Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology.


Supplementary material to

# Sources and ecological risk of polycyclic aromatic hydrocarbons in water and air of the Yangtze River

## Contents

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## Abbreviation List

| Full Name | Abbreviation |
|---|---|
| Polycyclic aromatic hydrocarbons | PAHs |
| Shanghai | SH |
| Nantong | NT |
| Changzhou | ChangZ |
| Zhenjiang | ZhJi |
| Nanjing | NJ |
| Wuhu | WHu |
| Chizhou | ChiZ |
| Anqing | AQ |
| Jiujiang | JJ |
| Huangshi | HS |
| Ezhou | EZ |
| Wuhan | WHan |
| Yueyang | YY |
| Jingzhou | JZ |
| Yichang | YC |
| Chongqing | CQ |
| Zhejiang | ZJ |
| Jiangsu | JS |
| Anhui | AH |
| Hubei | HB |
| Hunan | HN |
| Jiangxi | JX |
| Principal component analysis | PCA |
| Absolute principal component score | APCS |
| Fugacity fractions | ff |
| Octanol–water partition coefficient | Kow |
| Dichloromethane | DCM |
| Gas chromatograph | GC |
| Mass spectrometer | MS |

## Table S1 List of 16 PAHs and their physicochemical properties

| | Congener | | Ring | H[a] (Pa/m³/mol) | Instrument detection limit (ng/mL) | Recovery rate (%) |
|---|---|---|---|---|---|---|
| 1 | Naphthalene | Nap | 2 | 55 | 0.38 | 82.1 ± 13 |
| 2 | Acenaphthylene | Acy | 3 | 16 | 0.19 | 117 ± 9.8 |
| 3 | Acenaphthene | Ace | 3 | 29 | 0.24 | 90.2 ± 1.5 |
| 4 | Fluorene | Fl | 3 | 10 | 0.20 | 93.1 ± 7.0 |
| 5 | Phenanthrene | Phe | 3 | 4.8 | 0.19 | 90.7 ± 4.2 |
| 6 | Anthracene | Ant | 3 | 5.2 | 0.21 | 89.6 ± 6.2 |
| 7 | Fluoranthene | Flu | 4 | 1.7 | 0.17 | 113 ± 8.4 |
| 8 | Pyrene | Pyr | 4 | 2.1 | 0.15 | 109.9 ± 16 |
| 9 | Benzo[a]anthracene | BaA | 4 | 0.45 | 0.14 | 69.9 ± 24 |
| 10 | Chrysene | Chr | 4 | 0.42 | 0.21 | 91.1 ± 4.7 |
| 11 | Benzo[b]fluoranthene | BbF | 5 | 0.18 | 0.74 | 108.1 ± 12 |
| 12 | Benzo[k]fluoranthene | BkF | 5 | 0.17 | 0.81 | 97 ± 6.9 |
| 13 | Benzo[a]pyrene | BaP | 5 | 0.21 | 1.05 | 90.8 ± 5.1 |
| 14 | Indeno[1,2,3-cd]pyrene | InP | 5 | 0.07 | 1.41 | 79.3 ± 19 |
| 15 | Dibenz[a,h]anthracene | DaA | 5 | 0.05 | 2.05 | 63.7 ± 9.7 |
| 16 | Benzo[ghi]perylene | BghiP | 6 | 0.09 | 3.37 | 70.4 ± 9.6 |

[a] Henty's law consistent at 25 °C [1]

**Table S2 Concentration of 16 PAHs in water (ng/L)**

| | Nap | Acy | Ace | Fl | Phe | Ant | Flu | Pyr | BaA | Chr | BbF | BkF | BaP | InP | DaA | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Mean | 153 | 5.56 | 5.44 | 4.67 | 18.2 | 1.25 | 4.04 | 5.50 | 0.15 | 0.51 | 0.08 | ND | 0.45 | ND | ND | ND |
| Std Dev | 113 | 1.91 | 1.67 | 2.02 | 16.7 | 1.55 | 3.69 | 5.56 | 0.19 | 0.14 | 0.05 | ND | 0.15 | 0.44 | 0.11 | ND |
| Max | 421 | 9.77 | 9.43 | 9.34 | 54.5 | 5.05 | 11.2 | 18.1 | 1.03 | 0.82 | 0.21 | ND | 0.74 | 2.39 | 0.39 | ND |
| Min | 11.7 | 2.21 | 2.88 | ND | ND | ND | ND | ND | ND | 0.31 | ND | ND | 0.20 | ND | ND | ND |
| Median | 137 | 5.66 | 5.57 | 4.71 | 15.10 | 0.48 | 4.14 | 5.73 | 0.12 | 0.48 | 0.07 | ND | 0.45 | ND | 0.21 | ND |

ND: Detected concentration below the method detection limit.

