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Occurrence, distribution and influence factors.**

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1 **PAEs and PAHs in the surface sediments of the East China Sea:**

2 **Occurrence, distribution and influence factors**

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25 **Abstract**

26 A total of 29 sediment samples were collected from the East China Sea (ECS),

27 with the Yangtze River estuary and the Zhejiang coastal area. These sediment samples
28 were analyzed for 6 phthalate esters (PAEs) and 16 polycyclic aromatic hydrocarbons
29 (PAHs): the Σ PAEs and Σ PAHs concentrations ranged between 1649.5–8451.5 ng g⁻¹
30 (mean = 3446.3 ng g⁻¹) and 57.5–364.5 ng g⁻¹ (mean = 166.2 ng g⁻¹), respectively.
31 Overall, the PAEs and PAHs concentrations gradually decreasing in the offshore and
32 southward directions: their compositions and distributions suggest they could have
33 mainly derived from the Yangtze River. In particular, their distribution was influenced
34 by the sources' proximity, hydrodynamics, and sediment geochemistry (i.e., TOC
35 content and grain size). A classical two-end member model was utilized to estimate the
36 fraction of terrestrial organic carbon in the sediments of the ECS. When the sediment
37 was dominated by terrestrial-derived organic matter (OM), the concentrations of PAEs
38 and PAHs were significantly correlated to the TOC content and grain size of the
39 sediments. In contrast, the poor correlation of TOC content and grain size with PAEs in
40 those sediments dominated by marine-derived OM, implied that the distribution of
41 PAEs in the ECS was mainly related to land-based inputs, (especially to that of the
42 Yangtze River). Regardless of the origin of most of the OM contained in the sediments,
43 we observed positive correlations between the TOC content, and grain size of those
44 containing PAHs. These results suggest that the distribution of PAHs in the ECS was
45 not only related to the Yangtze River input, but also to the geochemical characteristics
46 of the sediments.

47 Keywords: PAEs; PAHs; distribution characteristics; sediment; ECS

48

49

50 **1. Introduction**

51 Organic contaminants enter the marine ecosystems through various pathways,

52 including wastewater and riverine inputs, overland flows, atmospheric transport and
53 deposition, or via adsorption onto particles and finally end up in the sediment (Zhang
54 et al., 2018a; Montuori et al., 2016). The relative importance of these pathways depends
55 largely on the distance from the input sources and on the characteristics of the emitted
56 particles; due to sedimentation dynamics, the contributions from lateral transport may
57 decrease with increasing distance (Liu et al., 2012). Marine sediments are important
58 sources of information, which can elucidates the fate of organic compounds over long
59 periods of time. The factors controlling the fate of organic contaminants in sediments
60 include, among others, the sediments' characteristics, the pollutants' geochemical
61 characteristics, and the environmental conditions (Louvado et al., 2015).

62 Phthalate esters (PAEs) are a group of chemical compounds that are widely used
63 in the manufacturing and processing of plastics products (e.g., plasticizers), which are
64 employed for a broad range of industrial and consumer products (Zhang et al., 2018a;
65 Chen et al., 2017). Previous studies have shown that PAEs are endocrine-disrupting
66 chemicals that pose potential health risks to humans and other organisms; for example,
67 they can disrupt the hormonal balance of mammalian species (Wang et al., 2016). PAEs
68 are easily degraded by light, as well as by degraded bacteria and actinomycetes;
69 therefore, they are not considered as persistent chemicals (Wezel et al., 2000). After
70 PAEs enter an aquatic system, given their low solubility, they are easily adsorbed onto
71 particles and eventually accumulate in the bottom sediments (Liu et al., 2014).

72 Polycyclic aromatic hydrocarbons (PAHs) are organic compounds with two or
73 more fused aromatic rings produced during the incomplete combustion of organic
74 matter (OM). PAHs are lipophilic, non-biodegradable, environmentally persistent, toxic
75 and a few of individual PAHs are categorized as carcinogen (Pérezfernández et al.,
76 2018). Sources of PAHs included, among others, motor vehicle exhausts, industrial

77 emissions, coal burning for commercial and household heating, forest fires and the
78 combustion of other biomass fuels. Consequently, PAH pollution mainly occurs in
79 densely populated areas, major manufacturing districts, intensive agricultural basins,
80 and other industrial centers (Han et al., 2017). Due to their low solubility and high
81 hydrophobic-lipophilic characteristics, in marine ecosystem PAHs tend to bound to
82 suspended particles and be subsequently deposited within sediments (Sun et al., 2018).
83 The East China Sea (ECS) is a typical marginal sea located in the western part of the
84 northwest Pacific Ocean, which receives large amounts of fresh water and sediment
85 discharges from the Yangtze River (Changjiang). This river, the third longest river in
86 the world, is heavily contaminated by industrial-agricultural discharges and by the
87 runoff of big cities along the river (Adeleye et al., 2016). Moreover, the ECS receives
88 a large amount of pollutants from the Yangtze River Delta, which is one of the most
89 urbanized and industrialized regions of China (Zhou et al., 2014). Due to the importance
90 of ECS in Chinese coastal, the levels, compositions, sources and ecological risk of
91 PAHs in ECS surface sediment have been studied. For example, Hung et al. (2011)
92 reported the distribution and levels of PAHs in sediments from ECS. Deng et al. (2013)
93 investigated the source apportionment of PAHs in mud areas of ECS. Li et al. (2017a)
94 studied the distribution, sources and ecological risk of PAHs in the estuarine-coastal
95 sediments of ECS. However, few PAE-related studies in ECS surface sediment have
96 been conducted, and these studies only focused on the levels and distribution. Yang et
97 al. (2011) reported the occurrence and distribution of PAEs in sediments from ECS, and
98 Zhang et al. (2018a) reported the occurrence and distribution of PAEs in sediments of
99 Changjiang River Estuary and its adjacent area.

