

Comprehensive Insights Into the Occurrence, Source, Distribution and Risk Assessment of Polycyclic Aromatic Hydrocarbons in a Large Drinking Reservoir System

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26 **Abstract**

27 The resource, environment, and ecological value of drinking reservoirs have received
28 widespread concerns due to the pollution of persistent organic pollutants such as
29 polycyclic aromatic hydrocarbons (PAHs). Therefore, we comprehensively studied the
30 occurrence, source, distribution and risk assessment of representative PAHs in
31 Fengshuba Reservoir (FSBR) (large drinking reservoir, China). The total concentrations
32 of 16 USEPA PAHs in the water phase, porewater phase, sediment phase and soil phase
33 were in ranges of 109.72-393.19 ng/L, 5.75-35.15 µg/L, 364.4-743.71 µg/kg and
34 367.81-639.89 µg/kg, respectively. The naphthalene (Nap) was the dominant PAHs in
35 the water phase, while it was Nap and phenanthrene (Phe) in porewater, sediment and
36 soil phase. The main sources of PAHs in FSBR were biomass combustion. Redundancy
37 analysis indicated that, the NTU, NO₂⁻, NH₄⁺, Chl- α and IC were the dominant factor
38 influencing the PAHs distribution in water phase and the PAHs in sediment phase was
39 affected by T and NO₃⁻. Pseudo-partitioning coefficients indicated that the PAHs in the
40 porewater phase was more likely to migrate to the sediment phase. Risk assessment
41 indicated that the PAHs both in the water and sediment phases were generally in a Low-
42 risk state, while the PAHs in the soil phase were in a Moderate-risk state and the Nap
43 was in a High-risk state, and exposure to the PAHs in FSBR through drinking and skin
44 exposure had little impact on consumers' health. In summary, Nap could be used as a
45 key indicator to evaluate the existence and potential risk of PAHs in FSBR.

46 **Key Word:** Polycyclic aromatic hydrocarbons; Fengshuba Reservoir; Occurrence;
47 Risk assessment; Indicator.

48 **1. Introduction**

49 Polycyclic aromatic hydrocarbons (PAHs) are persistent pollutants that contain two
50 or more fused rings (Cao et al., 2017, El-Saeid et al., 2015), and are widely present in
51 various environmental media (Han et al., 2019, Saeedi et al., 2012, Mojiri et al., 2019,
52 Bianco et al., 2020). Due to their carcinogenicity, teratogenicity and mutagenicity and
53 the ability to migrate over long distances (He et al., 2020, Tobiszewski and Namiesnik,
54 2012), PAHs has attracted much attention for the harm to ecological environment and
55 human health (Lu et al., 2019). The United States Environmental Protection Agency
56 (USEPA) lists 16 kinds of PAHs as priority pollutants (Keith and Telliard, 1979), and
57 the International Agency for Research Cancer (IARC) lists many PAHs such as benzo
58 (a) pyrene (Bap) as carcinogenic pollutants. The sources of PAHs include natural
59 processes such as forest fires, as well as anthropogenic processes such as incomplete
60 combustion of fossil fuels and biomass (Dat and Chang, 2017), which can enter the
61 natural environment such as water body through a variety of ways (Palm et al., 2004).
62 With the world's socio-economic development and increasing demand for energy, the
63 high emissions of PAHs pose an increasing threat to global health (Idowu et al., 2019,
64 Kim et al., 2013).

65 The PAHs in the water environment tend to adsorb in the sediment due to the high
66 PAHs stability, low solubility and strong hydrophobicity (Doong and Lin, 2004). At the
67 same time, due to the frequent substances exchange between the sediment phase and
68 the aqueous phase (including the water and porewater phase), the adsorbed PAHs in
69 sediment phase will also be released into water for biological utilization, resulting in

70 secondary pollution and harm to ecological environment and human health (Hussain et
71 al., 2016, Yim et al., 2007, Han et al., 2020). The importance of drinking water
72 reservoirs to human beings is self-evident. However, the reports of PAHs in the water
73 environment are mainly focused on rivers (Doong and Lin, 2004, Sun et al., 2009),
74 lakes (Li et al., 2017, Qiao et al., 2006) and oceans (Araghi et al., 2014, Yuan et al.,
75 2015). There are few comprehensive studies on PAHs in reservoir systems, especially
76 large drinking water reservoirs (Jiao et al., 2009, Seopela et al., 2020) and the soil phase
77 of the water level drop zone of the reservoir. The Fengshuba Reservoir (FSBR) is
78 located at the source of the Dongjiang River Basin in southern China, providing
79 drinking water sources for nearly 40 million people in surrounding cities (Longchuan,
80 Guangzhou and Shenzhen). With the rapid economic development of Dongjiang River
81 Basin (He et al., 2013), the FSBR may be polluted by PAHs, which will threaten the
82 ecological environment and human health in the basin (Zheng et al., 2013). Hence,
83 assessing the occurrence and risk levels of PAHs in the FSBR will become crucial.

84 The purpose of this study is to systematically explore the occurrence, source, and
85 distribution of 16 USEPA PAHs in water phase, porewater phase, sediment phase and
86 soil phase in FSBR, and at the same time to evaluate the risks of the PAHs in water
87 phase and solid phase of the reservoir to ecology and human health. The relationship
88 between the PAHs and environmental parameters in different environmental media, and
89 finally determined the potential applicability of this potential indicator in the
90 management and control of the PAHs pollution in drinking water reservoirs. These
91 studies contribute to a better understanding of the PAHs environmental behavior in

92 reservoir systems and are expected to enrich the study of the PAHs in the water
93 environment.

94 **2. Materials and Methods**

95 **2.1 Study area and sampling**

96 The main water collecting sources of FSBR are Beiling River (basin area: 2363 km²)
97 and Xinwu River (basin area: 2697 km²). The water surface area of FSBR is about 16.45
98 km², and the water depth is between 70 m and 100 m (Chen et al., 2017). [Table S1](#)
99 showed the hydrological information of the reservoir in detail. Samples were taken from
100 five sampling sites (S1-S5) from July 18 to 20, 2015 (summer), November 25 to 27,
101 2015 (autumn) and March 14 to 16, 2016 (spring), respectively. For the water phase, 3
102 L of water samples were taken from the surface (50 cm below the water surface), the
103 middle (half of the water depth) and the bottom (2-4 m from the bottom), respectively.
104 And then the three water samples were mixed to form 9 L of water samples (S1-S5) at
105 the point. For sediment phase, a gravity sedimentation sampler was used to take
106 sediment samples (S1-S5) about 15 cm from the bottom of the water. In addition, only
107 in March 2016 (spring), when the water yield of the reservoir increased, that is, the
108 water level of the reservoir decreased, the surface soil samples (15 cm below the surface)
109 were taken from the water level drop zone at the same sampling point (S1-S5) (Chen et
110 al., 2020). In order to minimize sampling errors, samples were collected three times at
111 each sampling site. In addition, the samples were immediately transferred to the
112 laboratory after collection and stored in acidic plastic drums at -20 °C.

113 **2.2 Chemicals and standard products**

114 This study involves the detection and quantification of 16 representative PAHs listed
115 by USEPA. PAHs Standards were classified according to the number of rings: 2-ring
116 PAHs (naphthalene (Nap)), 3-ring PAHs (acenaphthylene (Acy), acenaphthene (Ace),
117 fluorene (Flo), phenanthrene (Phe), anthracene (Ant)), 4-ring PAHs (fluoranthene (Fla),
118 pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr)), 5-ring PAHs
119 (benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP),
120 dibenz[a,h]anthracene (DahA)), 6-ring PAHs (indeno[1,2,3-cd]pyrene (IcdP),
121 benzo[g,h,i]perylene (BghiP)). Standard Solution of the PAHs Isotope Tracer:
122 naphthalene-d8 (Nap-d8), acenaphthylene-d10 (Ace-d10), phenylene-d10 (Phe-d10)
123 and pyrene-d12 (Pyr-d12) were soluble in Methanol. All the abovementioned
124 compounds were purchased from Dr.Ehrenstorfer (Augsburg, Germany). Quantitative
125 internal standard: Anthracene-d10 was purchased from German DR Company as the
126 quantitative internal standard on machine. All the organic reagents in this experiment
127 were chromatography-grade, and the solid phase extraction HC-C18 column was
128 purchased from Shanghai Anpu Experimental Scientific Instrument Co., Ltd. The
129 physical and chemical properties of these 16 USEPA PAHs were listed in [Table S2](#).

130 **2.3 Environmental Factor Detection**

131 The sediment samples were centrifuged at 5000 rpm for 10 min and the supernatant
132 was collected. Porewater is known as the filtrate obtained by filtered the supernatant
133 through a 0.45 μm glass fiber filter. Using Hydrolab multi-parameter water quality
134 analyzer (Hash, USA), the temperature (T), oxidation reduction potential (ORP),
135 dissolved oxygen (DO), pH, electrical conductivity (EC), chlorophyll- α (Chl- α) and

136 turbidity of the water samples and porewater samples were measured. Other
137 environmental parameters were measured according to the national standard methods:
138 dissolved total nitrogen (DTN), dissolved organic carbon (DOC), total phosphorus. In
139 addition, the sediment and soil samples were freeze-dried, and then ground and passed
140 through 100-mesh sieve. According to other studies, the basic environmental
141 parameters of the solid phase are T, EC, ORP, pH, total organic carbon (TOC), total
142 nitrogen (TN), total phosphorus (TP), and the percentage of clay, sand and silt.

143 **2.4 Sample handling and analysis**

144 **2.4.1 Solid phase extraction of water samples**

145 The water samples (1 L in triplicate) were filtered by 0.45 μm aqueous phase filter
146 membrane, and the filtrate was transferred into brown bottles, and 5 mL of methanol
147 was added to each bottle, and then a certain amount of standard solution of isotope
148 tracer was added and fully mixed before use. 5 mL of N-hexane: Dichloromethane (1:1),
149 5 mL of methanol, and 5 mL of distilled water were respectively used to activate the
150 HC-C18 column for 3 min. After connected the large-capacity sampling tube,
151 maintained the flow rate of 3-5 mL/min for water sample enrichment. After sample
152 enrichment was completed, the inner wall rinsed by ultrapure water and continued the
153 enrichment. and then use the vacuum pump to drain it after the completion 30min. Then,
154 use 5 mL of n-hexane: dichloromethane (1:1), and 5 mL of dichloromethane were used
155 for elution successively, and the elution was performed for about 3 minutes each time.
156 The eluent was collected in the test tube. Then, when the eluent was concentrated to
157 about 1 mL with a nitrogen blower, transferred to a quantitative concentration tube, and

158 added 3 mL of n-hexane: acetone (1:1) as the conversion solvent, continued to use the
159 nitrogen blower to concentrate to about 0.3 mL, added 50 μ L internal standard
160 anthracene-d10 (800 μ g/L) made the final concentrated solution 80 μ g/L, and finally
161 calibrated to 0.5 mL with n-hexane: acetone (1:1), and then transferred to a brown
162 machine vial for later use.

163 **2.4.2 Solid phase extraction of water samples**

164 For the solid samples (sediment and soil samples), first freeze-dried for 24 hours,
165 ground them in a mortar and pass through a 100-mesh sieve, and then collected them
166 for later use. Then, took 1 g of the treated samples and 5 g of diatomite and mixed them
167 thoroughly, and put them into a 0.45 μ m glass fiber membrane accelerated solvent
168 extraction cell, a certain amount of internal tracer markers was added and left overnight
169 before an accelerated solvent extractor was added. Then put it into an accelerated
170 solvent extraction instrument for extraction. Extraction conditions: the extraction
171 solvent was dichloromethane: acetone: n-hexane (1:1:1, V/V/V) mixed solution,
172 pressure 1500 psi, temperature 100°C, 5 min of preheating equilibrium time, 6 min of
173 static extraction time, the elution volume was 60% of the volume of the extraction tank,
174 the nitrogen purge time was 60 s, and 2 cycles of extraction process. Finally, the
175 extracted solution was subjected to the same concentration process as the above water
176 sample.