**Table S3 Concentration ranges of PAHs in water dissolved phase collected from different rivers in China**

| Site | Range | Number of PAHs | Sampling year | References |
|---|---|---|---|---|
| Yangtze River | 19.9–468 | 16 | 2019 | This study |
| Yangtze River | 2.4–761.2 | 16 | 2017, 2018 | [2] |
| Rivers in Shanghai [Y] | 46.5-460 | 15 [a] | 2015 | [3] |
| Taihu lake [Y] | 45.4-233 | 16 | 2010 | [4] |
| Poyang lake [Y] | 5.56-266 | 16 | 2012 | [5] |
| Songhua River | 182-397 | 16 | 2010 | [6] |
| Pearl River | 92.8-324 | 16 | 2016 | [7] |
| Yellow River | 64.8-335 | 16 | 2007 | [3] |
| Huaihe River | 79.9-421 | 16 | 2013, 2014 | [8] |
| Weihe River | 351-4427 | 16 | 2014 | [9] |
| Daliao River | 71.1-4250 | 16 | 2013 | [6] |
| Jinjiang River | 42-63 | 16 | 2011 | [9] |

[Y] Tributary of Yangtze River;

a 15 priority PAHs except Nap.

**Table S4 Concentrations of PAHs in the atmosphere collected from China**

| Sites | Ranges (ng/m$^3$) | Mean (ng/m$^3$) | Number of PAHs | Sampling year | References |
|---|---|---|---|---|---|
| Yangtze River | 21.4 - 209 | 104 | 16 | 2019 | Present study |
| North China | 6.3 – 2180[a] | 239 [b] | 16 | 2008, 2009 | [10] |
| | 13 - 785 [a] | 254 [b] | 15 [a] | 2011 | [11] |
| Taiyuan | | 76.5 in spring, 87.4 in winter [c, a] | 15 [c] | 2009, 2010 | [12] |
| | | 142 [c] | 21 [d] | | [13] |
| Wuwei | | 88 [c] | 21 [d] | | |
| Yinchuan | | 72 [c] | 21 [d] | | |
| Beijing | | 118 [c] | 21 [d] | | |
| Dezhou | | 96 [c] | 21 [d] | | |
| South China | 15.2 - 937 [a] | 165 [b] | 16 | 2008, 2009 | [10] |
| Guangzhou | 49.6 - 585 | 313 [c] | 16 | 2001, 2002 | [14] |
| Shanghai | 33.6 - 55.4 | 36.0 [c] | 13 | 2007 | [15] |

[b] Total PAHs of gaseous phase plus particle phase;

[c] PAHs in gaseous phase;

[d] 21 PAHs including 15 priority PAHs except Nap, and retene (RET), perylene (PER), benzo(e)pyrene (BeP), dibenzo(a,l)pyrene (dBalP), dibenzo(a,e) pyrene (dBaeP), dibenzo(a,h)pyrene (dBahP).

**Table S5 Concentration of 16 PAHs in air (ng/m$^3$)**

| | Nap | Acy | Ace | Fl | Phe | Ant | Flu | Pyr | BaA | Chr | BbF | BkF | BaP | InP | DaA | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Mean | 57.1 | 2.11 | 1.92 | 3.69 | 13.6 | 12.1 | 6.19 | 5.27 | 0.65 | 0.53 | 0.41 | 0.16 | 0.23 | 0.12 | 0.03 | 0.09 |
| Std Dev | 42.0 | 2.74 | 1.50 | 2.54 | 9.13 | 11.0 | 4.67 | 4.28 | 0.79 | 0.48 | 0.51 | 0.17 | 0.28 | 0.13 | 0.03 | 0.10 |
| Max | 157 | 10.2 | 6.08 | 11.2 | 34.4 | 40.2 | 17.9 | 14.1 | 2.73 | 1.8 | 1.99 | 0.67 | 0.87 | 0.48 | 0.10 | 0.36 |
| Min | 4.68 | 0.12 | 0.53 | 0.52 | 2.30 | 0.50 | 1.19 | 0.83 | 0.06 | 0.08 | 0.04 | 0.02 | 0.01 | 0.02 | ND | 0.01 |
| Median | 59.0 | 1.32 | 1.31 | 2.89 | 12.1 | 10.8 | 5.04 | 3.50 | 0.39 | 0.35 | 0.22 | 0.13 | 0.07 | 0.08 | 0.02 | 0.06 |