100 Marginal seas play an important role in the transport and storage of terrestrial input
101 organic materials, since they represent transition zones between the continent and the

102 open sea (Hung et al., 2011). Thus, the ECS can act as an important sinks of pollutants
103 that originate from land-based pollution sources, and affect the transportation of
104 pollutants into the open sea. In addition, the Zhejiang-Fujian Coastal Current (ZFCC),
105 the Jiangsu Coastal Current (JCC), and the Taiwan Warm Current (TWC) also
106 significantly affect the study area (Zhang et al., 2015). ECS as one of an active interface
107 between terrestrial and oceanic environments have complex biogeochemical processes.
108 In order to reveal the fate of organic pollutants in the ECS, a few studies have been
109 conducted. Still, local controls on distribution of organic pollutants in the sediments
110 (e.g., different OM sources or geochemical characteristics of sediment) remain unclear.

111 In this study, we analyzed the occurrence of sixteen PAHs and six PAEs in
112 sediment samples collected from the ECS. The main objectives of this work were to
113 investigate the contamination status and to compare the distribution characteristics of
114 PAHs and PAEs in the surface sediments, as well as to identify the potential factors
115 influencing their distribution.

116

117 **2. Experimental**

118 **2.1. Sample collection and preparation**

119 A total of 29 representative surface sediment samples (0–2 cm) were collected
120 using a box sampler from 29 sampling locations in the ECS between May–June 2017
121 (Table S1). After homogenization, the sediment samples were enclosed in a pre-cleaned
122 aluminum foil and immediately stored frozen until analysis. Before extraction, the
123 samples were freeze-dried, grinded, and sieved.

124 Approximately 0.5 g of grated sediment sample were placed in a glass vial, mixed
125 with dichloromethane and ultrasonic-extracted. The extract was evaporated with a
126 stream of nitrogen until reaching a volume of ~100 μ L; then, they were further purified

127 through gas purge microsyringe extraction (GP-MSE) (Yang et al., 2011). Details about
128 the GP-MSE are presented in the [Supplementary material](#). All samples were spiked
129 prior to extraction with a mixture of isotopically labeled compounds, which were used
130 as internal standards (²H₁₀-acenaphthene, ²H₁₀-fluoranthene and ²H₁₀-perylene,
131 (Accustandard, New Haven, CT, USA).

132 **2.2. Total organic carbon (TOC) and grain size of sediments**

133 The freeze-dried and grated sediment samples were treated with 4 N HCl to
134 remove any carbonate, were rinsed with deionized water, and subsequently dried at
135 60 °C for 12 h. The resulting carbonate-free samples were analyzed for the TOC content
136 using a Vario EL-III Elemental Analyzer. A stable carbon isotope analysis of the
137 organic carbon was also performed on these samples using a Thermo Deltaplus XL
138 mass spectrometer operating in a continuous flow mode.

139 The grain size of the sediment samples was determined using a laser particle size
140 analyzer (Mastersizer 2000, Malvern Instruments Ltd., Malvern, Worcestershire, UK).

141 **2.3. Classical two-end member model of the OM δ¹³C signatures**

142 The δ¹³C values change depending on the carbon sources; hence, they can be used
143 effectively to distinguish different sources (marine or terrestrial) of the bulk
144 sedimentary OM. The terrestrial OM fraction (F_t) in a sample is given by:

$$145 F_t = (\delta^{13}C_s - \delta^{13}C_m) / (\delta^{13}C_t - \delta^{13}C_m) \times 100\% \quad (1)$$

146 Where δ¹³C_m and δ¹³C_t are the δ¹³C values of the marine and terrestrial end members,
147 and δ¹³C_s is that of the sediment sample. We assumed that the OM sample was
148 composed exclusively of terrestrial OM delivered by the Yangtze River (δ¹³C_t =
149 -25.6‰) and of marine OM derived from local sources (δ¹³C_m = -20.0‰) (Wu et al.,
150 2007; Wang et al., 2018). The fraction of marine OM (F_m) was given by $F_m = 1 - F_t$.

151 **2.4. Chemicals and materials**

152 Six PAEs and 16 PAHs standard mixtures were purchased from AMP (USA) and
153 Accustandard (New Haven, CT, USA), respectively. The purity of all standards was
154 higher than 99%. A mixture standard solution of PAEs was prepared in hexane at a
155 concentration of 1000 mg L⁻¹, while a mixture standard solution of PAHs was prepared
156 in acetone at a concentration of 100 mg L⁻¹. Detail information about the PAHs and
157 PAEs standards are provided in [Table S2](#). The standard solutions were stored in the dark
158 at 4°C until use. HPLC grade organic solvents (e.g., dichloromethane and hexane) were
159 purchased from Caledon (Georgetown, Canada).

160 **2.5. Instrumental analysis**

161 The analysis and detection of the PAEs and PAHs were conducted using a gas
162 chromatography coupled to a quadrupole mass spectrometer with electron impact
163 ionization (Shimadzu GC 2010 System connected with a Shimadzu QPMS 2010 MS).
164 The separation was achieved within a DB5 fused-silica capillary column (30 m × 0.25
165 mm × 0.25 mm). Details about the GC-MS analysis are presented in the [Supplementary](#)
166 [material](#).