177 **2.4.3 GC-MS instrumental analysis conditions**

178 Gas chromatographic conditions: the temperature of the injection port was 280 °C,
179 and splitless sampling; the carrier gas was high-purity helium with a flow rate of 1.0

180 mL/min; heating program: kept at 60 °C for 2 min, then increased to 210 °C at the rate
181 of 15 °C/min and kept for 2 min, and increased to 300 °C at the rate of 3 °C/min and
182 kept for 2 min, and ran at 315 °C for 5 min. Mass spectrometry conditions: the
183 temperature of the quadrupole was 150 °C, the temperature of the ion source was 230 °C,
184 and the temperature of the transmission line was 300 °C, and the ionization energy was
185 70 eV. Data collection method: adopt ion scanning mode, in which the solvent delay
186 time was 5 minutes, the peak time of the target compound, the quantification ion and
187 the qualifier ion were listed in [Table S3](#).

188 Establishment of standard curve: with reference to the above conditions of instrument
189 analysis within the standard method of quantitative, and established standard curves of
190 internal standard method. The quantitative analysis of water samples: the sample was
191 carried out according to the above-mentioned instrument analysis conditions, and
192 quantitative analysis using internal standard method with the curve, and used the tracer
193 recovery rate as the correction factor, and divided by the concentration factor of 2000,
194 which was the actual concentration (ng/L) of water samples. The quantitative analysis
195 of solid samples: samples with reference to the above analysis instrument analysis
196 conditions. The internal standard method was used to mark the curve for quantitative
197 analysis, and the tracer recovery was used as the correction factor. The concentration in
198 the sample divided by the concentration ratio was the actual concentration in the
199 sediment (µg/kg).

200 **2.5 Sources of PAH**

201 A large number of studies have shown that the ratio isomers of the PAHs is of great

202 significance for study the source of the PAHs (Hartmann et al., 2004, Zhang et al., 2006,
203 Yunker et al., 2002, Sarria-Villa et al., 2016). These ratios such as Fla/(Fla + Pyr),
204 Phe/Ant, Nap/Phe, Ant/(Ant + Phe), Fla/(Fla + Pyr), InP/(InP + BghiP) can be used to
205 study the sources of the PAHs in water, sediment and soil (Budzinski et al., 1997,
206 Tobiszewski and Namiesnik, 2012). Since no high-ring PAHs such as IcdP and BghiP
207 were detected in the water phase, the ratio of Fla/(Fla + Pyr) and Ant/(Ant + Phe) were
208 used to study the main sources of water and solid phase of FSBR. The ratio of Fla/(Fla
209 +Pyr) < 0.4 (De La Torre-Roche et al., 2009) indicates that petroleum sources are
210 dominant, and the ratio of Flu/(Flu +Pyr) between 0.4 to 0.5 indicates that liquid
211 petrochemical combustion is dominant, and the ratio of Fla/(Fla +Pyr) > 0.5 indicates
212 that biomass burning such as grass, wood, and coal is dominant. The ratio of Ant/(Ant
213 + Phe) < 0.1 (Pies et al., 2008) indicates that the petroleum source is dominant, and the
214 ratio of Ant/(Ant + Phe) > 0.1 indicates that the pyrolysis source is dominant.

215 **2.6 Risk assessment**

216 Ecological risk assessment: the risk quotient (RQ) method was used to evaluate the
217 ecological toxicity of the PAHs in the FSBR (Cao et al., 2010, Zhang et al., 2017, Yu et
218 al., 2021). The risk level of the PAHs was characterized by the RQ, which was defined
219 as follows:

$$220 \quad RQ = \frac{C_{PAHs}}{C_{Qv}} \quad (1)$$

221 C_{PAHs} and C_{Qv} in the above formula were expressed as the actual concentration of
222 certain PAHs in each medium and the corresponding quality value of certain PAHs in
223 each medium. The ratio of the actual concentrations (ACs) of the PAHs to the negligible

224 concentrations (NCs) and maximum permissible concentrations (MPCs) of the PAHs
 225 in the corresponding medium was used to evaluate the risk level of the PAHs in water,
 226 sediment and soil phase, which was defined as follows:

$$227 \quad RQ_{NCs} = \frac{C_{ACs}}{C_{NCs}} \quad (2)$$

$$228 \quad RQ_{MPCs} = \frac{C_{ACs}}{C_{MPCs}} \quad (3)$$

229 Furthermore, according to the current research, the following formulas (4), (5) and
 230 (6) were used to calculate $RQ_{\Sigma PAHs}$, $RQ_{\Sigma PAHs (NCs)}$ and $RQ_{\Sigma PAHs (MPCs)}$ in various
 231 environmental medium, which were defined as follows:

$$232 \quad RQ_{\Sigma PAHs} = \sum_{i=1}^{16} RQ \quad (RQ > 1) \quad (4)$$

$$233 \quad RQ_{\Sigma PAHs(NCs)} = \sum_{i=1}^{16} RQ_{NCs} \quad (RQ_{NCs} > 1) \quad (5)$$

$$234 \quad RQ_{\Sigma PAHs(MPCs)} = \sum_{i=1}^{16} RQ_{MPCs} \quad (RQ_{MPCs} > 1) \quad (6)$$

235 Health risk assessment: the incremental lifetime cancer risk (ILCRs) recommended
 236 by USEPA combined with toxic equivalent of BaP (TEQ_{BaP}) was used to quantitatively
 237 characterize the potential carcinogenic risk of human body by the PAHs in the water of
 238 FSBR (Nadal et al., 2004, Meng et al., 2019, Yu et al., 2021). Human body are mainly
 239 exposed to the PAHs in surface water and groundwater bodies through ingestion and
 240 skin contact, resulting in potential carcinogenic risks. The calculation formulas (3), (4)
 241 and (5) are as follows:

$$242 \quad TEQ_{BaP} = \sum_{i=1}^{16} C_i \times TEF \quad (7)$$

$$243 \quad ILCRs_{ingestion} = \frac{TEQ_{BaP} \times IRW \times CSF_i \times EF \times ED}{BW \times AT} \quad (8)$$

$$244 \quad ILCRs_{skin \ contact} = \frac{TEQ_{BaP} \times SA \times K_p \times ABS \times ET \times CSF_i \times EF \times ED}{BW \times AT} \quad (9)$$

245 where TEQ_{BaP} represents the toxicity equivalent concentration (ng/L), $ILCRs_{ingestion}$

246 represents the incremental lifetime cancer risk caused by ingestion and $ILCR_{S_{skin\ contact}}$

247 represents the incremental lifetime cancer risk caused by contact.

248 **2.7 Data analysis**

249 Before the statistical analysis of the data, first of all, SPSS 26.0 (IBM, USA) Shapiro-
250 Wilk test was used to check whether the measured data met the normal distribution, and
251 then the One-way ANOVA test (for normal distribution) or the Kruskal-Wallis test (for
252 non-normal distribution) were used to analyze the inherent correlation of the data. Use
253 Origin 9 software (Originlab, USA) and R language (Microsoft, USA) for data
254 visualization. Use Canoco 5 (Microcomputer Power, UBA) software and R language
255 (Microsoft, USA) to perform redundancy analysis (RDA) and ANOSIM analysis on the
256 measured data of PAHs. If P value is involved, then $P \leq 0.05$ or $P \leq 0.01$ indicates that
257 the test results of the measured data were statistically significant.

258 **3. Result and discussions**

259 **3.1 Distribution of the PAHs**

260 **3.1.1 The PAHs in aqueous phase**

261 The concentrations of 16 PAHs in the water samples from FSBR were shown in the
262 [Fig. 2 \(A\)](#) and [Table S4](#). Among the 16 PAHs, 6 of the PAHs were not detected (5-ring:
263 BbF, BkF, BaP, IcdP; 6-ring: DahA, BghiP), the detection rate of BaA was 50%, and
264 the detection rate of the remaining PAHs were 100%. The total concentrations of 10
265 remaining PAHs ($\sum 10PAHs$) ranged from 110.04 ng/L to 389.88 ng/L, with an average
266 of 213.14 ng/L and a median of 198.37 ng/L. Therefore, it can be seen from the results
267 that the FSBR as a source of drinking water had been contaminated. Among these 10

268 PAHs, the concentration of Nap ranged from 62.09 to 256.44 ng/L (average: 125.72
269 ng/L, median: 138.44 ng/L), which was significantly higher than others, indicating that
270 Nap abound in the reservoir. The possible reason was that Nap has the highest solubility
271 in water (Tobiszewski and Namiesnik, 2012). In addition, the 5-ring and 6-ring PAHs
272 were not detected in any water samples because of their extremely low solubility in
273 water, which makes it easier for high-ring PAHs to be enriched in the solid phase. This
274 phenomenon also occurs in the Moscow River, Russia water body, which the high-ring
275 PAHs were not detected (Eremina et al., 2016). Comparing with other studies, the total
276 concentration of the PAHs in the water phase of FSBR was similar to the total
277 concentration of the PAHs in Luan River Basin, China (Σ PAHs: 9.75-309.75 ng/L) (Li
278 et al., 2010), which was higher than that in River Tiber, Italy (23.9-72.0 ng/L)
279 (Patrolecco et al., 2010), Ili-Balkhash Basin, arid Central Asia (7.58-70.98 ng/L) (Shen
280 et al., 2021), and lower than that in Loskop dam, South Africa (0.150-49.8 μ g/L)
281 (Seopela et al., 2020).

282 In porewater samples (Fig. 2 (B) and Table S4), except for the detection rate of IcdP
283 and DahA at 93.3%, the detection rate of the remaining 14 PAHs was 100%. The total
284 concentrations of the 16 USEPA the PAHs (Σ 16PAHs) ranged from 5.75 μ g/L to 34.96
285 μ g/L, with an average of 18.33 μ g/L and a median of 18.56 μ g/L. These data were
286 significantly higher than those in the water phase, mainly because of sediments. The
287 sediment phase was also a sink of the PAHs in the water body, and the porewater was
288 obtained by centrifugation of the sediment phase (Bigus et al., 2014, Guo et al., 2017).
289 In fact, this was consistent with what has been reported by other studies (Maskaoui et

290 al., 2002). Similar to the aqueous phase, the concentration of Nap was also the highest
291 (average: 9.42 µg/L, median: 10.16 µg/L), followed by Phe (2.80 µg/L, 2.88 µg/L) in
292 porewater phase of FSBR. Looking at the water and porewater phase, it can be inferred
293 that the PAHs in the water phase were an important source of PAHs in the porewater
294 phase. Comparing with other studies, the total concentration of the PAHs in the
295 sediments of FSBR was higher than that in Lake Taihu, China (Σ 16PAHs: 2.01-19.90
296 µg/L) (Zhang et al., 2011), and lower than that in Lanzhou Reach of Yellow River,
297 China (48.2-206 µg/L) (Yu et al., 2009).

