 and Table 4). PCBs were detected in 60% of the samples collected in NAG. 1 to 6 PCBs were identified in these samples with very high concentration in PCB52 and the most frequently detected appears to be PCB105. Individual PCB concentration was in the range 0.6 to 50 ng.g$^{-1}$ while total PCBs concentrations revealed to be in the range 0.6 to 59 ng.g$^{-1}$ in accordance with the literature (Hirai 2011). Moreover, the total abundance of PCBs was considered regarding PCBs distribution between high-chlorinated, HC-PCBs, more than 5 chlorine atoms

*i.e.* from PCB1 to PCB126 and low-chlorinated, LC-PCBs, up to than 5 chlorine atoms *i.e.* from PCB127 to PCB209. Thus, figure 4 puts in evidence that NAG samples mainly contain tetra and pentachlorinated PCBs *i.e.* LC-PCBs (except sample NA-1).

**Table 3.** PAHs concentration in ng per ICC plastic debris gram

| MT | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|
| N | 421.2 | 162.5 | 83.4 | 1262.4 | 72.6 |
| Ace | - | 3.9 | - | - | - |
| F | 43.66 | 3.75 | 2.23 | 23.17 | 4.07 |
| P | 45.38 | 32.28 | 10,56 | 147.62 | 27.36 |
| Fl | 45.06 | 12.68 | 16.67 | 162.99 | 11.85 |
| Pyr | 62.60 | 11.21 | 17.61 | 148.33 | 10.97 |
| Chr | 22.62 | 0.60 | 8.69 | - | - |
| BbFl | - | 5.05 | - | - | 4.90 |
| BkFl | 20.73 | 4.64 | 3.31 | 17.97 | 2.48 |
| BaPyr | 52.85 | 12.51 | 9.25 | 50.22 | 3.86 |
| DBahAnt | 44.83 | 8.89 | 4.81 | 69.16 | 11.52 |
| BghiPer | 72.12 | 19.94 | 9.26 | 58.83 | 3.90 |
| Ipyr | 57.69 | 30.18 | 8.22 | 62.15 | - |
| **ΣPAHs** | **888.7** | **308.0** | **174.0** | **2,002.8** | **153.5** |



**Figure 2.** PAHs distribution in mass percentage in ICC plastic debris: light (blue), heavy (light blue).
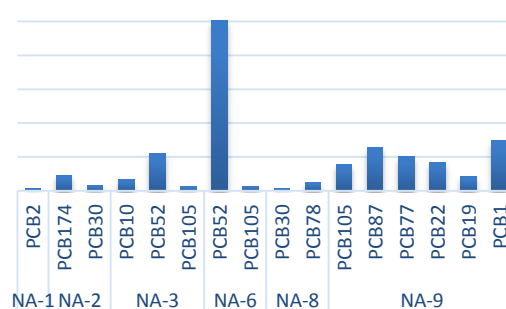


**Figure 3.** PCBs concentration in ng per NAG plastic debris gram.

PCBs were detected in all ICC samples as showed on figure 5. PCB101 was always detected and other PCBs were often detected in these samples: PCB4, 6, 29, 77, 101, 147 and 173. Individual PCB concentration was in the range 2.1 to 15,664 ng.g$^{-1}$ while total PCBs concentrations revealed to be in the range 1,243 to 18,699 ng.g$^{-1}$. These concerned concentrations are around two times higher than

those cited in the literature (Rios 2014, Hirai 2011, Teuten 2009).
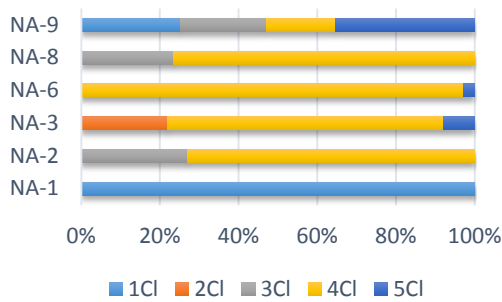


**Figure 4.** PCBs distribution in mass percentage in NAG plastic debris as a function of Cl atom number.
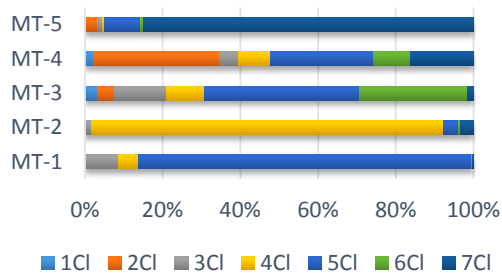