167 **2.6. Quality control and quality assurance (QA/QC)**

168 All the analyses were performed according to the quality assurance and quality
169 control measures. The quantification was done following an internal standard method,
170 and the correlation coefficients (r^2) of all the PAEs and PAHs calibration curves were
171 higher than 0.99. The recoveries of the three internal standards for the sediment samples
172 were in the range of 59.8–104.8%, 62.0–89.2%, and 51.1–80.2%, respectively. The
173 analyses were simultaneously carried out for every set of blank, laboratory blank spiked,
174 laboratory duplicate, and matrix spiked samples. The recoveries for the laboratory
175 blank spiked samples ranged between 67.8–111.9%, while those of the matrix spiked
176 samples between 62.3–124.9%. The in-lab reproducibility was in the range of 2.6–

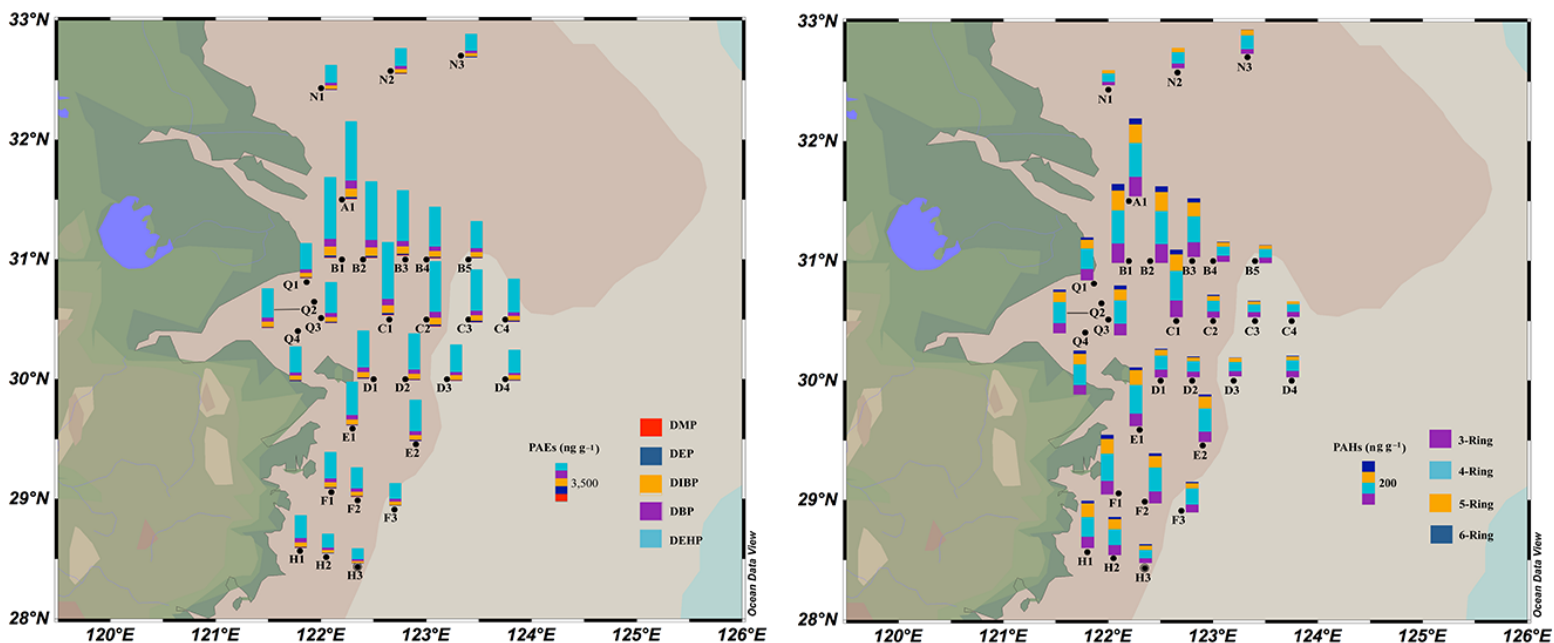
177 13.1%.

178

179 **3. Results**

180 **3.1. PAEs in ECS sediments**

181 Five out of the 6 target PAE compounds (except di-n-octyl phthalate (DnOP)) were
182 detected in the 29 sediment samples collected from the ECS. [Table S3](#) summarizes the
183 concentration of the individual and Σ PAEs in the surface sediments; the Σ PAEs
184 concentrations in the sediments oscillated between 1550.4–7381.5 ng g⁻¹ (mean =
185 3873.9 ng g⁻¹). The highest concentration was found at site B1 (in the southeastern area
186 of the Yangtze River estuary), while the lowest concentration was recorded at site H3
187 (in the southern part of the Zhejiang Coastal area) ([Figure 1](#)). Among the individual
188 PAEs, di-(2-ethylhexyl)-phthalate (DEHP) was the most abundant (1100.6–5643.4 ng
189 g⁻¹; mean = 2916.8 ng g⁻¹), followed by diisobutyl phthalate (DIBP) (202.3–822.6 ng
190 g⁻¹; mean = 466.2 ng g⁻¹) and dibutyl phthalate (DBP) (205.5–714.1 ng g⁻¹; mean =
191 380.1 ng g⁻¹). These compounds account on average for 73.5% (59.1–81.7%), 12.8%
192 (9.0%–20.8%) and 10.6% (7.6%–15.6%) of the total PAEs concentration, respectively
193 ([Figure S1](#)). Such results are consistent with the commonly accepted notion that DBP,
194 DIBP and DEHP are the dominant PAE components in sediments ([Li et al., 2016a](#); [Li](#)
195 [et al., 2017b](#); [Zhang et al., 2018a](#)). In addition, we noted higher concentrations of PAEs
196 in the sediment samples collected from the Yangtze River estuary and a decrease of
197 these concentrations northward, in the direction of the outer shelf.