298 **3.1.2 PAHs in solid phase**

299 As shown in Fig. 2 (C) and Table S4, all 16 PAHs were detected in all sediment
300 samples. The total concentrations of these 16 PAHs (Σ 16PAHs) ranged from 364.4
301 µg/kg to 743.71 µg/kg, with an average of 506.81 µg/kg and a median of 470.22 µg/kg.
302 Similar to the occurrence of the PAHs in the porewater phase, the concentration of Nap
303 (114.43 µg/kg, 120.77 µg/kg) and Phe (112.26 µg/kg, 74.57 µg/kg) were significantly
304 higher than the others. Due to the occurrence of the PAHs in aqueous phase, Nap and
305 Phe may have a tendency to accumulate from aqueous phase to sediment phase (Han et
306 al., 2020). Comparing with other studies, the total concentration of the PAHs in the
307 sediments of FSBR higher than that in Chenab River, Pakistan (Σ 16PAHs: 7.41-600
308 µg/kg) (Hussain et al., 2016), Gorgan Bay, Caspian Sea (107.87-516.18 µg/kg) (Araghi
309 et al., 2014), and lower than that in Dianchi Lake, China (210-11070 µg/kg) (Zhao et
310 al., 2014) and Loskop dam, South Africa (61.6-2618 µg/kg) (Seopela et al., 2020).

311 For the solid phase (Fig. 2 (D) and Table S4), the 16 PAHs were detected in all

312 samples. The total concentration of these 16 PAHs ($\Sigma 16\text{PAHs}$) ranged from 367.81
313 $\mu\text{g/kg}$ to 639.89 $\mu\text{g/kg}$, with an average of 485.18 $\mu\text{g/kg}$ and a median of 453.29 $\mu\text{g/kg}$.
314 Comparing with the sediment, the overall occurrence level of PAHs in the soil phase in
315 the drawdown area was similar to that in the sediment phase. The distribution of the
316 PAHs in soil phase of FSBR was closely related to human activities (Li et al., 2020).
317 There are more intensive human activities at S5, including agriculture, fishing and fish
318 breeding, while the human activities at S4 were lesser. Among them, the point with the
319 highest concentration was S5 (528.18 $\mu\text{g/kg}$), and the point with the lowest
320 concentration was S4 (400.33 $\mu\text{g/kg}$). Similar to the occurrence of the PAHs in
321 porewater and sediment samples, the Nap (170.25 $\mu\text{g/kg}$, 154.92 $\mu\text{g/kg}$) and Phe
322 (108.43 $\mu\text{g/kg}$, 100.98 $\mu\text{g/kg}$) have the highest concentration. It can be seen from the
323 results that the occurrence of Nap and Phe in soil phase was the same as that of Nap
324 and Phe in water and sediment phase, which also proves that PAHs are more likely to
325 be deposited in solid phase in water environment (Doong and Lin, 2004). However,
326 since there are few reports on the occurrence of PAHs in the soil in the drawdown area
327 of reservoirs, the results of this study cannot be compared with those of other studies.
328 At the same time, it also reflects that the pollution of PAHs in soil in water-leveling and
329 fluctuating areas is worthy of further study, especially for those large drinking water
330 reservoirs.

331 **3.2 Sources of PAHs**

332 For the water phase, from [Fig. 3 \(A\)](#) and [Table S5](#), the ratio of Fla/(Fla + Pyr) in the
333 water samples was 0.54-0.63 > 0.5, indicating that the PAHs in the water phase were

334 mainly derived from grass, wood and coal combustion, and the ratio of Ant/(Ant + Phe)
335 in the water samples was 0.03-0.22, indicating that the PAHs in the water phase also
336 came from combustion of petroleum. For the porewater phase, according to Fig. 3 (B)
337 and Table S5, the ratio of Fla/(Fla + Pyr) in the porewater samples was 0.58-0.70 > 0.5,
338 indicating that the PAHs in the porewater phase were mainly derived from the grass,
339 wood and coal combustion and the ratio of Ant/(Ant + Phe) in the porewater samples
340 was almost all < 0.1, indicating that PAHs in the porewater phase were also mainly
341 derived from petroleum sources. For the sediment phase (Fig. 3 (C) and Table S5), the
342 ratio of Fla/(Fla + Pyr) in sediment samples was 0.52-0.57 > 0.5, indicating that the
343 PAHs in the sediment phase were derived from the grass, wood and coal combustion
344 and the ratio of Ant/(Ant + Phe) in the sediment samples was 0.82-0.94 > 0.1, indicating
345 that the PAHs in the sediment phase were mainly derived from combustion. For the soil
346 phase, according to Fig. 3 (D) and Table S5, the ratio of Flu/(Flu + Pyr) in soil samples
347 was 0.46-0.56, indicating that the PAHs in the soil phase were mainly derived from the
348 mixed sources of petroleum combustion and grass, wood and coal combustion, and the
349 ratio of Ant/(Ant + Phe) in the soil samples were 0.88-0.96 > 0.1, indicating that PAHs
350 in the soil phase were also mainly derived from combustion. In conclusion, the PAHs
351 in FSBR were mainly derived from biomass combustion sources such as grass, wood
352 and coal, which may be due to the fact that FSBR is located in the natural environment
353 area at the source of Dongjiang River, far away from mega-cities such as Guangzhou,
354 Shenzhen and Hong Kong, and the forest coverage rate of Dongjiang River Basin
355 reached 71.8% in 2009 (Ding et al., 2016). Compared with the four water bodies' bank

356 in Jamshedpur, India, where urbanization was accelerating, the main source of PAHs in
357 water bodies was related to combustion and diagenesis (Ambade and Sethi, 2021), and
358 for the Lake Ijssel, the Netherlands (Niu and van Gelder, 2020), its water source mainly
359 comes from the Rhine River and passes through big cities like Amsterdam, making the
360 PAHs in the lake mainly come from the coal combustion and petroleum source.

361 **3.3 Seasonal variations of PAHs in reservoirs**

362 For the water phase, the concentration of 2-ring PAHs in spring (191.45 ± 75.00 ng/L)
363 was significantly lower than the concentration of 2-ring PAHs in summer ($125.20 \pm$
364 11.46 ng/L) and autumn (96.64 ± 23.83 ng/L) (one-way ANOVA, $F = 5.42$, $P < 0.05$)
365 (Fig. 4 (A)). The concentration of 3-ring PAHs in autumn (38.79 ± 5.51 ng/L) was also
366 significantly lower than the concentration of 3-ring PAHs in summer (58.88 ± 3.35 ng/L)
367 and spring (63.39 ± 13.17 ng/L) (one-way ANOVA, $F = 11.96$, $P < 0.01$). The
368 concentration of 4-ring PAHs in summer (17.38 ± 1.47 ng/L) was lower than the
369 concentration of 4-ring PAHs in spring (23.25 ± 5.31 ng/L) (one-way ANOVA, $F = 5.61$,
370 $P < 0.05$). In addition, this distribution trend was similar to the order of local rainfall:
371 Autumn (28.5 mm) < Summer (218 mm) < Spring (316 mm) (Table S1). It also
372 indicated that season had a significant effect on the distribution of total concentration
373 of PAHs in the aqueous phase (Li et al., 2020).

374 For the PAHs in the porewater phase, the concentrations of 3-ring PAHs
375 hydrocarbons in spring (4.71 ± 0.81 $\mu\text{g/L}$) were lower than those in summer ($5.75 \pm$
376 0.22 $\mu\text{g/L}$) and autumn (5.60 ± 0.68 $\mu\text{g/L}$) (one-way ANOVA, $F = 4.14$, $P < 0.043$) (Fig.
377 3 (B)). The concentration of 6- ring PAHs in spring (0.04 ± 0.01 $\mu\text{g/L}$) was significantly

378 lower than that of 6-ring PAHs in summer ($0.19 \pm 0.12 \mu\text{g/L}$) and autumn (0.06 ± 0.03)
379 (one-way ANOVA, $F = 6.57$, $P < 0.05$). In addition, the variation trend of the total
380 concentration of the PAHs was similar to that of Hydraulic retention time (days) in
381 FSBR: 49 days in spring $<$ 122 days in summer $<$ 246 days in autumn (Table S1).

382 For the PAHs in sediments, the concentration of 2-ring PAHs in autumn ($88.26 \pm$
383 $29.30 \mu\text{g/kg}$) was significantly lower than that in spring ($114.43 \pm 24.45 \mu\text{g/kg}$) (one-
384 way ANOVA, $F = 6.10$, $P < 0.05$) (Fig3 (C)). The 3-ring PAHs concentrations in
385 summer ($253.98 \pm 53.14 \mu\text{g/kg}$) were significantly higher than those in autumn (124.16
386 $\pm 11.08 \mu\text{g/kg}$) and spring ($120.15 \pm 7.21 \mu\text{g/kg}$) (Kruskal-Wallis H, $P < 0.05$). The
387 concentrations of 6-ring PAHs in summer ($27.40 \pm 2.78 \mu\text{g/kg}$) were significantly
388 higher than those in spring ($22.88 \pm 1.83 \mu\text{g/kg}$) (one-way ANOVA, $F = 4.17$, $P < 0.05$).

389 It is worth noting that there were significant differences in the PAH composition
390 between the water phase and the pore water phase and the sediment and soil phases
391 (ANOSIM, $P < 0.01$). It was further confirmed that the environmental behavior of the
392 PAHs in storage systems with different environmental media was complex and diverse.
393 At the same time, this also emphasized that when facing the problem of the PAHs
394 pollution in large drinking water reservoirs, we should comprehensively consider the
395 impact of multiple environmental media (environmental factors) on their environmental
396 behavior.

397 **3.4 Relationship between environmental parameters and PAHs**

398 Redundancy analysis (RDA) is a commonly used multivariate analysis method that
399 can be used to analyze the relationship between two sets of variables (Takane and

400 Hwang, 2005). This study used RDA to further explore the relationship between
401 environmental parameters and the PAHs in various media of FSBR (Opere et al., 2020).
402 The blue arrows indicate specific PAHs, and the red arrows indicate specific
403 environmental parameters. And these environmental parameters are shown in detail in
404 [Table S5-S8](#).

405 For the water phase, the RDA results ([Fig. 5 \(A\)](#)) showed that 49.7% of the changes
406 in PAHs were explained by these five significant environmental parameters: NTU
407 (24.0%, $P = 0.002$), NO_2^- (20.3%, $P = 0.004$), Chl- α (16.4%, $P = 0.002$), NH_4^+ (5.3%,
408 $P = 0.036$), IC (4.3%, $P = 0.05$). This result indicated that these environmental
409 parameters have a significant impact on the distribution of the PAHs in the water phase
410 of the FSBR. At the same time, it was worth noting that NTU and Chl- α were related
411 to turbidity, thus affecting the photodegradation of PAHs in water (Sanches et al., 2011).
412 The distribution of PAHs in the porewater phase was slightly affected by environmental
413 parameters, such as pH (4.0%, $P = 0.09$) and NO_3^- (3.0%, $P = 0.166$), and the P value
414 was greater than 0.05 for porewater phase ([Fig. 5 \(B\)](#)). For the sediment phase, the RDA
415 results showed ([Fig. 5 \(C\)](#)) that 26.8% of the changes in PAHs can be explained by
416 environmental parameters, such as T (35.6%, $P = 0.002$) and NO_3^- (17.9%, $P = 0.006$).
417 For the soil in the drawdown zone, the results of RDA showed ([Fig. 4 \(D\)](#)) that these
418 environmental parameters have little effect on the PAHs in the soil in the drawdown
419 zone of FSBR, but the changes in pH, DOC, TOC and NO_3^- had significant influence
420 on the distribution of Acy.