ND: Detected concentration below the method detection limit

## Table S6 Component matrix of PAHs in water by PCA analysis

| | $R^2$ | R1 | R2 | R3 | R4 | R5 | R6 |
|---|---|---|---|---|---|---|---|
| Nap | 0.90** | 0.68** | 0.47** | -0.24** | 2.5** | -0.15* | -0.25** |
| Acy | 0.88** | -0.40** | 0.44** | -0.07 | -0.04 | 0.60** | 0.41** |
| Ace | 0.81** | 0.41** | 0.50** | -0.21* | 0.56** | 0.08 | -0.16 |
| Fl | 0.92** | 0.41** | -0.07 | 0.21** | 0.62** | 0.55** | 0.15* |
| Phe | 0.93** | 0.94** | -0.08 | 0.18** | 0.08 | 0.04 | 0.07 |
| Ant | 0.95** | 0.93** | -0.07 | 0.20** | 0.05 | -0.06 | 0.17** |
| Flu | 0.97** | 0.88** | 0.20** | 0.10* | -0.37** | -0.02 | 0.10* |
| Pyr | 0.95** | 0.88** | 0.19** | 0.08 | -0.35** | -0.02 | 0.12 |
| BaA | 0.83** | -0.30** | 0.65** | 0.52** | -0.04 | -0.11 | 0.17 |
| Chr | 0.78** | -0.28** | 0.61** | 0.34** | -0.37** | 0.25* | -0.09 |
| BbF | 0.90** | -0.10 | 0.87** | -0.03 | 0.13 | -0.27** | -0.19** |
| BkF | 0.76** | -0.42** | 0.39** | 0.14 | 0.20 | -0.39** | 0.48** |
| BaP | 0.68** | -0.23 | 0.50** | -0.55** | 0.15 | 0.13 | -0.19 |
| InP | 0.83** | -0.14 | -0.04 | 0.62** | -0.15 | 0.26** | -0.58** |
| DaA | 0.77** | -0.36** | -0.53** | 0.31** | 0.48** | -0.16 | -0.02 |
| BghiP | 0.76** | 0.01 | 0.18 | 0.79** | 0.29** | -0.14 | -0.04 |
| Cumulative variance | | 30.1% | 49.1% | 61.9% | 71.8% | 18.8% | 85.1% |

## Table S7 Linear regression coeffficient of PAHs in water by PCA-APCS

| | PC 1 | PC 2 | PC 3 | PC 4 | PC 5 | PC 6 |
|---|---|---|---|---|---|---|
| Nap | 0.684 | 0.473 | -0.235 | 0.252 | -0.149 | -0.252 |
| Acy | -0.400 | 0.437 | -0.065 | -0.039 | 0.598 | 0.406 |
| Ace | 0.410 | 0.500 | -0.210 | 0.564 | 0.081 | -0.156 |
| Fl | 0.407 | -0.065 | 0.209 | 0.621 | 0.547 | 0.154 |
| Phe | 0.938 | -0.075 | 0.179 | 0.082 | 0.037 | 0.072 |
| Ant | 0.931 | -0.073 | 0.197 | 0.045 | -0.064 | 0.168 |
| Flu | 0.880 | 0.204 | 0.101 | -0.365 | -0.016 | 0.101 |
| Pyr | 0.878 | 0.193 | 0.075 | -0.351 | -0.021 | 0.115 |
| BaA | -0.302 | 0.652 | 0.523 | -0.037 | -0.113 | 0.167 |
| Chr | -0.284 | 0.613 | 0.344 | -0.370 | 0.252 | -0.092 |
| BbF | -0.102 | 0.873 | -0.033 | 0.127 | -0.273 | -0.190 |
| BkF | -0.419 | 0.386 | 0.136 | 0.196 | -0.385 | 0.477 |
| BaP | -0.226 | 0.503 | -0.547 | 0.145 | 0.129 | -0.187 |
| InP | -0.137 | -0.043 | 0.621 | -0.145 | 0.260 | -0.577 |
| DaA | -0.358 | -0.532 | 0.309 | 0.484 | -0.163 | -0.017 |
| BghiP | 0.013 | 0.178 | 0.793 | 0.289 | -0.138 | -0.035 |

### Table S8 Component matrix of PAHs in air by PCA analysis

| | PC 1 | PC 2 | PC 3 |
|---|---|---|---|
| Nap | 0.258 | 0.456 | 0.549 |
| Acy | 0.412 | 0.603 | 0.567 |
| Ace | 0.510 | 0.595 | -0.550 |
| Fl | 0.708 | 0.547 | -0.389 |
| Phe | 0.926 | -0.146 | -0.081 |
| Ant | 0.515 | -0.675 | 0.072 |
| Flu | 0.982 | -0.086 | -0.099 |
| Pyr | 0.932 | 0.099 | 0.071 |
| BaA | 0.973 | -0.032 | -0.095 |
| Chr | 0.982 | -0.087 | -0.061 |
| BbF | 0.962 | -0.223 | 0.061 |
| BkF | 0.960 | -0.165 | 0.029 |
| BaP | 0.971 | 0.095 | 0.039 |
| InP | 0.977 | -0.089 | 0.124 |
| DaA | 0.960 | -0.036 | -0.079 |
| BghiP | 0.951 | -0.067 | 0.243 |