199 Figure 1 Concentration of the PAEs and PAHs in the ECS surface sediment

200 3.2. PAHs in ECS sediments

201 Sixteen target PAH compounds were detected in the 25 sediment samples collected
 202 from the ECS. Among them Naphthalene (Nap) did not satisfy the criteria of QA/QC
 203 considered in this study: the blank values were three times higher than those of the
 204 method detection limit. Thus, the concentration of Nap was not determined and this
 205 compound was not considered in further discussions.

206 Table S4 summarizes the concentration of individual and total PAHs in the
 207 sediment; the \sum PAHs concentrations in the sediments were comprised between 70.7–
 208 383.6 ng g⁻¹ (mean = 179.9 ng g⁻¹). The highest concentration was recorded at site B1
 209 (in southeastern area of the Yangtze River estuary), while the lowest in the lowest
 210 longitude-sampling site (C4) (Figure 1). Phenanthrene (Phe) (14.8–65.2 ng g⁻¹; mean
 211 = 32.6 ng g⁻¹), pyrene (Pyr) (15.1–56.8 ng g⁻¹; mean = 31.8 ng g⁻¹) and fluoranthene
 212 (Fluo) (10.5–52.1 ng g⁻¹; mean = 26.8 ng g⁻¹) were the most abundant PAHs detected,
 213 followed by benzo[a]pyrene (BaP) (4.6–41.7 ng g⁻¹; mean = 17.0 ng g⁻¹),
 214 benzo[b]fluoranthene (BbF) (3.7–34.8 ng g⁻¹; mean = 16.1 ng g⁻¹), and
 215 benzo[a]anthracene (BaA) (3.7–26.0 ng g⁻¹; mean = 12.8 ng g⁻¹). As shown in Figure

216 S2, 4-ring PAHs (41.2%–56.3%; mean = 47.4%) were the most abundant type of PAHs,
217 followed by 3- and 5-ring PAHs (3-ring: 20.5%–33.1%; mean = 25.6%; 5-ring: 16.9%–
218 28.7%; mean = 22.4%) in most of the sampling stations. Generally, higher
219 concentrations of PAHs with relatively high molecular weights are observed in marine
220 sediments (Huang et al., 2012). Moreover, relatively high concentrations of PAHs were
221 found in the sediment samples collected from the Yangtze River estuary (sampling sites
222 A1, B1 and B2) and the Zhejiang Coastal area (sampling sites E1, F1 and H1); the PAHs
223 concentrations decreased similarly toward the outer shelf.

224

225 **4. Discussion**

226 **4.1. Concentration levels of PAEs and PAHs in the ECS and other areas**

227 The Σ PAEs and Σ PAHs concentrations in the ECS sediments were compared with
228 those previously reported in other parts of China and abroad. The results are shown in
229 Table 1. Generally, the Σ PAEs concentrations in the sediments were found to be slightly
230 lower in the ECS (1550.4–7381.5 ng g⁻¹) than in other seas, such as the Bohai and
231 Yellow Sea in northern China (1.24–15.8 μ g g⁻¹), the Pearl River estuary in southern
232 China (0.88–13.6 μ g g⁻¹), the Kaohsiung Harbor in Taiwan (1.27 \pm 0.76–51.94 \pm 29.21
233 μ g g⁻¹) and the coastal areas of Kuwait (2.15 to 15.72 μ g g⁻¹); however, they are similar
234 to those observed in the Southern Yellow Sea (311.4–6156.5 ng g⁻¹). Moreover, the
235 Σ PAEs concentrations in the ECS sediments were 2–7 times lower than those in the
236 sediments of the Pu River in northeastern China (3.71–46.9 μ g g⁻¹) and of the Pearl
237 River Delta (PRD) region (0.567–47.3 μ g g⁻¹), but comparable to those in the sediments
238 of the Qiantang River in eastern China (0.59 to 6.74 μ g g⁻¹).

239 The Σ PAHs concentrations observed during our study (58.6–351.6 ng g⁻¹) were
240 similar to those detected in the Liaodong Bay in China (88.5–347.1 ng g⁻¹), Daya Bay

241 in China (140.0–491.0 ng g⁻¹), the Libyan coastal area in the Mediterranean Sea (41.8–
 242 388 ng g⁻¹) and in the Tiber River estuary in Italy (36.2–545.6 ng g⁻¹). Moreover, the
 243 Σ PAHs concentrations in the sediments of the ECS were 2–4 times lower than those in
 244 the Bohai Sea in China (149.2–1211.8 ng g⁻¹), Rizhao coastal area in China (79.3–853.0
 245 ng g⁻¹), the northern part of the Yellow Sea in China (148.3–907.5 ng g⁻¹) and the Pearl
 246 River estuary in southern China (144.0–1289.0 ng g⁻¹). Finally, the Σ PAHs
 247 concentrations observed in the ECS were slightly higher than those on the Chinese
 248 continental shelf (e.g., the inner Yellow Sea (53–224 ng g⁻¹) and the South China Sea
 249 (27–110 ng g⁻¹).