421 **3.5 Pseudo-partitioning coefficient calculation**

422 In order to better understand the potential dynamics between the PAHs in sediment
423 and aqueous phase (water and porewater phase), the pseudo-partition coefficient (*P-PC*)
424 method (Bai et al., 2014) was used. According to the following formula to explore their
425 distribution behaviors: $P-PC_{s,w}$ (sediment phase to water phase) = C_s/C_w , $P-PC_{s,p}$
426 (sedimentary phase to porewater phase) = C_s/C_p , where C_s , C_w and C_p refer to the
427 actual measured concentration of the PAHs in sediment phase ($\mu\text{g}/\text{kg}$), water phase
428 ($\mu\text{g}/\text{L}$) and porewater phase ($\mu\text{g}/\text{L}$), respectively. The data of $P-PC_{s,w}$ and $P-PC_{s,p}$
429 were shown in [Table S9 and S10](#).

430 At the same time, some studies have reported that the distribution of the PAHs
431 between sediment and aqueous phase may also be affected by basic environmental
432 factors (such as pH, TOC and DOC) (Ekpo et al., 2012, Nascimento et al., 2017). In
433 this study, Pearson analysis was used to explore the relationship between *P-PC* ($\text{Log}P-$
434 $PC_{s,w}$ and $\text{Log}P-PC_{s,p}$) and the level of pH in the corresponding system. In the
435 sediment to water phase system, only the value of Phe ($\text{Log}P - PC_{s,w}$) and the pH value
436 of water phase were positively correlated. It was indicated that the pH of water phase
437 may affect the distribution behavior of Phe between water phase and sediment phase,
438 while other target PAHs had no significant correlation with the level of pH. In the
439 sediment to porewater system, it can be seen that the values of Ant, Fla, Pyr, BkF, BaP
440 and DahA ($\text{Log}P - PC_{s,p}$) were positively correlated with the pH of the porewater phase,
441 while the values of Flo and Phe ($\text{Log}P - PC_{s,p}$) were negatively correlated with the pH
442 of porewater ([Fig. 6](#)), indicating that the pH of porewater may affect the distribution of
443 Ant, Fla, Pyr, BkF, BaP, DahA, Flo, and Phe between the porewater phase and the

444 sedimentary phase.

445 **3.6 Ecological and health risk assessment**

446 **3.6.1 Ecological Risk Assessment**

447 [Table S11](#) and [Fig. 7](#) showed the NCs and MECs of the 16 USEPA PAHs in different
448 environmental media, as well as the RQ_{NCs} , RQ_{MECs} and $RQ_{\Sigma PAHs}$ of the USEPA PAHs
449 in water phase, sediment phase and soil phase of FSBR. As can be seen from [Table S11](#),
450 [12](#) and [Fig. 7](#), RQ_{NCs} and RQ_{MECs} in the water samples were both significantly greater
451 than 1, and RQ_{MECs} were also significantly lower than 1. Meanwhile, the total RQ of
452 PAHs in the water phase of FSBR were found to be $RQ_{\Sigma PAH(NCs)} = 88.88$ and
453 $RQ_{\Sigma PAH(MECs)} = 0$, therefore, the PAHs in the water phase of FSBR was in a state of
454 Low-risk (Tobiszewski and Namiesnik, 2012). The value of RQ_{NCs} of Chr, BkF, BaP,
455 IcdP, BghiP in sediment samples were approximately 0 and the value of RQ_{MECs} were
456 almost 0, so their impact on the ecological environment was very low, while the other
457 PAHs were in a state of Moderate-risk. And the total RQ of the PAHs in the sediment
458 phase were found to be $RQ_{\Sigma PAH(NCs)} = 276.30$ and $RQ_{\Sigma PAH(MECs)} = 0$, so the PAHs of the
459 sediment phase in FSBR was in a state of Low-risk level. The value of RQ_{NCs} of Chr,
460 BkF, IcdP, and BghiP in the soil samples were approximately 0 and the value of RQ_{MECs}
461 were almost 0, which had a very low impact on the ecological environment, and other
462 PAHs were in a state of Moderate-risk in the ecological environment, except for Nap
463 ($RQ_{NCs} = 121.60$, $RQ_{MECs} = 1.22$) was in a High-risk state. And the total RQ of PAHs in
464 the soil phase of the drawdown zone were found to be $RQ_{\Sigma PAH(NCs)} = 324.22$ and
465 $RQ_{\Sigma PAH(MECs)} = 1.22$, therefore, the PAHs in the soil phase of FSBR was in a Moderate-

466 risk₁ state (Table S13). Compared with the FSBR in the nature reserve, the RQs study
467 of PAHs in coastline waters on Khark Island, SW Iran (Akhbarizadeh et al., 2016) that
468 is affected by the petroleum industry shows that PAHs are at a medium to high potential
469 risk. This also reflects from the side that it was very correct to list the FSBR as a
470 protected area (Chen et al., 2019).

471 3.6.2 Health risk assessment

472 According to equations (7), (8) and (9) and other parameters listed in Table S13, the
473 total carcinogenic risk (\sum ILCRs) of PAHs in the FSBR was $4.53 \times 10^{-8} \text{ a}^{-1}$ to 2.27×10^{-7}
474 a^{-1} , with an average of $9.34 \times 10^{-8} \text{ a}^{-1}$ and a median of $8.07 \times 10^{-8} \text{ a}^{-1}$. The carcinogenic
475 risk value (ILCR_{Sintake}) produced by direct ingestion was $4.53 \times 10^{-8} \text{ a}^{-1}$ to $2.27 \times 10^{-7} \text{ a}^{-1}$,
476 with an average of $9.34 \times 10^{-8} \text{ a}^{-1}$ and a median of $8.07 \times 10^{-8} \text{ a}^{-1}$; the carcinogenic risk
477 value (ILCR_{Sskin contact}) produced through skin contact was $1.30 \times 10^{-13} \text{ a}^{-1}$ to 6.53×10^{-13}
478 a^{-1} , with an average of $2.68 \times 10^{-13} \text{ a}^{-1}$ and a median of $2.32 \times 10^{-13} \text{ a}^{-1}$. By comparison, it
479 can be concluded that the risk of carcinogenesis through ingestion was about 100 times
480 that of skin exposure. The total carcinogenic risk (\sum ILCRs) of PAHs in the water of
481 FSBR was significantly lower than the carcinogenic risk recommended by USEPA (10^{-6} - 10^{-4} a^{-1}), indicating that the impact of PAHs on health risks was negligible. Compared
482
483 with the PAHs in the Shitoumen Reservoir, China (Sun et al., 2015) that posed a
484 potential carcinogenic risk to local residents, the PAHs in the water body of the FSBR
485 had almost no harm to the surrounding residents.

486 4. Conclusions

487 The total concentrations of 16 USEPA PAHs in water phase, porewater phase,

488 sediment phase and soil phase of FSBR ranged from 109.72 – 393.19 ng/L, 5.75 – 35.15
489 $\mu\text{g/L}$, 364.4 - 743.71 $\mu\text{g/kg}$, 367.81 - 639.89 $\mu\text{g/kg}$, respectively. The Nap was the
490 dominant PAHs in the water phase, while it was Nap and Phe were dominant in
491 porewater, sediment and soil phase. The main sources of PAHs in FSBR were grass,
492 wood, biomass and coal combustion. In addition, the distribution of the PAHs in the
493 water phase was affected by seasonal hydrological conditions, and Redundancy
494 analysis showed that, the PAHs in the water phase was also affected by environmental
495 parameters such as turbidity, NO_2^- , NH_4^+ , Chl- α and IC. The distribution of the PAHs
496 in sediment phase was affected by T and NO_3^+ , while the PAHs in porewater and soil
497 phase was slightly affected by environmental parameters. Pseudo-partitioning
498 coefficients indicated that the PAHs in the porewater phase were more likely to migrate
499 to the sediment phase. Risk assessment showed that the PAHs in the water and sediment
500 phases were generally in a Low-risk state, while the PAHs in the soil phase were
501 generally in a Moderate-risk₁ state and the Nap was in a High-risk state. The health risk
502 assessment indicated that exposure to the PAHs in FSBR through drinking and skin
503 exposure had little impact on consumers' health. In summary, Nap could be used as a
504 key indicator to evaluate the existence and potential risk of PAHs in FSBR.

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510 **Declaration**

511 ● Ethics approval and consent to participate
512 Not applicable

513

514 ● Consent for publication
515 Not applicable

516

517 ● Availability of data and materials

518 1. The datasets generated during and/or analysed during the current study are
519 available from the corresponding author on reasonable request.

520 2. All data generated or analysed during this study are included in this published
521 article (and its supplementary information files).

522

523 ● Competing interests

524 The authors declare that they have no competing interests.

525

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530

531 ● Authors' contributions

532 Yiliang He and Kangping Cui conceived of the presented idea. Material preparation,
533 data collection and analysis were performed by Yihan Chen. The first draft of the
534 manuscript was written by Xiangyang Xu. Xing Chen, Zhi Guo, Hongjie Chen and
535 Guangwei Deng commented on previous versions of the manuscript. All authors
536 read and approved the final manuscript.

Reference:

- Akhbarizadeh, R., Moore, F., Keshavarzi, B. & Moeinpour, A. (2016) Aliphatic and polycyclic aromatic hydrocarbons risk assessment in coastal water and sediments of Khark Island, SW Iran. *Mar Pollut Bull*, **108** (1-2), 33-45. <https://doi.org/10.1016/j.marpolbul.2016.05.004>
- Ambade, B. & Sethi, S. S. (2021) Health Risk Assessment and Characterization of Polycyclic Aromatic Hydrocarbon from the Hydrosphere. *Journal of Hazardous, Toxic, and Radioactive Waste*, **25** (2). [https://doi.org/10.1061/\(ASCE\)HZ.2153-5515.0000586](https://doi.org/10.1061/(ASCE)HZ.2153-5515.0000586)
- Araghi, P. E., Bastami, K. D. & Rahmanpoor, S. (2014) Distribution and sources of polycyclic aromatic hydrocarbons in the surface sediments of Gorgan Bay, Caspian Sea. *Mar Pollut Bull*, **89** (1-2), 494-498. <https://doi.org/10.1016/j.marpolbul.2013.12.001>
- Bai, Y., Meng, W., Xu, J., Zhang, Y., Guo, C., Lv, J. & Wan, J. (2014) Occurrence, distribution, environmental risk assessment and source apportionment of polycyclic aromatic hydrocarbons (PAHs) in water and sediments of the Liaohe River Basin, China. *Bull Environ Contam Toxicol*, **93** (6), 744-751. <https://doi.org/10.1007/s00128-014-1390-7>
- Bianco, F., Race, M., Papiro, S. & Esposito, G. (2020) Removal of polycyclic aromatic hydrocarbons during anaerobic biostimulation of marine sediments. *Sci Total Environ*, **709**, 136141. <https://doi.org/10.1016/j.scitotenv.2019.136141>
- Bigus, P., Tobiszewski, M. & Namiesnik, J. (2014) Historical records of organic pollutants in sediment cores. *Mar Pollut Bull*, **78** (1-2), 26-42. <https://doi.org/10.1016/j.marpolbul.2013.11.008>
- Budzinski, H., Jones, I., Bellocq, J., Piérard, C. & Garrigues, P. (1997) Evaluation of sediment contamination by polycyclic aromatic hydrocarbons in the Gironde estuary. *Marine Chemistry*, **58** (1), 85-97. [https://doi.org/10.1016/S0304-4203\(97\)00028-5](https://doi.org/10.1016/S0304-4203(97)00028-5)
- Cao, H., Chao, S., Qiao, L., Jiang, Y., Zeng, X. & Fan, X. (2017) Urbanization-related changes in soil PAHs and potential health risks of emission sources in a township in Southern Jiangsu, China. *Sci Total Environ*, **575**, 692-700. <https://doi.org/10.1016/j.scitotenv.2016.09.106>
- Cao, Z., Liu, J., Luan, Y., Li, Y., Ma, M., Xu, J. & Han, S. (2010) Distribution and ecosystem risk assessment of polycyclic aromatic hydrocarbons in the Luan River, China. *Ecotoxicology*, **19** (5), 827-837. <https://doi.org/10.1007/s10646-010-0464-5>
- Chen, Y., Cui, K., Huang, Q., Guo, Z., Huang, Y., Yu, K. & He, Y. (2020) Comprehensive insights into the occurrence, distribution, risk assessment and indicator screening of antibiotics in a large drinking reservoir system. *Sci Total Environ*, **716**, 137060. <https://doi.org/10.1016/j.scitotenv.2020.137060>
- Chen, Y., Li, P., Huang, Y., Yu, K., Chen, H., Cui, K., Huang, Q., Zhang, J., Yew-Hoong Gin, K. & He, Y. (2019) Environmental media exert a bottleneck in driving the dynamics of antibiotic resistance genes in modern aquatic environment. *Water Res*, **162**, 127-138. <https://doi.org/10.1016/j.watres.2019.06.047>
- Chen, Y., Yu, K., Zhou, Y., Ren, L., Kirumba, G., Zhang, B. & He, Y. (2017) Characterizing spatiotemporal variations of chromophoric dissolved organic matter in headwater catchment of a key drinking water source in China. *Environ Sci Pollut Res Int*, **24** (36), 27799-27812. <https://doi.org/10.1007/s11356-017-0307-5>
- Dat, N. D. & Chang, M. B. (2017) Review on characteristics of PAHs in atmosphere, anthropogenic sources and control technologies. *Sci Total Environ*, **609**, 682-693. <https://doi.org/10.1016/j.scitotenv.2017.07.204>