### Table S9 Linear regression coeffficient of PAHs in AIR by PCA-APCS

| | $R^2$ | R1 | R2 | R3 |
|---|---|---|---|---|
| Nap | 0.576* | 0.258 | 0.457 | 0.548* |
| Acy | 0.855** | 0.411** | 0.604** | 0.567** |
| Ace | 0.916** | 0.509** | 0.594** | -0.552** |
| Fl | 0.950** | 0.707** | 0.547** | -0.389** |
| Phe | 0.885** | 0.926** | -0.146 | -0.080 |
| Ant | 0.644** | 0.515* | -0.675** | 0.072 |
| Flu | 0.976** | 0.982** | -0.087 | -0.098* |
| Pyr | 0.884** | 0.932** | 0.099 | 0.071 |
| BaA | 0.958** | 0.974** | -0.030 | -0.097 |
| Chr | 0.976** | 0.982** | -0.091 | -0.060 |
| BbF | 0.978** | 0.962** | -0.222** | 0.063 |
| BkF | 0.951** | 0.960** | -0.172* | 0.031 |
| BaP | 0.953** | 0.971** | 0.095 | 0.040 |
| InP | 0.978** | 0.977** | -0.080 | 0.127* |
| DaA | 0.923** | 0.959** | -0.05 | -0.056 |
| BghiP | 0.967** | 0.951** | -0.059 | 0.244** |

**Table S10 Acute and chronic Species Sensitivity Distribution parameters and evaluation criteria**

| | Acute risk | | | | Chronic risk | | | |
|---|---|---|---|---|---|---|---|---|
| Compounds | μ (μg/L) | σ (μg/L) | number of species | MoA classification | μ (μg/L) | σ (μg/L) | number of species | MoA classification |
| Nap | 3.5699786 | 0.6007781 | 10 | 1 | 3.0880456 | 0.6366096 | 8 | 1 |
| Pyr | 1.4792086 | 0.6741506 | 4 | 1 | 0.1583625 | 0.8494513 | 3 | 2 |
| Ace | 2.806683 | 0.9237374 | 4 | 2 | 1.4696098 | 0.6646106 | 4 | 1 |
| Fl | 3.3710679 | 0.8147327 | 7 | 2 | 2.462398 | 1.0993242 | 5 | 3 |
| Phe | 2.7516372 | 0.903772 | 17 | 2 | 2.72022 | 0.8936563 | 8 | 2 |
| Ant | 1.5520191 | 0.8080877 | 7 | 2 | 1.3121805 | 1.2003597 | 4 | 3 |
| Flu | 2.0700339 | 0.3509727 | 8 | 3 | 1.69897 | 0.9911127 | 15 | 2 |
| BaP | 1.4668676 | 1.4444968 | 8 | 4 | 1.3914137 | 1.7698241 | 10 | 4 |
| BaA | 1.5793093 | 1.4836184 | 4 | 4 | 0.860585 | 1.2062285 | 4 | 3 |
| BbF | 0.4019431 | 1.5670758 | 3 | 4 | -0.598056 | 1.8077062 | 3 | 4 |