Table 1 Concentration levels of PAEs and PAHs in ECS and other different areas

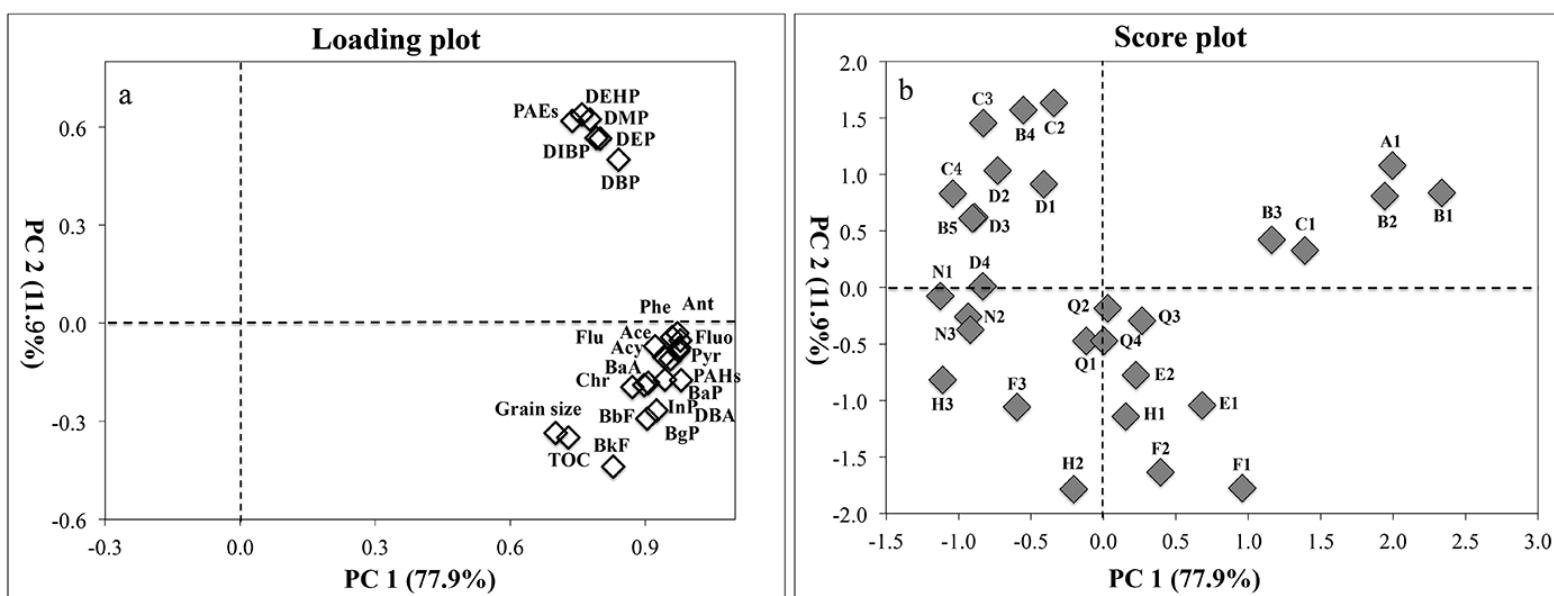
	Location	Research area	Sample	Concentration	Refrence
PAEs	North China	Bohai and Yellow Sea	Sediment	1.24–15.8 (16)	Zhang et al. (2018a)
	Yellow Sea, China	Southern yellow sea	Sediment	0.31–6.15 (4)	Li et al. (2014)
	South China Sea	Pearl River Estuary	Sediment	0.88–13.6 (6)	Li et al. (2016a)
	Taiwan, China	Kaohsiung Harbor	Sediment	1.27 ± 0.76–51.94 ± 29.21 (10)	Chen al. (2017)
	Kuwait	Kuwait's coastal areas	Sediment	2.15–15.72 (6)	Saeedet al. (2017)
	Northeast China	Pu River	Sediment	3.71–46.9 (6)	Li et al. (2016b)
	South China	Pearl River Delta (PRD) region	Sediment	0.567–47.3 (16)	Liu et al. (2014)
	East China	Qiantang River	Sediment	0.59–6.74 (16)	Sun et al. (2013)
	East China	East China Sea	Sediment	1.55–7.38 (6)	This Study
PAHs	Bohai Sea, China	Bohai sea	Sediment	149.24–1211.81 (16)	Li et al. (2016c)
		Liaodong Bay		88.5–347.1 (16)	Zhang et al. (2018b)
	Yellow Sea, China	Rizhao coastal area	Sediment	79.3–853 (16)	Chen et al. (2012)
		Northern part of the Yellow Sea		148.28–907.47 (16)	Li et al. (2015)
	South China Sea	Pearl River Estuary	Sediment	144–1289 (16)	Yuan et al. (2015)
		Daya Bay		140–491 (16)	
	Mediterranean Sea	Libyan coastal area	Sediment	41.8–388 (16)	Bonsignore et al. (2018)
	Italy	Tiber River estuary	Sediment	36.2–545.6 (16)	Montuori et al. (2016)
	Algeria	Annaba Bay	Sediment	250.16–509.58 (16)	Khaled-Khodja and Rouibah. (2018)
	East China	East China Sea	Sediment	70.7–383.6 (16)	This Study

The numbers in brackets indicate the number of PAEs and PAH compounds studied.

The unit of PAEs in sediment is $\mu\text{g g}^{-1}$; The unit of PAHs in sediment is ng g^{-1} .

250 4.2. Spatial distribution of PAEs and PAHs

251 As shown in Figure 1, the PAEs and PAHs concentrations decreased with
252 increasing longitude, indicating lower pollution levels in the offshore rather than in the
253 inshore areas. This result illustrates that the potential source (e.g., terrestrial emissions)
254 of pollution might have been located closed to the inshore sites; therefore, the
255 concentration of ΣPAEs and ΣPAHs was higher than that in offshore sites. Moreover,
256 PAEs and PAHs were detected in all of the surface sediment samples, suggesting their
257 widespread distribution in the marine sediments of the ECS region.