- De La Torre-Roche, R. J., Lee, W. Y. & Campos-Diaz, S. I. (2009) Soil-borne polycyclic aromatic hydrocarbons in El Paso, Texas: analysis of a potential problem in the United States/Mexico border region. *J Hazard Mater*, **163** (2-3), 946-958. <https://doi.org/10.1016/j.jhazmat.2008.07.089>
- Ding, J., Jiang, Y., Liu, Q., Hou, Z., Liao, J., Fu, L. & Peng, Q. (2016) Influences of the land use pattern on water quality in low-order streams of the Dongjiang River basin, China: A multi-scale analysis. *Sci Total Environ*, **551-552**, 205-216. <https://doi.org/10.1016/j.scitotenv.2016.01.162>
- Doong, R. A. & Lin, Y. T. (2004) Characterization and distribution of polycyclic aromatic hydrocarbon contaminations in surface sediment and water from Gao-ping River, Taiwan. *Water Res*, **38** (7), 1733-1744. <https://doi.org/10.1016/j.watres.2003.12.042>
- Ekpo, B. O., Oyo-Ita, O. E., Oros, D. R. & Simoneit, B. R. (2012) Distributions and sources of polycyclic aromatic hydrocarbons in surface sediments from the Cross River estuary, S.E. Niger Delta, Nigeria. *Environ Monit Assess*, **184** (2), 1037-1047. <https://doi.org/10.1007/s10661-011-2019-5>
- El-Saeid, M. H., Al-Turki, A. M., Nadeem, M. E., Hassanin, A. S. & Al-Wabel, M. I. (2015) Photolysis degradation of polyaromatic hydrocarbons (PAHs) on surface sandy soil. *Environ Sci Pollut Res Int*, **22** (13), 9603-9616. <https://doi.org/10.1007/s11356-015-4082-x>
- Eremina, N., Paschke, A., Mazlova, E. A. & Schuurmann, G. (2016) Distribution of polychlorinated biphenyls, phthalic acid esters, polycyclic aromatic hydrocarbons and organochlorine substances in the Moscow River, Russia. *Environ Pollut*, **210**, 409-418. <https://doi.org/10.1016/j.envpol.2015.11.034>
- Guo, J., Chen, J. & Wang, J. (2017) Sedimentary records of polycyclic aromatic hydrocarbons in China: A comparison to the worldwide. *Critical Reviews in Environmental Science and Technology*, **47** (17), 1612-1667. <https://doi.org/10.1080/10643389.2017.1393262>
- Han, B., Liu, A., He, S., Li, Q. & Zheng, L. (2020) Composition, content, source, and risk assessment of PAHs in intertidal sediment in Shilaoren Bay, Qingdao, China. *Mar Pollut Bull*, **159**, 111499. <https://doi.org/10.1016/j.marpolbul.2020.111499>
- Han, J., Liang, Y., Zhao, B., Wang, Y., Xing, F. & Qin, L. (2019) Polycyclic aromatic hydrocarbon (PAHs) geographical distribution in China and their source, risk assessment analysis. *Environ Pollut*, **251**, 312-327. <https://doi.org/10.1016/j.envpol.2019.05.022>
- Hartmann, P. C., Quinn, J. G., Cairns, R. W. & King, J. W. (2004) The distribution and sources of polycyclic aromatic hydrocarbons in Narragansett Bay surface sediments. *Mar Pollut Bull*, **48** (3-4), 351-358. <https://doi.org/10.1016/j.marpolbul.2003.08.014>
- He, Y., Lin, K. & Chen, X. (2013) Effect of Land Use and Climate Change on Runoff in the Dongjiang Basin of South China. *Mathematical Problems in Engineering*, **2013**, 1-14. <https://doi.org/10.1155/2013/471429>
- He, Y., Yang, C., He, W. & Xu, F. (2020) Nationwide health risk assessment of juvenile exposure to polycyclic aromatic hydrocarbons (PAHs) in the water body of Chinese lakes. *Sci Total Environ*, **723**, 138099. <https://doi.org/10.1016/j.scitotenv.2020.138099>
- Hussain, I., Syed, J. H., Kamal, A., Iqbal, M., Eqani, S. A., Bong, C. W., Taqi, M. M., Reichenauer, T. G., Zhang, G. & Malik, R. N. (2016) The relative abundance and seasonal distribution correspond with the sources of polycyclic aromatic hydrocarbons (PAHs) in the surface sediments of Chenab River, Pakistan. *Environ Monit Assess*, **188** (6), 378. <https://doi.org/10.1007/s10661-016-5359-3>

- Idowu, O., Semple, K. T., Ramadass, K., O'Connor, W., Hansbro, P. & Thavamani, P. (2019) Beyond the obvious: Environmental health implications of polar polycyclic aromatic hydrocarbons. *Environ Int*, **123**, 543-557. <https://doi.org/10.1016/j.envint.2018.12.051>
- Jiao, W., Lu, Y., Wang, T., Li, J., Han, J., Wang, G. & Hu, W. (2009) Polycyclic aromatic hydrocarbons in soils around Guanting Reservoir, Beijing, China. *Chemistry and Ecology*, **25** (1), 39-48. <https://doi.org/10.1080/02757540802657193>
- Keith, L. & Telliard, W. (1979) ES&T Special Report: Priority pollutants: I-a perspective view. *Environmental Science & Technology*, **13** (4), 416-423. <https://doi.org/10.1021/es60152a601>
- Kim, K. H., Jahan, S. A., Kabir, E. & Brown, R. J. (2013) A review of airborne polycyclic aromatic hydrocarbons (PAHs) and their human health effects. *Environ Int*, **60**, 71-80. <https://doi.org/10.1016/j.envint.2013.07.019>
- Li, C., Huo, S., Yu, Z., Xi, B., Yeager, K. M., He, Z., Ma, C., Zhang, J. & Wu, F. (2017) National investigation of semi-volatile organic compounds (PAHs, OCPs, and PCBs) in lake sediments of China: Occurrence, spatial variation and risk assessment. *Sci Total Environ*, **579**, 325-336. <https://doi.org/10.1016/j.scitotenv.2016.11.097>
- Li, R., Hua, P., Zhang, J. & Krebs, P. (2020) Effect of anthropogenic activities on the occurrence of polycyclic aromatic hydrocarbons in aquatic suspended particulate matter: Evidence from Rhine and Elbe Rivers. *Water Res*, **179**, 115901. <https://doi.org/10.1016/j.watres.2020.115901>
- Li, Y., Liu, J., Cao, Z., Lin, C. & Yang, Z. (2010) Spatial distribution and health risk of heavy metals and polycyclic aromatic hydrocarbons (PAHs) in the water of the Luanhe River Basin, China. *Environ Monit Assess*, **163** (1-4), 1-13. <https://doi.org/10.1007/s10661-009-0811-2>
- Lu, J., Zhang, C., Wu, J., Lin, Y., Zhang, Y., Yu, X. & Zhang, Z. (2019) Pollution, sources, and ecological-health risks of polycyclic aromatic hydrocarbons in coastal waters along coastline of China. *Human and Ecological Risk Assessment: An International Journal*, **26** (4), 968-985. <https://doi.org/10.1080/10807039.2018.1548899>
- Maskaoui, K., Zhou, J. L., Hong, H. S. & Zhang, Z. L. (2002) Contamination by polycyclic aromatic hydrocarbons in the Jiulong River Estuary and Western Xiamen Sea, China. *Environmental Pollution*, **118** (1), 109-122. [https://doi.org/10.1016/S0269-7491\(01\)00208-1](https://doi.org/10.1016/S0269-7491(01)00208-1)
- Meng, Y., Liu, X., Lu, S., Zhang, T., Jin, B., Wang, Q., Tang, Z., Liu, Y., Guo, X., Zhou, J. & Xi, B. (2019) A review on occurrence and risk of polycyclic aromatic hydrocarbons (PAHs) in lakes of China. *Sci Total Environ*, **651** (Pt 2), 2497-2506. <https://doi.org/10.1016/j.scitotenv.2018.10.162>
- Mojiri, A., Zhou, J. L., Ohashi, A., Ozaki, N. & Kindaichi, T. (2019) Comprehensive review of polycyclic aromatic hydrocarbons in water sources, their effects and treatments. *Sci Total Environ*, **696**, 133971. <https://doi.org/10.1016/j.scitotenv.2019.133971>
- Nadal, M., Schuhmacher, M. & Domingo, J. L. (2004) Levels of PAHs in soil and vegetation samples from Tarragona County, Spain. *Environ Pollut*, **132** (1), 1-11. <https://doi.org/10.1016/j.envpol.2004.04.003>
- Nascimento, R. A., de Almeida, M., Escobar, N. C. F., Ferreira, S. L. C., Mortatti, J. & Queiroz, A. F. S. (2017) Sources and distribution of polycyclic aromatic hydrocarbons (PAHs) and organic matter in surface sediments of an estuary under petroleum activity influence, Todos os Santos Bay, Brazil. *Mar Pollut Bull*, **119** (2), 223-230. <https://doi.org/10.1016/j.marpolbul.2017.03.069>
- Niu, L. & van Gelder, P. (2020) Distribution and source assessment of polycyclic aromatic hydrocarbons levels from Lake IJssel (the Netherlands) and their responses to hydrology. *Water Environ Res*, **92** (8), 1214-1229. <https://doi.org/10.1002/wer.1317>