**Fig. S1 Detailed sampling locations of water sampling sites in the Yangtze River**

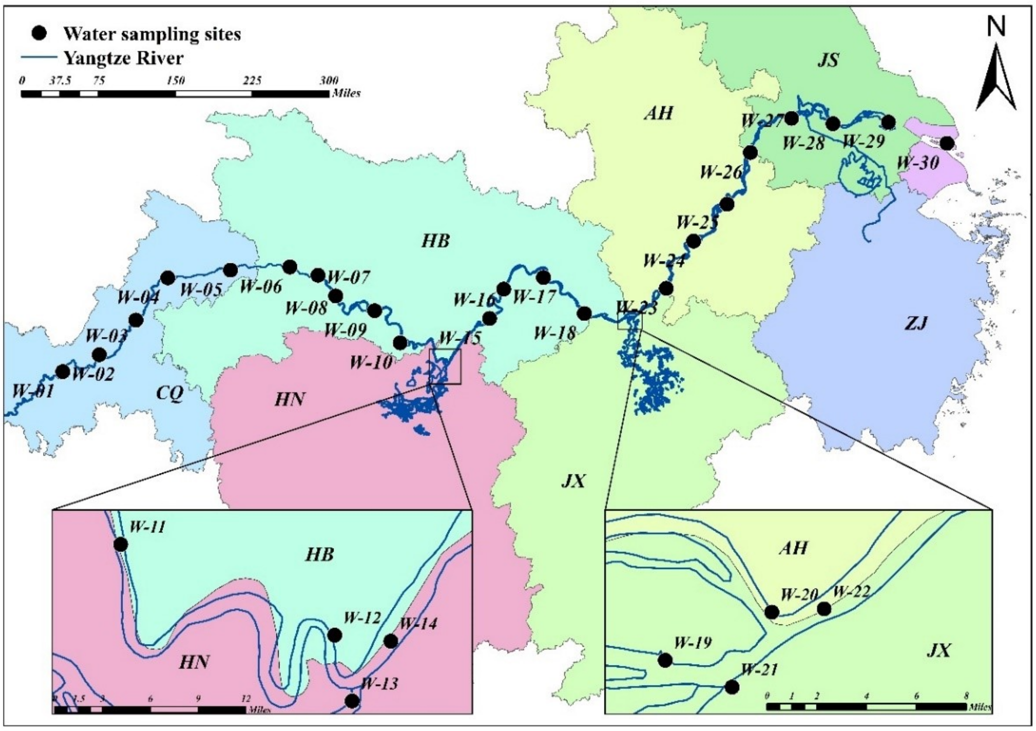
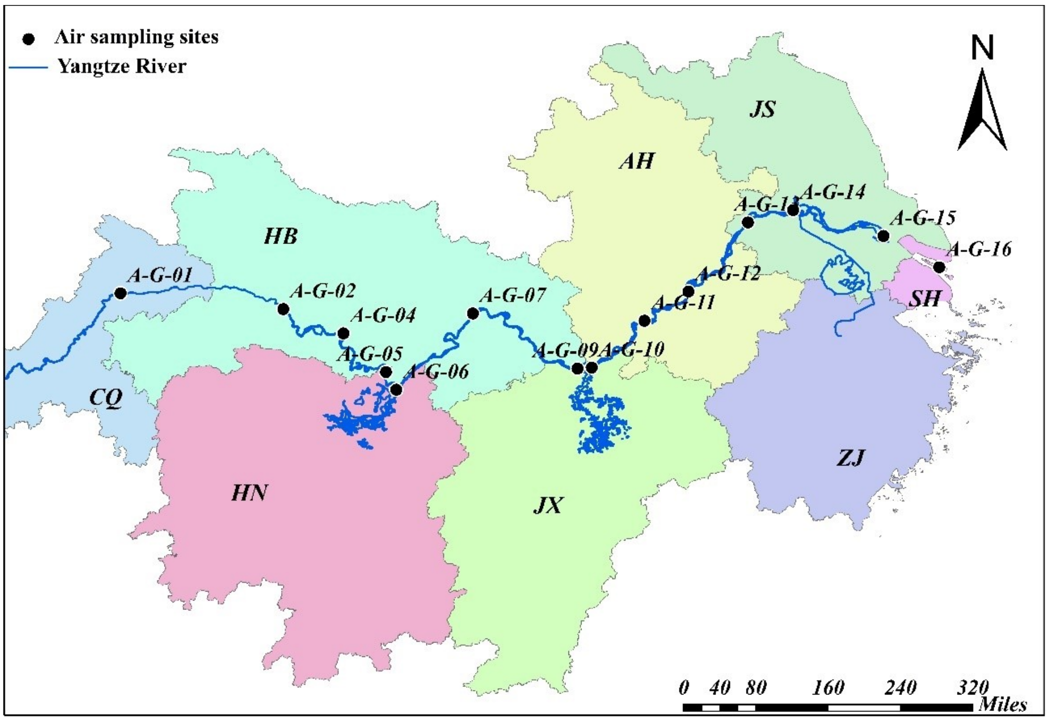



**Fig. S2 Detailed sampling end locations of air sampling sites in the Yangtze River**




* Air sampling point A-G-03 and A-G-08 were fixed point samples, which were in the same position as the end of sample A-G-02 and A-G-07, respectively. Therefore, they were not analyzed in this study. All analysis in this study were other 14 samples except A-G-03 and A-G-08.