259 Figure 2 Loadings (a) and score (b) plot of the principal components

260 A Principal Component Analysis (PCA) was conducted to analyze the spatial
261 distribution of PAEs and PAHs in the sediments. The PCA was based on the individual
262 and total PAEs and PAHs concentrations, on the TOC content and on the grain size data
263 obtained from the 29 sediment sampling sites in the ECS. The loading and factor score
264 plots of the PAEs and PAHs in the ECS sediments are presented in Figure 2. The PCA
265 resulted in the extraction of two principal components (PCs) with eigenvalues >1 ,
266 which explained 89.9% of the total variation in the analyzed data. The individual and

267 total PAHs, PAEs, as well as the TOC and grain size values were mainly associated
268 with PC1, which explained 77.9% of the variation. Half of the sampling sites plotted in
269 the PCA corresponded to positive values of the PC1 axis, and were consistent with the
270 PAHs, PEAs, TOC and grain size vectors. The samples corresponding to the positive
271 section of the PC1 axis were characterized by fine-grained sediments, high TOC levels
272 and high concentrations of PAEs and PAHs. As shown in [Figure 2](#), the higher PC1
273 scores (larger than 0.5) were recorded in the Yangtze River estuary (sampling sites A1,
274 B1, B2, B3 and C1) and in the inshore zone of the Zhejiang coastal area (sampling sites
275 E1, E2 and F1). Relatively high concentrations of PAEs, PAHs and TOC, as well as fine
276 grain sizes were also observed at these sampling sites: they represent significant
277 deposition centers of Yangtze-derived fine-grained sediments in the ECS ([Liu et al.,](#)
278 [2006](#)). Organic pollutants, especially high molecular weight ones with high K_{OW} values,
279 can easily adsorb to particles due to their high hydrophobicity ([Duan et al., 2013](#); [Zheng](#)
280 [et al., 2014](#)). Given this characteristic, organic pollutants can be rapidly and abundantly
281 transported downward by sinking particles, once they are discharged into water bodies
282 ([Liu et al., 2014](#)). The PAEs and PAHs discharged into the ECS should have been
283 quickly incorporated into the sediments and preferentially deposited near the coast,
284 justifying their relatively high concentrations at the inshore sites, especially those in the
285 Yangtze River estuary.

286 PC 2 explained 11.9% of the total variation. The sampling sites plotted in the PCA
287 in correspondence of the positive PC 2 axis were consistent with the PAEs and 3-ring
288 PAH vectors, but opposite to the PAHs, TOC and grain size vectors. The negative part
289 of the PC 2 axis showed that the distribution of the PAHs was influenced by sediment
290 geochemistry (i.e., TOC content and grain size). When PC1 was plotted against PC2,
291 we noticed that the sediment samples from the inshore sites formed different groups

292 (one for those of the Yangtze River estuary and another for those of the Zhejiang costal
293 area) and were clearly separated from those collected in the offshore region (Figure 2b).
294 These groupings reflected the PAEs and PAHs loadings of sediments collected from
295 inshore locations, the PAHs loadings of sediments collected from the Zhejiang Coastal
296 sites, and the PAEs loadings of sediments collected from the Yangtze River estuary sites
297 (Figure 2b).

298 **4.3. Effect of sediment geochemistry (TOC content and grain size) on distribution** 299 **of PAEs and PAHs**

300 A previous study has inferred that the TOC and grain size (Mz) of sediments are
301 two important factors controlling the spatial distribution of persistent organic pollutants
302 (POPs) in aquatic environment (Zheng et al., 2014). Like POPs, PAEs and PAHs can
303 adsorb onto particulate matters due to their high hydrophobicity. The concentrations of
304 TOC, PAEs and PAHs in sediments of the ECS were examined through Spearman's
305 correlations (Table 2). The concentration of Σ PAHs presented significant positive
306 correlations with the TOC content ($p < 0.01$, $r^2 = 0.73$) and the grain size ($p < 0.01$, $r^2 =$
307 0.69); on the contrary, the concentration of PAEs showed no significant correlations
308 with neither the TOC content or the grain size. These results indicated that the
309 distribution of PAHs in the ECS sediments was controlled by the sediment
310 geochemistry; moreover, a unique distributional pattern of the PAEs could be observed
311 within specific regions of the ECS.

312

313

Table 2 Spearman correlation for sediment geochemistry and target compound in surface sediments