- Opere, W. M., John, M. & Ombori, O. (2020) Occurrence of Enteric Viruses in Surface Water and the Relationship with Changes in Season and Physical Water Quality Dynamics. *Adv Virol*, **2020**, 9062041. <https://doi.org/10.1155/2020/9062041>
- Palm, A., Cousins, I., Gustafsson, O., Axelman, J., Grunder, K., Broman, D. & Brorstrom-Lunden, E. (2004) Evaluation of sequentially-coupled POP fluxes estimated from simultaneous measurements in multiple compartments of an air-water-sediment system. *Environ Pollut*, **128** (1-2), 85-97. <https://doi.org/10.1016/j.envpol.2003.08.023>
- Patrolecco, L., Ademollo, N., Capri, S., Pagnotta, R. & Polesello, S. (2010) Occurrence of priority hazardous PAHs in water, suspended particulate matter, sediment and common eels (*Anguilla anguilla*) in the urban stretch of the River Tiber (Italy). *Chemosphere*, **81** (11), 1386-1392. <https://doi.org/10.1016/j.chemosphere.2010.09.027>
- Pies, C., Hoffmann, B., Petrowsky, J., Yang, Y., Ternes, T. A. & Hofmann, T. (2008) Characterization and source identification of polycyclic aromatic hydrocarbons (PAHs) in river bank soils. *Chemosphere*, **72** (10), 1594-1601. <https://doi.org/10.1016/j.chemosphere.2008.04.021>
- Qiao, M., Wang, C., Huang, S., Wang, D. & Wang, Z. (2006) Composition, sources, and potential toxicological significance of PAHs in the surface sediments of the Meiliang Bay, Taihu Lake, China. *Environ Int*, **32** (1), 28-33. <https://doi.org/10.1016/j.envint.2005.04.005>
- Saeedi, M., Li, L. Y. & Salmanzadeh, M. (2012) Heavy metals and polycyclic aromatic hydrocarbons: pollution and ecological risk assessment in street dust of Tehran. *J Hazard Mater*, **227-228**, 9-17. <https://doi.org/10.1016/j.jhazmat.2012.04.047>
- Sanches, S., Leitao, C., Penetra, A., Cardoso, V. V., Ferreira, E., Benoliel, M. J., Crespo, M. T. & Pereira, V. J. (2011) Direct photolysis of polycyclic aromatic hydrocarbons in drinking water sources. *J Hazard Mater*, **192** (3), 1458-1465. <https://doi.org/10.1016/j.jhazmat.2011.06.065>
- Sarria-Villa, R., Ocampo-Duque, W., Paez, M. & Schuhmacher, M. (2016) Presence of PAHs in water and sediments of the Colombian Cauca River during heavy rain episodes, and implications for risk assessment. *Sci Total Environ*, **540**, 455-465. <https://doi.org/10.1016/j.scitotenv.2015.07.020>
- Seopela, M. P., McCrindle, R. I., Combrinck, S. & Augustyn, W. (2020) Occurrence, distribution, spatio-temporal variability and source identification of n-alkanes and polycyclic aromatic hydrocarbons in water and sediment from Loskop dam, South Africa. *Water Res*, **186**, 116350. <https://doi.org/10.1016/j.watres.2020.116350>
- Shen, B., Wu, J., Zhan, S. & Jin, M. (2021) Residues of organochlorine pesticides (OCPs) and polycyclic aromatic hydrocarbons (PAHs) in waters of the Ili-Balkhash Basin, arid Central Asia: Concentrations and risk assessment. *Chemosphere*, **273**, 129705. <https://doi.org/10.1016/j.chemosphere.2021.129705>
- Sun, C., Zhang, J., Ma, Q. & Chen, Y. (2015) Human Health and Ecological Risk Assessment of 16 Polycyclic Aromatic Hydrocarbons in Drinking Source Water from a Large Mixed-Use Reservoir. *Int J Environ Res Public Health*, **12** (11), 13956-13969. <https://doi.org/10.3390/ijerph121113956>
- Sun, J. H., Wang, G. L., Chai, Y., Zhang, G., Li, J. & Feng, J. (2009) Distribution of polycyclic aromatic hydrocarbons (PAHs) in Henan Reach of the Yellow River, Middle China. *Ecotoxicol Environ Saf*, **72** (5), 1614-1624. <https://doi.org/10.1016/j.ecoenv.2008.05.010>
- Takane, Y. & Hwang, H. (2005) On a test of dimensionality in redundancy analysis. *Psychometrika*, **70** (2), 271-281. <https://doi.org/10.1007/s11336-003-1089-x>

- Tobiszewski, M. & Namiesnik, J. (2012) PAH diagnostic ratios for the identification of pollution emission sources. *Environ Pollut*, **162**, 110-119. <https://doi.org/10.1016/j.envpol.2011.10.025>
- Yim, U. H., Hong, S. H. & Shim, W. J. (2007) Distribution and characteristics of PAHs in sediments from the marine environment of Korea. *Chemosphere*, **68** (1), 85-92. <https://doi.org/10.1016/j.chemosphere.2006.12.032>
- Yu, H., Liu, Y., Han, C., Fang, H., Weng, J., Shu, X., Pan, Y. & Ma, L. (2021) Polycyclic aromatic hydrocarbons in surface waters from the seven main river basins of China: Spatial distribution, source apportionment, and potential risk assessment. *Sci Total Environ*, **752**, 141764. <https://doi.org/10.1016/j.scitotenv.2020.141764>
- Yu, Y., Xu, J., Wang, P., Sun, H. & Dai, S. (2009) Sediment-porewater partition of polycyclic aromatic hydrocarbons (PAHs) from Lanzhou Reach of Yellow River, China. *J Hazard Mater*, **165** (1-3), 494-500. <https://doi.org/10.1016/j.jhazmat.2008.10.042>
- Yuan, K., Wang, X., Lin, L., Zou, S., Li, Y., Yang, Q. & Luan, T. (2015) Characterizing the parent and alkyl polycyclic aromatic hydrocarbons in the Pearl River Estuary, Daya Bay and northern South China Sea: influence of riverine input. *Environ Pollut*, **199**, 66-72. <https://doi.org/10.1016/j.envpol.2015.01.017>
- Yunker, M. B., Macdonald, R. W., Vingarzan, R., Mitchell, R. H., Goyette, D. & Sylvestre, S. (2002) PAHs in the Fraser River basin: a critical appraisal of PAH ratios as indicators of PAH source and composition. *Organic Geochemistry*, **33** (4), 489-515. [https://doi.org/10.1016/S0146-6380\(02\)00002-5](https://doi.org/10.1016/S0146-6380(02)00002-5)
- Zhang, H. B., Luo, Y. M., Wong, M. H., Zhao, Q. G. & Zhang, G. L. (2006) Distributions and concentrations of PAHs in Hong Kong soils. *Environ Pollut*, **141** (1), 107-114. <https://doi.org/10.1016/j.envpol.2005.08.031>
- Zhang, J., Liu, G., Wang, R. & Huang, H. (2017) Polycyclic aromatic hydrocarbons in the water-SPM-sediment system from the middle reaches of Huai River, China: Distribution, partitioning, origin tracing and ecological risk assessment. *Environ Pollut*, **230**, 61-71. <https://doi.org/10.1016/j.envpol.2017.06.012>
- Zhang, Y., Lu, Y., Xu, J., Yu, T. & Zhao, W. (2011) Spatial distribution of polycyclic aromatic hydrocarbons from Lake Taihu, China. *Bull Environ Contam Toxicol*, **87** (1), 80-85. <https://doi.org/10.1007/s00128-011-0292-1>
- Zhao, S. M., Wang, B., Wang, D. W., Li, X. M., Huang, B., Hu, P., Zhang, L. W. & Pan, X. J. (2014) Environmental Behavior of PAHs in Dianchi Lake Distributions, Sources and Risk Assessment of Polycyclic Aromatic Hydrocarbons in Surface Sediments from Dianchi Lake, China. *International Journal of Environmental Research*, **8** (2), 317-328. <https://dx.doi.org/10.22059/IJER.2014.722>
- Zheng, T., Ran, Y. & Chen, L. (2013) Polycyclic aromatic hydrocarbons (PAHs) in rural soils of Dongjiang River Basin: occurrence, source apportionment, and potential human health risk. *Journal of Soils and Sediments*, **14** (1), 110-120. <https://doi.org/10.1007/s11368-013-0753-8>

Figures

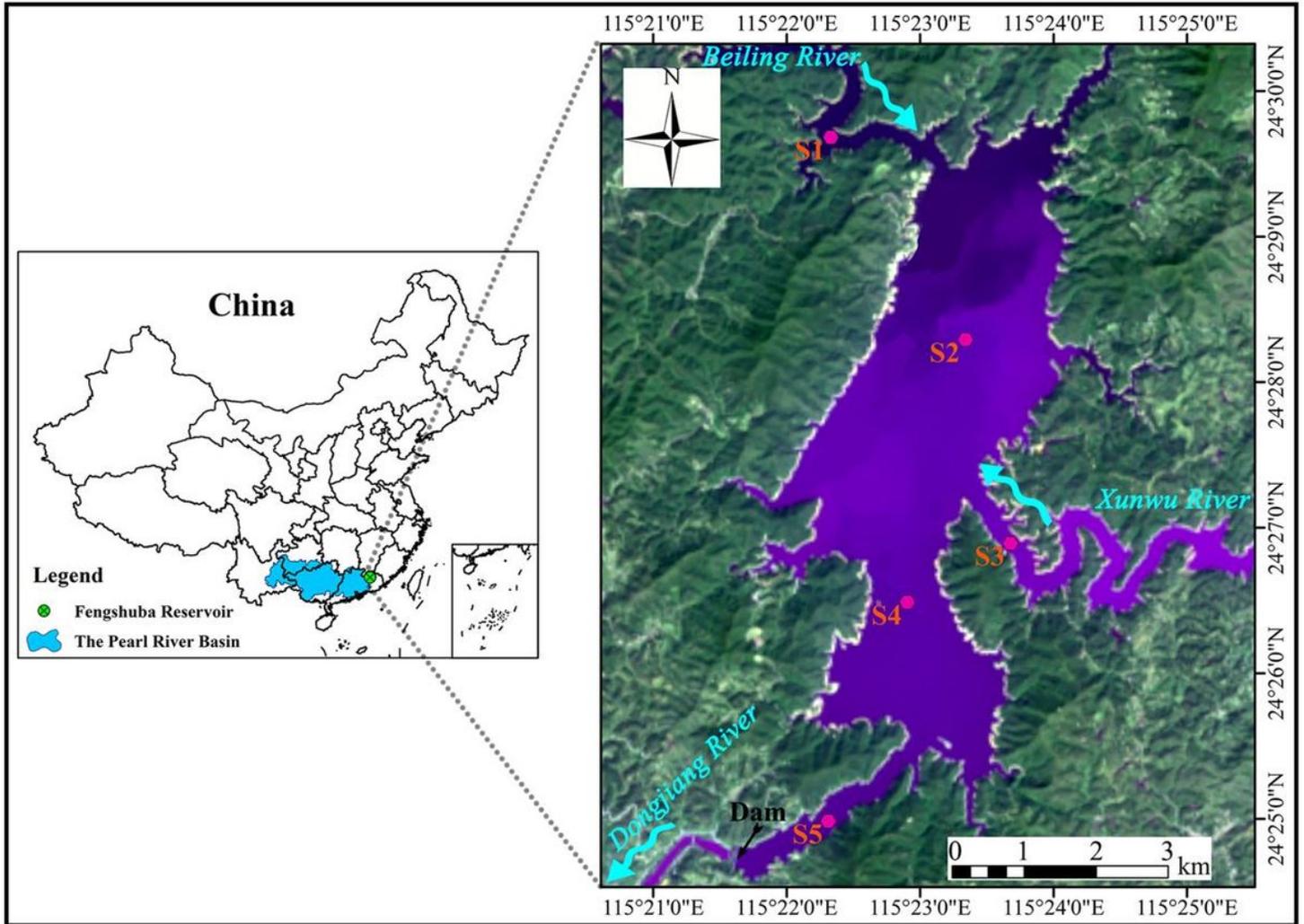


Figure 1

Map showing the location of Fengshuba Reservoir and sampling stations. S1 (northwest inlet of reservoir); S2 (upstream of reservoir); S3 (southeast inlet of reservoir); S4 (midstream of reservoir); S5 (downstream of reservoir). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