**Fig. S3 The ternary plots of PAH compositions in water (a) and air (b) from the Yangtze River**

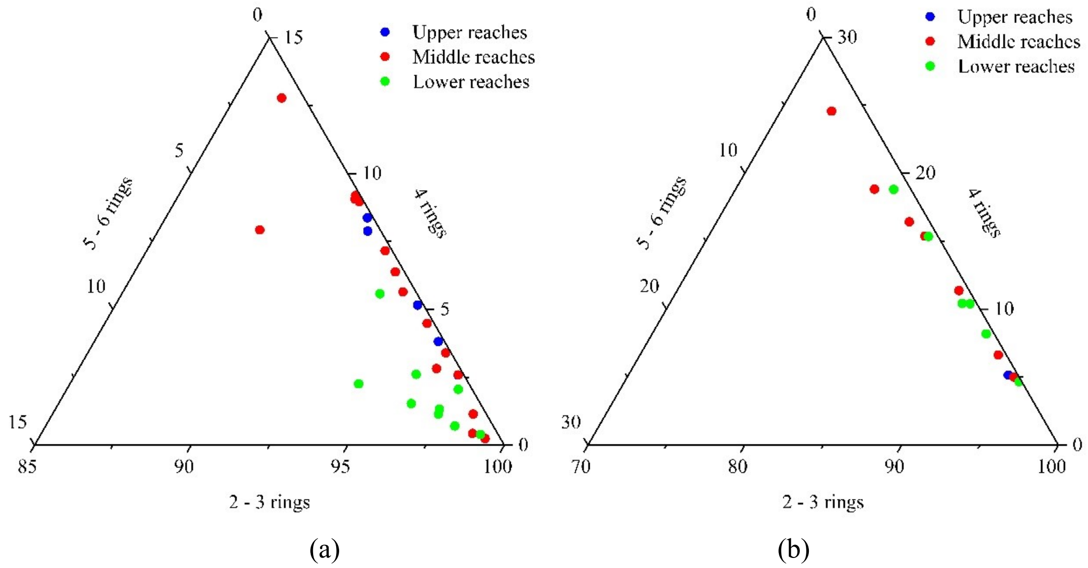



**Fig. S4 Correlation analysis between the PAHs concentration in air over the Yangtze River and GDP of along cities**

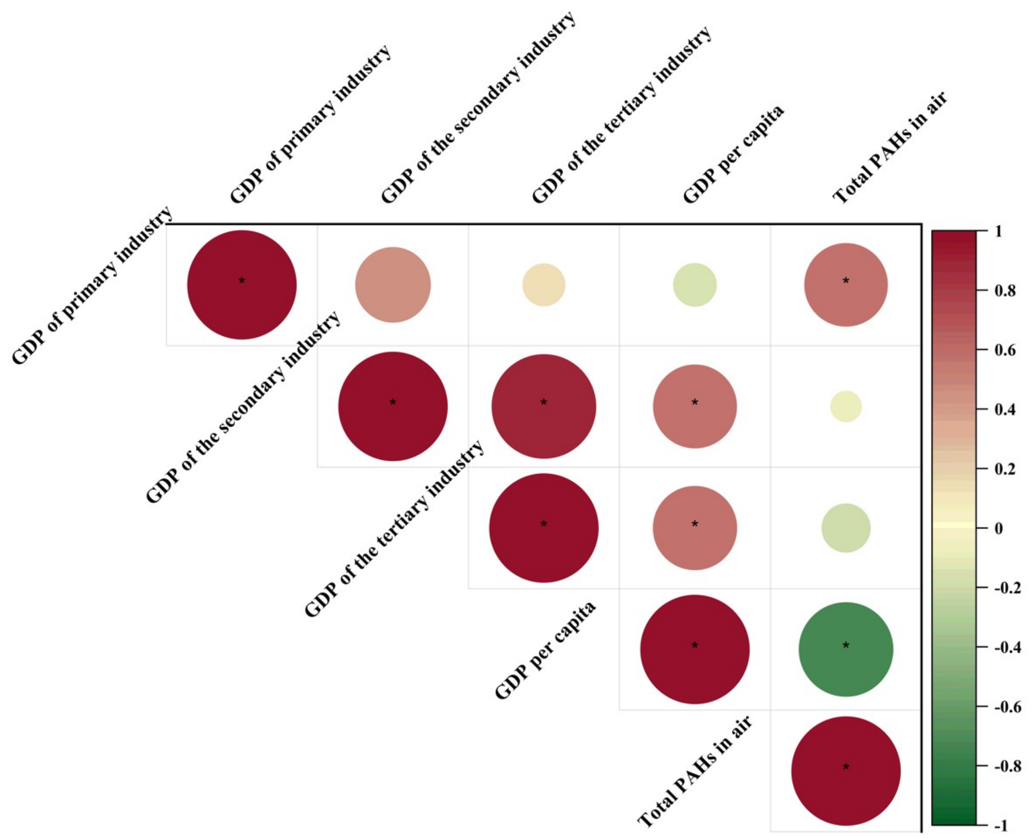