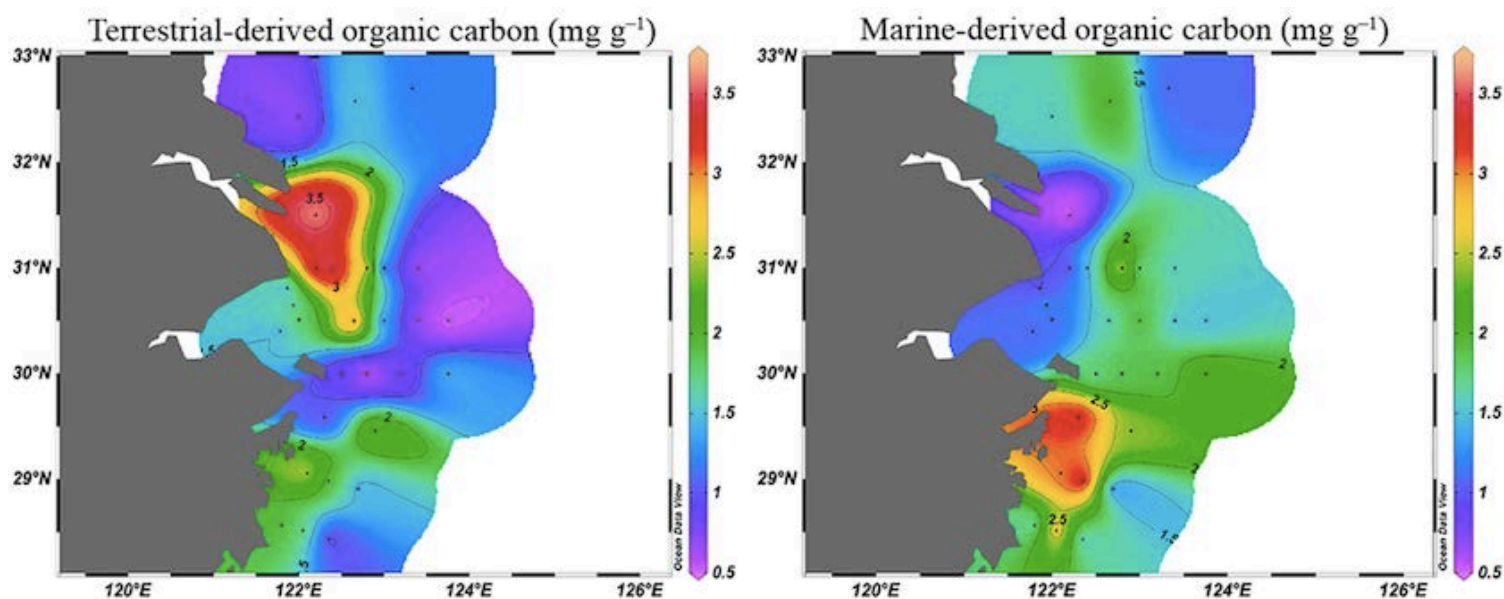
TOC	OC-T	OC-M	Grain size	PAEs	PAHs
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All site	PAEs	0.25	0.38*	-0.04	0.21	1.00	
	PAHs	0.73**	0.92**	0.01	0.69**	0.39*	1.00
Inshore site	PAEs	0.62**	0.86**	-0.03	0.43*	1.00	
	PAHs	0.76**	0.97**	0.12	0.68**	0.84**	1.00
Offshore site	PAEs	-0.24	-0.41	0	-0.05	1.00	
	PAHs	0.88**	0.78**	0.89**	0.82**	0.01	1.00

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

314 Most of the OM that reaches the ECS marginal sea is brought by the Yangtze River.
315 The terrestrial inputs were expected to dominate over the marine inputs in both the
316 suspended and surface sediments of the inner estuary (Wu et al., 2007; Wang et al.,
317 2018). The two primary end-members of the OM in the ECS sediments were terrestrial
318 matter (mainly transported from the Yangtze River) and marine autochthonous matter
319 (Wu et al., 2007). A classical two-end member model of the OM $\delta^{13}\text{C}$ signatures was
320 used to estimate the fractions of terrestrial (F_t) and marine organic carbon (F_m) in the
321 ECS surface sediments (Eq. (1)); the corresponding results are presented in Table S1.
322 According to these data, the terrestrial-derived organic carbon (OC-T) generally
323 occurred in the inshore zones of the Yangtze River estuary and of the Zhejiang coastal
324 area, while the marine-derived organic carbon (OC-M) was occurred in the offshore
325 zones of the Yangtze River estuary and of the Zhejiang Coastal area (Figure 3).
326



328 Figure 3 Terrestrial-derived organic carbon (OC-T) and marine-derived organic carbon (OC-M)
 329 in the ECS surface sediment

330 Although no significant relationship was found between the TOC and the PAEs
 331 concentration in the ECS surface sediments, we discovered that the Σ PAEs
 332 concentrations were positively correlated ($p < 0.05$, $r^2 = 0.38$) with the OC-T, especially
 333 in the terrestrial OM-dominated area ($p < 0.01$, $r^2 = 0.86$). Another positive correlation
 334 ($p < 0.05$, $r^2 = 0.43$) was also observed between the Σ PAEs concentration and the grain
 335 size in the terrestrial OM-dominated area. However, no significant correlation was
 336 found between the Σ PAEs concentration and the OC-M. One of the possibilities is that
 337 the pollutants adsorbed onto sediment particles did not reach the adsorption-desorption
 338 equilibrium during particle settling, due to the high sedimentation rates characterizing
 339 the inshore and estuary areas (Duan et al., 2013). These statistical results indicated that
 340 the PAEs and OC-T should have had similar sources, or that they were subjected to the
 341 same transport processes. Hence, the distribution of PAEs in the ECS surface sediments
 342 may be more prominently influenced by direct and continuous regional inputs, rather
 343 than by the natural sorption process on particulate organic carbon. This suggests that
 344 the Yangtze River has a great impact on the input of PAEs in these areas. Likewise, the
 345 Σ PAHs concentrations were significantly and positively correlated with OC-T in the

346 terrestrial OM-dominated area ($p < 0.01$, $r^2 = 0.97$), but had no relationship with OC-M
347 in this same area. Additionally, the Σ PAHs concentrations showed significant positive
348 correlations ($p < 0.01$, $r^2 = 0.88$) with the TOC content in the marine OM-dominated
349 area; this can be attributed to the correlation between Σ PAHs and OC-M ($p < 0.01$, r^2
350 $=0.89$) and that between Σ PAHs and OC-T ($p < 0.01$, $r^2 = 0.78$). Moreover, a significant
351 positive correlation was found between Σ PAHs and grain size in both the marine ($p <$
352 0.01 , $r^2 = 0.82$), and terrestrial ($p < 0.01$, $r^2 = 0.68$) OM-dominated areas. In comparison
353 to the distribution of PAEs, that of PAHs in the ECS surface sediments was influenced
354 by the continuous regional input of PAHs from the Yangtze River (especially in the
355 inshore region, which is a terrestrial OM-dominated area) and by the sediment
356 geochemistry (particularly in the offshore region, which is a marine OM-dominated
357 area). Previous soil studies have demonstrated that strong relationships between
358 contaminant and organic carbon concentrations in the soil could occur under a number
359 of scenarios; one of these scenarios consists in a stable chemistry that does not allow
360 degradation (Sweetman et al., 2005). Similarly, relatively persistent PAHs could have
361 achieved steady-state conditions between seawater and sediment on a timescale,
362 strengthening the correlation between PAH and sediment organic carbon concentrations.