**Figure 5.** PCBs distribution in mass percentage in ICC plastic debris as a function of Cl atom number.

Figure 5 clearly shows that hexa and heptachlorinated PCBs are present in non-negligible amount in most of the samples. This tends to underline the predominance of HC-PCBs in ICC debris, mainly the ones collected close to the coast (MT-3 to 5). PAHs detected in plastic debris differed from sampling area and their total concentrations revealed to be 10 times higher in Indonesian samples than NAG ones. The presence of many PAHs at very high concentration in plastic debris from urban beaches highlights risks associated with these POPs. However, the presence of H-PAHs in plastic debris collected in remote ocean such as BaPyr must be considered regarding its high toxicity. Even if urban beach samples mainly contain L-PAHs while gyre samples contain H-PAHs, the concentration of L and H-PAHs in coastal plastic debris generally remain higher than in open ocean ones. Such results could be assigned to: (*i*) short residence time in urban beaches that did not allow to reach equilibrium; (*ii*) release or degradation of chemicals from coast to gyre during transport by ocean current; (*iii*) great affinity of H-PAHs for PE and PP debris because of their remaining in open ocean debris. Such affinity has been demonstrated for PE but further studies are needed to address other issues (Choi 2013). Furthermore, it is on interest to note that PCBs detected in plastic debris are also chemically distinct in both sources and their concentrations are 300 times greater in Indonesia samples than others. ICC plastic debris have high PCB concentration - PCB52, 101 and 153 in a lesser extent, that belong to the list of six indicator PCBs-is of huge worry regarding the plastic pollution level in this part of the world (Jambeck 2015). In addition, the detection of PCB77 and 105 in plastic debris collected in

gyre is of great concern because they are listed in Water Framework Directive (2013/39/EU) as dioxin-like compounds. Moreover, the presence of LC-PCBs such as PCB52 and 105 in NAG debris is worried because of their demonstrated ingestion by marine organisms (Yamashita, 2011).

**Table 4.** PCBs concentration in ng per ICC plastic debris gram.

| MT | 1 | 2 | 3 | 4 | 5 |
|---|---|---|---|---|---|
| PCB1 | - | - | 42.0 | 207.6 | 2.2 |
| PCB3 | 7.4 | - | - | - | - |
| PCB4 | - | - | - | 1,872.8 | 363.9 |
| PCB6 | 10.6 | - | 55.2 | 261.4 | 213.7 |
| PCB10 | - | - | - | 1,025.1 | - |
| PCB16 | - | 2.3 | 44.3 | - | 6.4 |
| PCB18 | - | 48.6 | - | 72.3 | 21.3 |
| PCB22 | - | - | 36.9 | 404.4 | - |
| PCB25 | - | - | 58.5 | - | - |
| PCB28 | - | 9.0 | 0.9 | - | 82.9 |
| PCB29 | 1,608.3 | - | 28.7 | - | 45.1 |
| PCB52 | - | 3,019.1 | - | - | - |
| PCB56 | - | 12.6 | - | - | 16.6 |
| PCB66 | - | 16.0 | - | 37.4 | - |
| PCB67 | 21.7 | - | - | 113.2 | - |
| PCB74 | - | 2.8 | - | 167.3 | - |
| PCB77 | 927.7 | - | 126.1 | 491.8 | 100.0 |
| PCB82 | 256.1 | - | - | - | - |
| PCB101 | 15,548.1 | 130.8 | 508.0 | 2,590.2 | 1,723.4 |
| PCB104 | 256.6 | - | - | - | - |
| PCB138 | - | 14.4 | - | - | - |
| PCB147 | - | 3.7 | 304.7 | 929.1 | 105.6 |
| PCB149 | - | - | 19.3 | - | - |
| PCB153 | 14.7 | - | 33.0 | - | 28.5 |
| PCB173 | 42.2 | 56.9 | - | 1,568.5 | 15,664.4 |
| PCB177 | 19.7 | - | - | - | - |
| PCB179 | - | 17.5 | 18.2 | - | - |
| PCB180 | - | 38.6 | - | - | - |
| ∑PCBs | **18,698.4** | **3,372.3** | **1,242.8** | **9,741.1** | **18,345.5** |

At least, coastal plastic debris chemical content revealed that HC-PCBs are predo-minant while the opposite was observed for gyre debris. One more time, previous hypotheses could explain such results. However, even if it has been proved that HC-PCBs have great affinity for polymer like PE (Choi 2013), their detection at level of traces may indicate that other processes are involved (Rios 2014)

**5. Conclusion**

The study of PAHs and PCBs contained in plastic debris collected in different areas clearly showed differences in

concentration, nature and distribution. Coastal debris revealed to contain high concentration in PCBs and PAHs with a predominance of L-PAHs and HC-PCBs while gyre plastic debris are less polluted samples and mainly contain H-PAHs and LC-PCBs. Such results suggest the degradation or release of chemicals during transport by ocean current but also underline the presence of high toxic POPs in marine plastic debris from Indonesian Cilacap coast and North Atlantic gyre.

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