**Fig. S5 Back trajectories by NOAA HYSPLIT MODEL**

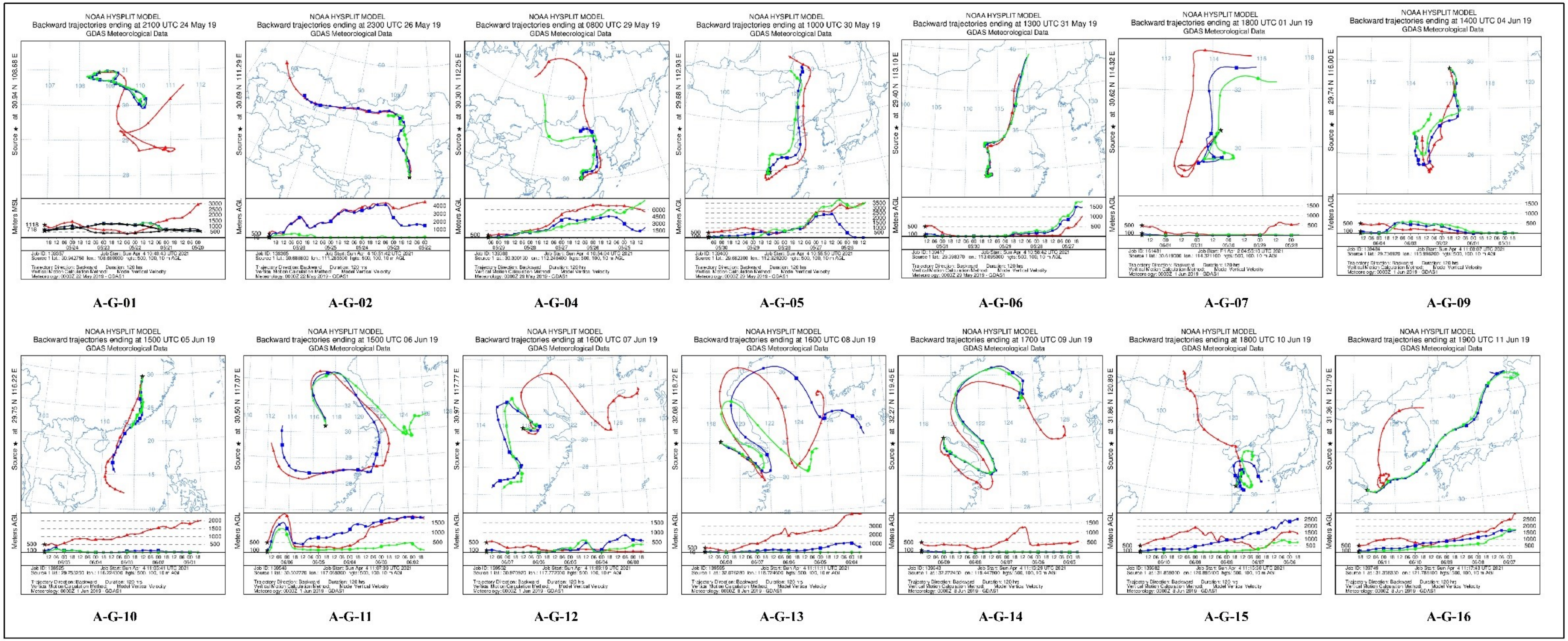


**Fig. S6 Fugacity fractions (ff) of 16 PAHs across the air-water interface during different seasons (ff =0.5 limits indicating equilibrium between the air and water)**