363 **4.4. Identification of the PAE and PAH sources**

364 As shown in Figure S1, the dominant PAE species was DEHP, followed by DIBP
365 and DBP. These results are consistent with those reported in previous studies, in which
366 DBP, DIBP and DEHP were the dominant PAEs components of the sediment (Li et al.,
367 2017; Zhang et al., 2018a). According to previous reports, DEHP, DIBP, and DBP are
368 the most widely used PAE plasticizers (Sun et al., 2013). Furthermore, DEHP and DBP
369 are the two main PAEs used in the manufacturing of mulch film, which is commonly
370 used in agricultural production (Xu and Liu, 2013). DMP, DEP, DIBP and DBP are also

371 used in cosmetics and personal care products; moreover, DIBP and DBP are used in
372 epoxy resins, cellulose esters and special adhesive formulations (Zeng et al., 2008). The
373 DEHP constituents represent about 50–60% of the total commercial PAEs being
374 produced (Zeng et al., 2008). Consequently, the high DEHP values reported in the
375 marine sediments of this study are not surprising.

376 Generally, high molecular weight PAHs are found in higher concentrations than
377 low molecular weight PAHs in marine sediments (Huang et al., 2012). As shown in
378 Figure S2, 4-ring PAHs were the most abundant compounds, followed by 3- or 5-ring
379 PAHs in most of the ECS stations. Based on the characteristics of PAH composition,
380 diagnostic ratios of PAHs isomers can be used to distinguish the probable sources of
381 the PAHs in the marine environment. The isomeric ratios of the selected compounds
382 have been considered good indicators of PAHs source. For example, $\text{Ant}/(\text{Ant} + \text{Phe}) <$
383 0.10 primarily indicates petroleum contamination, while $\text{Ant}/(\text{Ant} + \text{Phe}) >$ 0.10
384 indicates that the contaminants come from a combustion source, $\text{Flu}/(\text{Flu} + \text{Pyr}) <$ 0.40
385 primarily indicates petroleum contamination, while values between 0.40 – 0.50 and $<$
386 0.50 indicate mixed and combustion sources, respectively (Yunker et al., 2002). When
387 $\text{BaA}/(\text{BaA} + \text{Chr}) >$ 0.35 , it implies combustion sources, while values between 0.20 –
388 0.35 and $<$ 0.20 indicate mixed and petroleum sources, respectively (Yunker et al.,
389 2002). As shown in Table 3, the ratios of $\text{Ant}/(\text{Ant} + \text{Phe})$ in the ECS sediments ranged
390 between 0.00 – 0.15 ; moreover, the sampling sites located inshore (value $<$ 0.10) and
391 offshore (value $>$ 0.10) indicated two distinct PAH source: petrogenic and combustion
392 sources, respectively. The ratios of $\text{Flu}/(\text{Flu} + \text{Pyr})$ in the ECS surface sediments ranged
393 between 0.31 – 0.53 ; indicating mostly a combustion source. The ratios of
394 $\text{BaA}/(\text{BaA} + \text{Chr})$ fall between 0.45 – 0.70 , indicating a combustion source for all the
395 sampling sites. Overall, these results suggest that pyrogenic PAHs can be important

Table 3 Isomeric ratios of Ant/(Ant + Phe), Flu/(Flu+Pyr) and BaA/(BaA + Chr)

Site	Ant/(Ant + Phe)	Flu/(Flu+Pyr)	BaA/(BaA + Chr)
A1	0.14	0.47	0.50
B1	0.15	0.48	0.50
B2	0.14	0.48	0.50
B3	0.14	0.49	0.48
B4	0.06	0.35	0.57
B5	0.08	0.32	0.57
C1	0.15	0.47	0.51
C2	0.07	0.45	0.60
C3	0.08	0.45	0.59
C4	0.07	0.45	0.62
D1	0.07	0.46	0.56
D2	0.07	0.46	0.56
D3	0.06	0.48	0.56
D4	0.17	0.45	0.59
E1	0.20	0.48	0.45
E2	0.18	0.48	0.45
F1	0.10	0.39	0.51
F2	0.10	0.39	0.50
F3	0.08	0.38	0.51
H1	0.13	0.45	0.57
H2	0.14	0.48	0.56
H3	0.15	0.48	0.59
N1	0.00	0.44	0.70
N2	0.00	0.44	0.69
N3	0.00	0.44	0.57
Q1	0.10	0.52	0.53
Q2	0.11	0.52	0.50
Q3	0.12	0.48	0.55
Q4	0.14	0.50	0.55

397

398 **5. Conclusion**

399 In this work, we analyzed the concentrations and compositions of 6 PAEs and 16 PAHs
400 contained in surface sediments collected from the ECS. The Yangtze River input is an
401 important factor affecting the distribution of PAEs and PAHs in the sediments of the
402 ECS. The \sum PAEs and \sum PAHs concentrations decreased gradually from inshore to
403 offshore and from north to south; their maximum values were registered in the

404 southeastern part of the Yangtze River estuary, indicating that source proximity is
405 another factor influencing the distribution of PAEs and PAHs within the ECS.
406 Sedimentary geochemistry also affected to the distribution PAHs in the ECS sediments,
407 but not that of the PAEs. Finally, DEHP was found to be the major source of PAEs,
408 while pyrogenic PAHs may be an important source of PAHs in the ECS.

409

410

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417

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