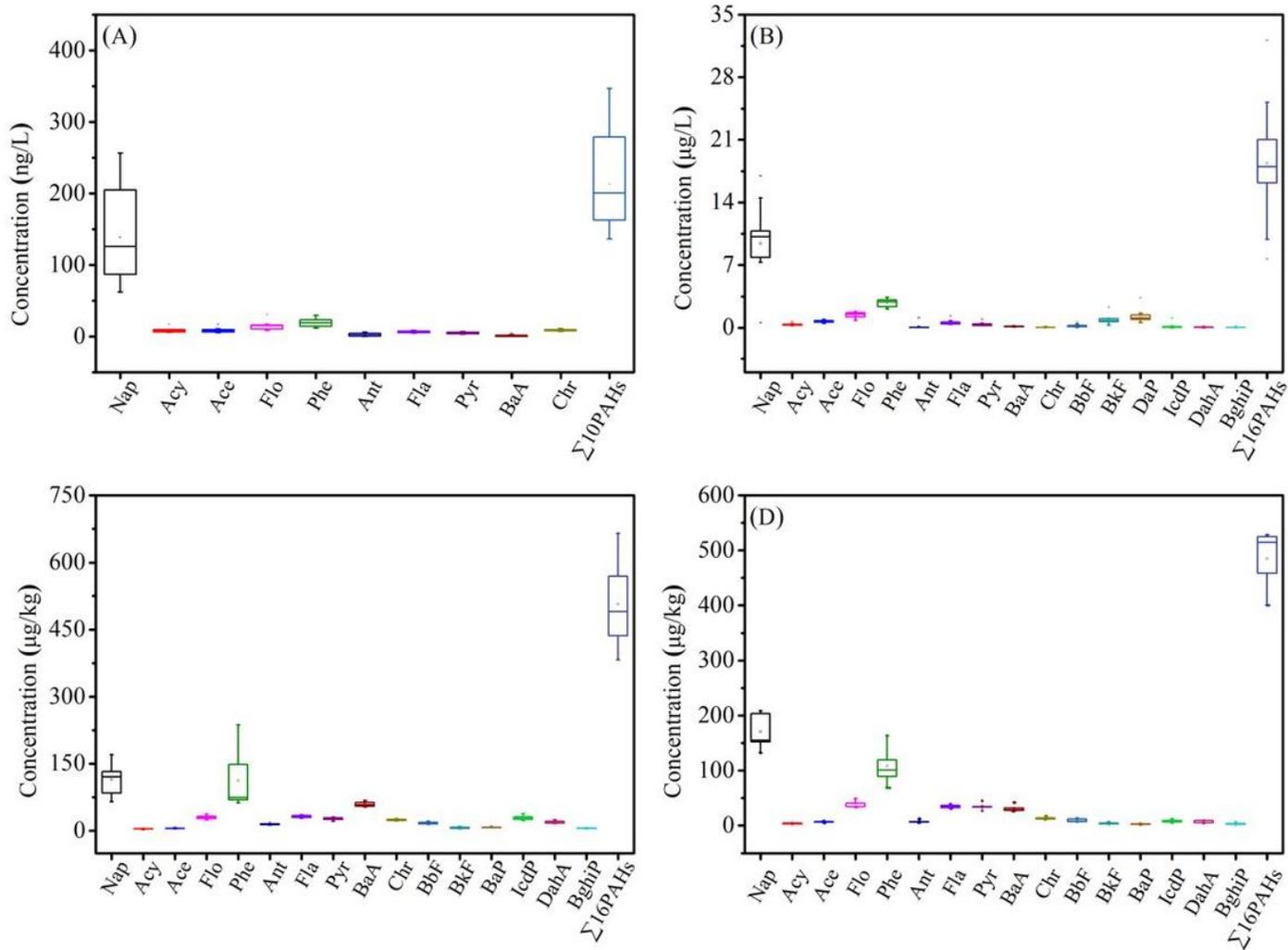


Figure 2

Concentrations of the PAHs in the waters (A), porewaters (B), sediments (C), and soils (D) of Fengshuba Reservoir.

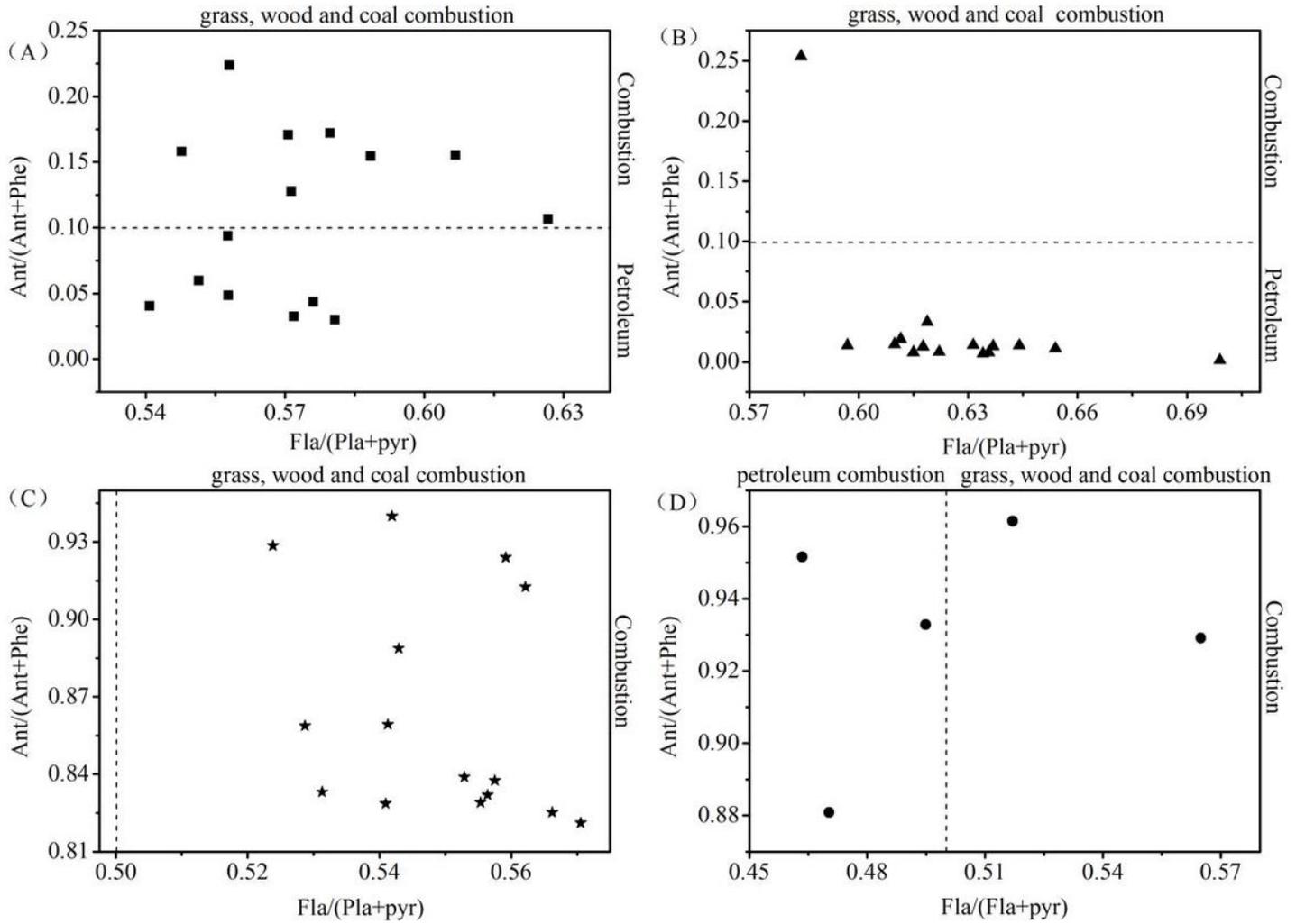


Figure 3

Plots of PAH isomer ratios for the identification of PAH sources: Fla/(Fla + Pyr) vs. Ant/(Ant + Phe) in water samples (A), porewater samples (B), sediments samples (C), and soil samples (D).

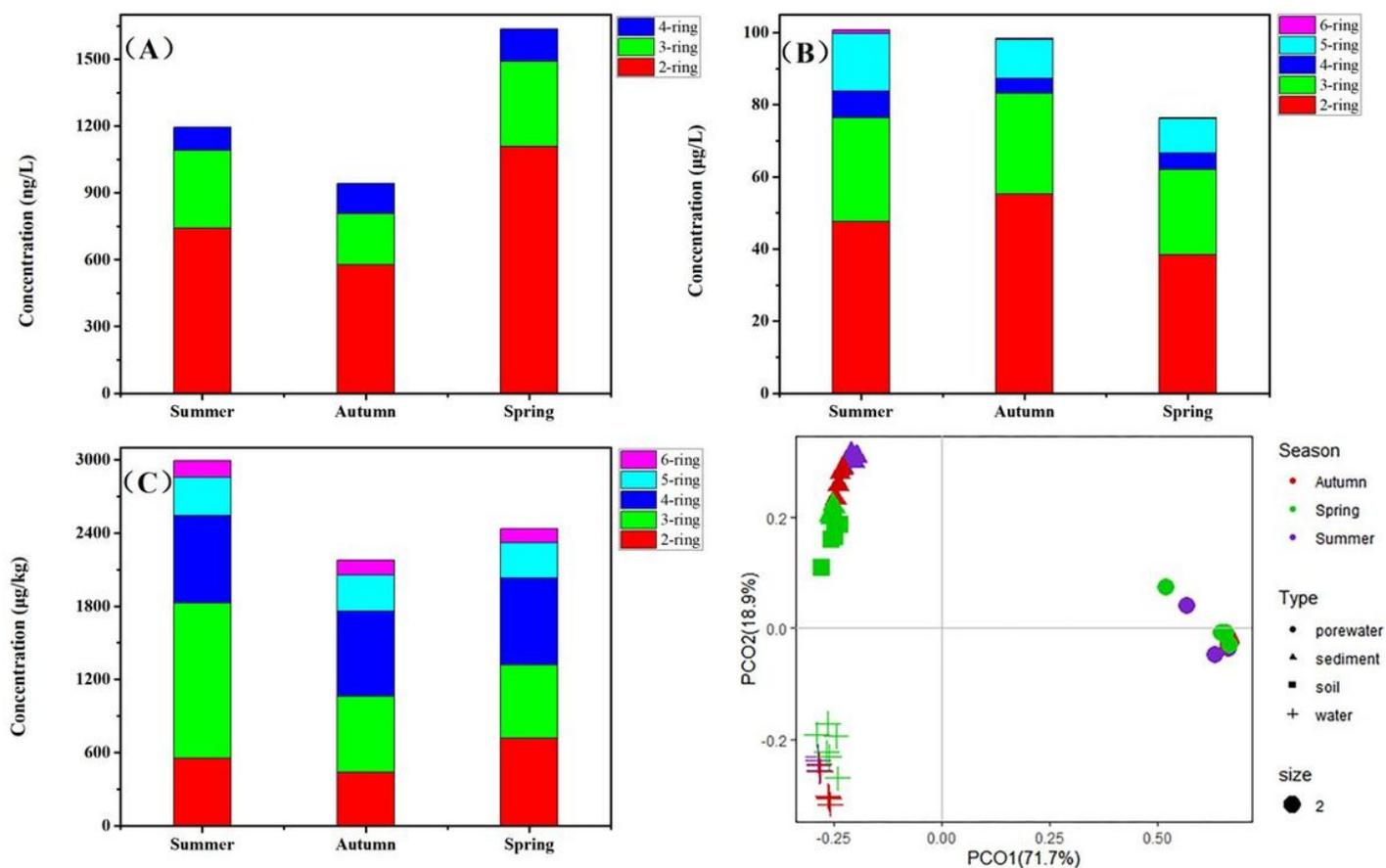


Figure 4

Concentrations of the PAHs in the waters (A), porewaters (B), and sediments (C) with the seasons. Compositions of the PAHs among the environmental compartments using the PCoA based on the Bray-Curtis dissimilarity matrices (D). Error bars represent standard error of the mean.

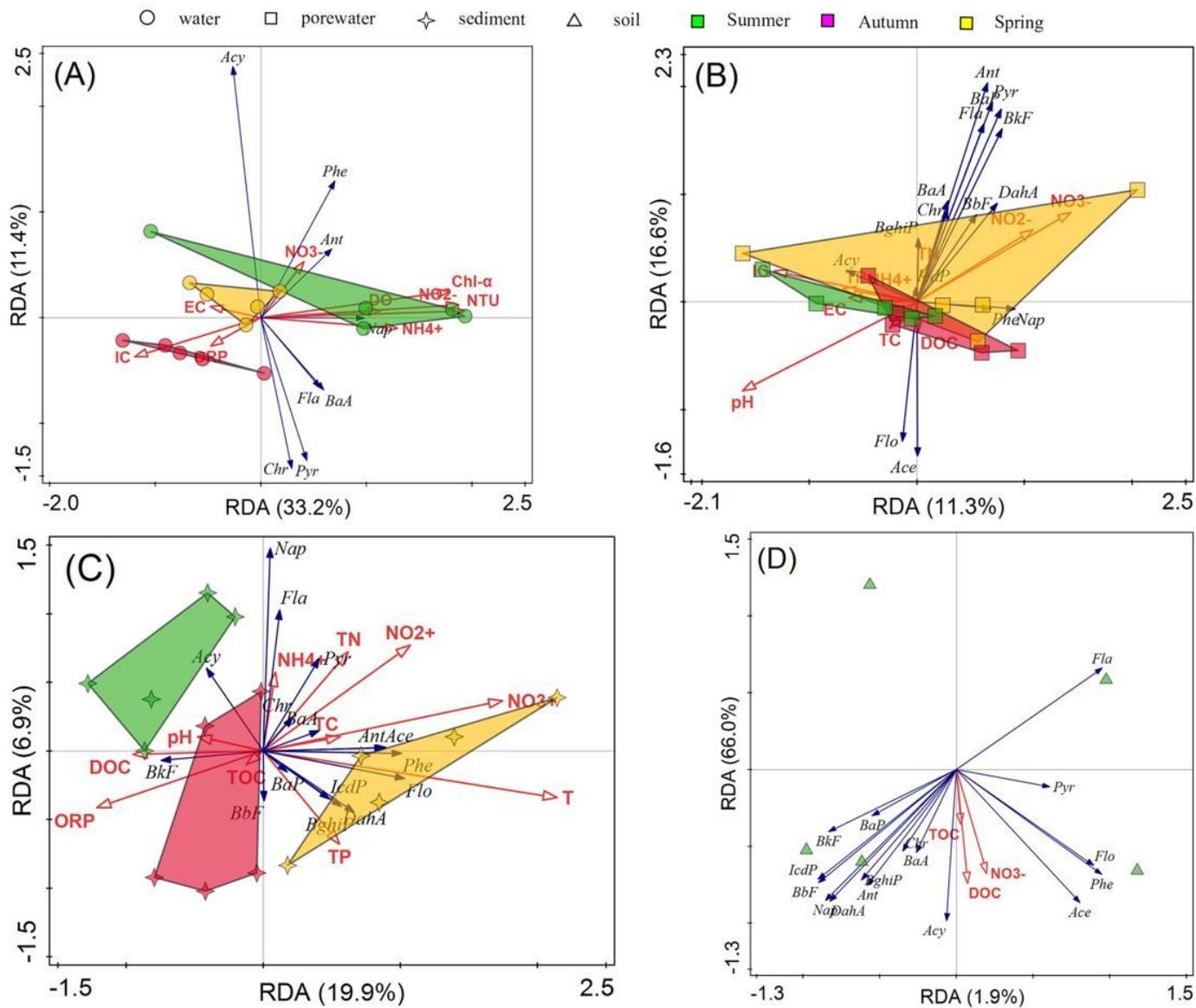


Figure 5

RDA of the PAHs concentrations and environmental concentrations in the waters (A), porewaters (B), sediments (C), and soils (D) of Fengshuba Reservoir.

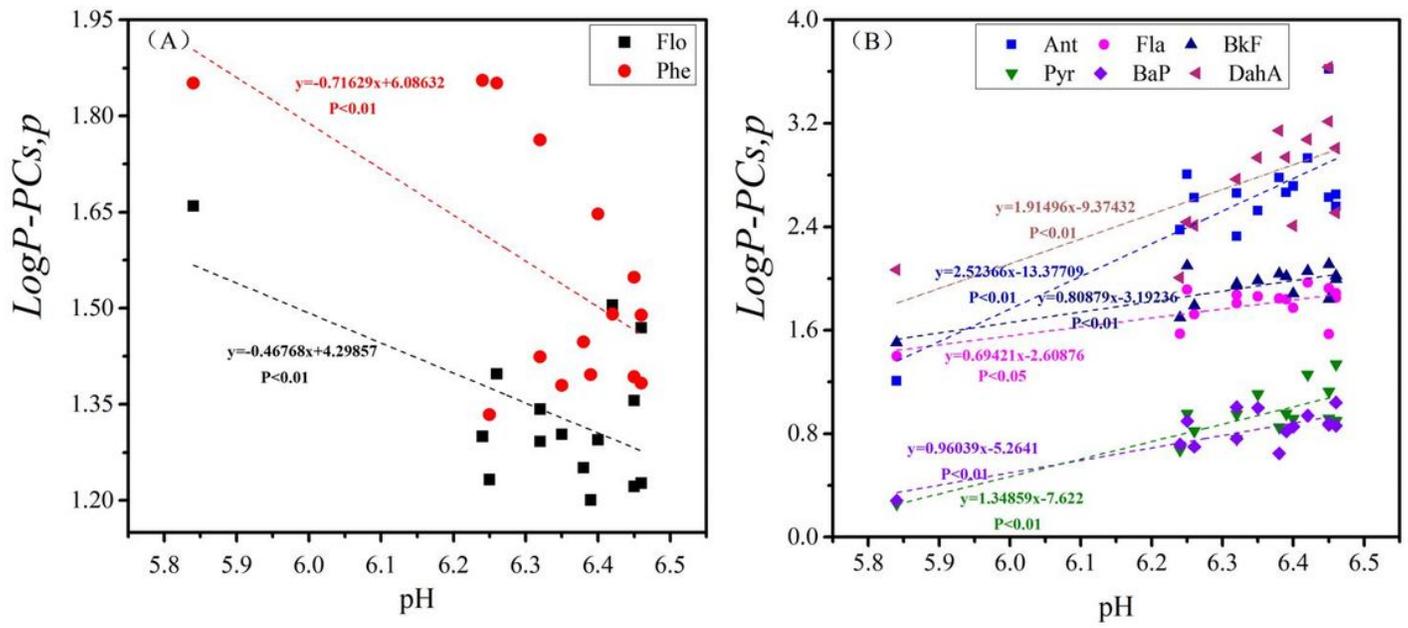


Figure 6

Linear correlation analysis of the Log P-PCs, w plotted against the pH level of in porewater phase.

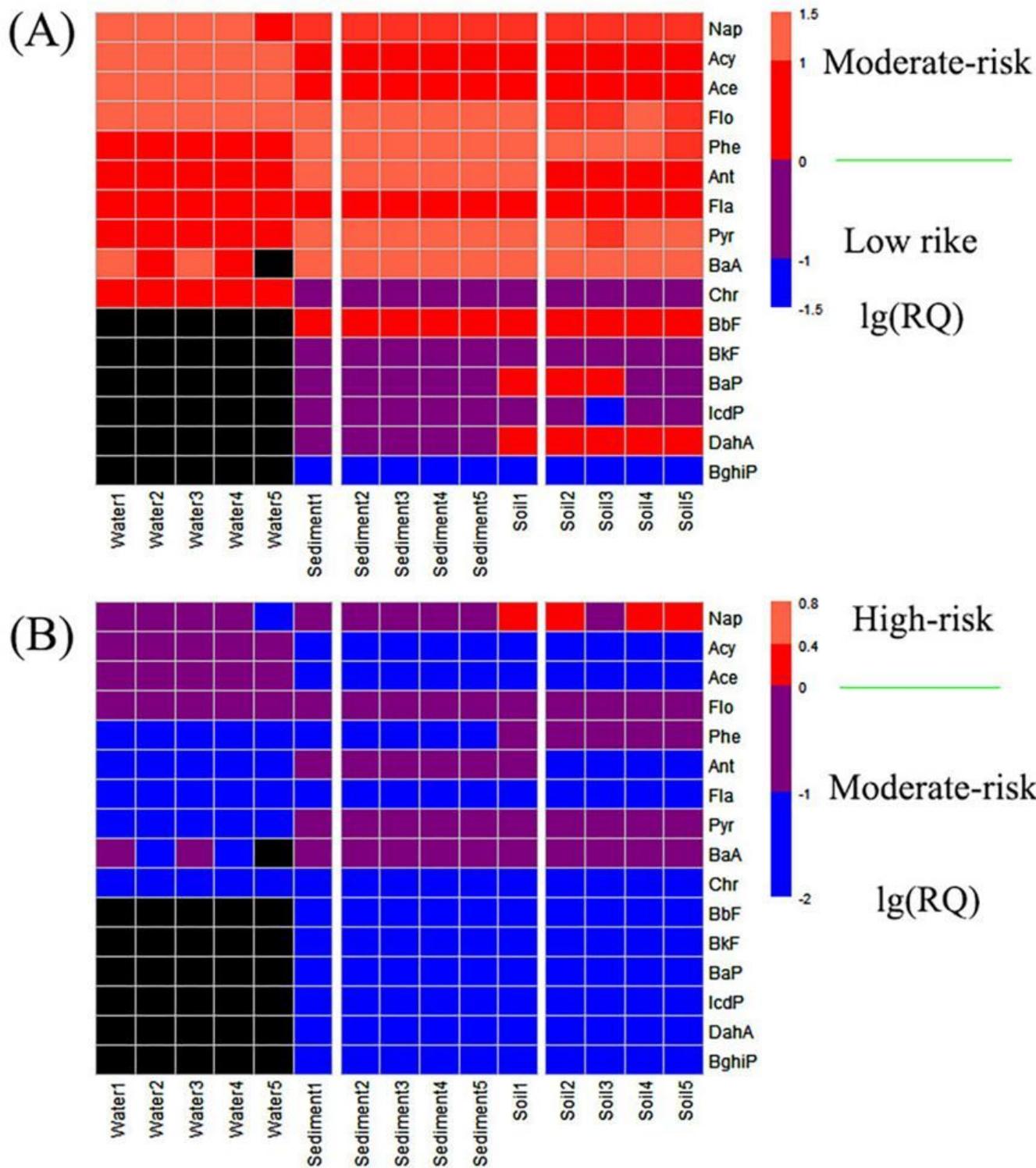


Figure 7

Calculated risk quotient (RQ) for the detected PAHs in the water, sediment, soil samples. Bottom right shows the color band indicator of the level of ecological risk based on a log scale. Black boxes indicate no ecological risks due to the corresponding PAHs undetected.

Supplementary Files

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