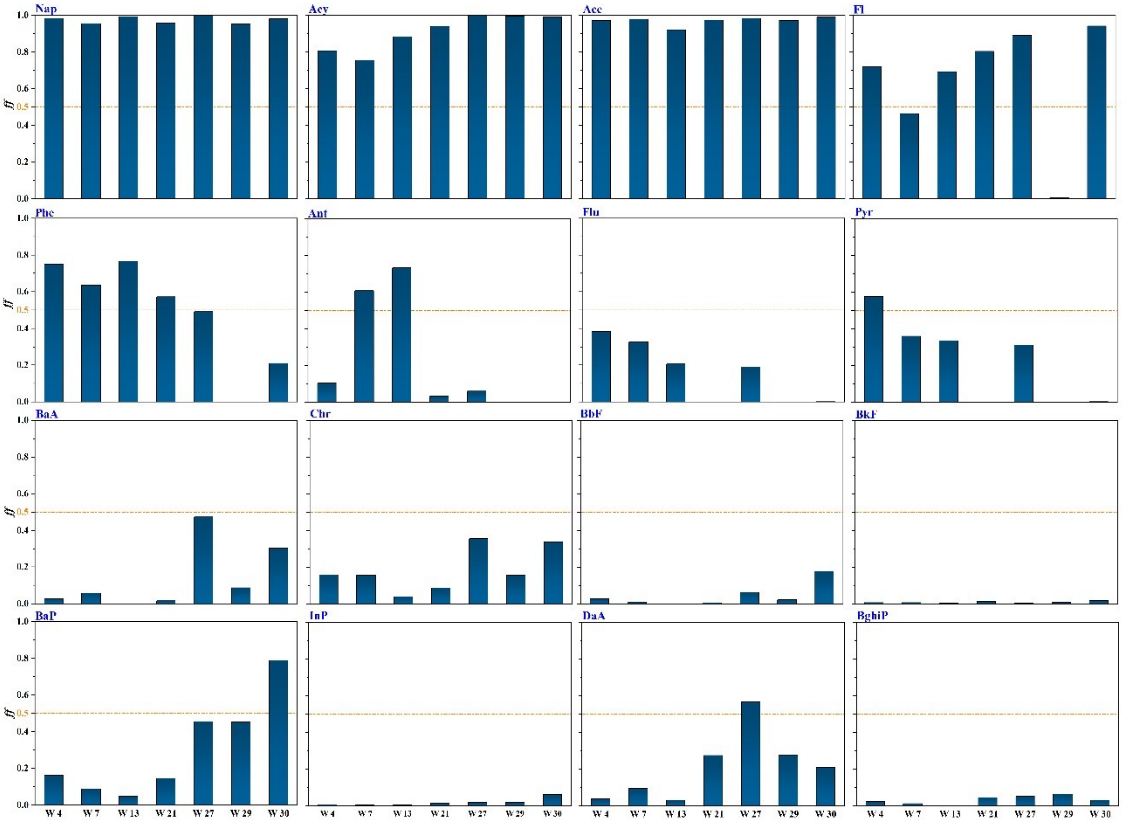
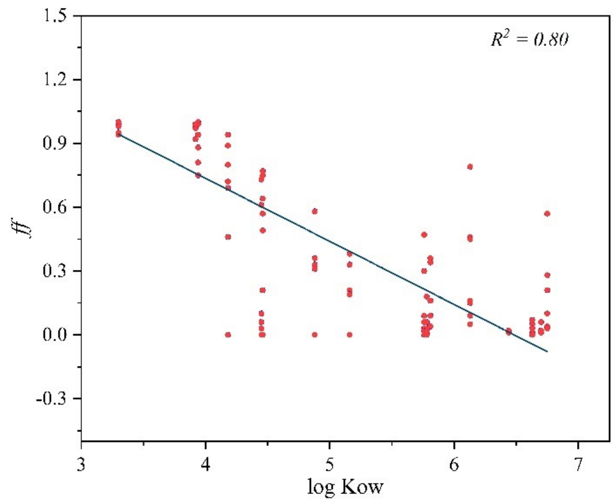



**Fig. S6 Liner regression between ff and log octanol–water partition coefficient (Kow)**



## Text S1 Extraction and instrument analysis

Water and air adsorption columns were free-dried firstly to remove the water, and then added internal standard into each sample. Spiked adsorption columns were extracted with 45 mL dichloromethane (DCM) three times by ultrasonic. Mixed extracts about 120 mL for each sample were concentrated to 1 mL under a gentle nitrogen gas flow. The concentrates were purified by the SPE cartridge (CNWBOND; ANPEL Laboratory Technologies, Shanghai, China), and concentrated again to 1 mL for instrument analysis.

Analysis for PAHs was conducted on an Agilent 7890 gas chromatograph (GC) coupled with Agilent 5,975 C mass spectrometer (MS) operated in the negative electron ionization mode. DB-5MS capillary column (30 m × 0.25 mm × 0.25 μm, Agilent Co., USA) was equipped to separate target compounds. The injection volume was 1.0 μL in the splitless mode, and helium was used as carrier gas at a flow rate of 0.9 mL/min. The temperature program was as follows: initial temperature of 80 °C for 2 min, increased at a rate of 20 °C/min to 180 °C, then held for 5 min, and raised to 300 °C at 8 °C/min followed by a hold of 4 min.

## Text S2 Air-water exchange fugacity fractions (ff) calculation

The air-water exchange of each PAHs has been studied in terms of *ff* using the formula reported elsewhere [16]:

$$f_w = C_w \times H$$

$$f_a = C_a \times RT$$

$$ff = \frac{f_w}{f_w + f_a}$$

Where Cw is the PAHs dissolved concentration in water (ng/m$^3$). Ca is the PAHs concentration in air phase (ng/m$^3$). R is the gas constant (8.314 Pa·m3/mol · K). H values were considered for 25 ℃, and T (K) is the absolute temperature. The values of H and T are given in Table S1. Theoretically, *ff* range between 0 and 1. ff value > 0.5 suggests a net volatilization from water to air, *ff* < 0.5 indicates a net deposition from water to air. Otherwise, a dynamic equilibrium is producing between air and water interface [17].

The results of the air-water exchange for PAHs using *ff* were shown in Fig. S5. The *ff* values had a wide range from 0 to 0.999. Nap, Acy and Ace displayed net volatilization from water into air. Flu, BaA, Chr, BbF, BkF, InP and BghiP showed net deposition from air into water. Other 6 PAHs exhibit complex air-water exchange process undergoing volatilization and deposition in different sampling sites. In addition, there was a potential tendency that air-water exchange process gradually transited from net volatilization to net deposition with the increase of molecular weight. For some PAHs, its air-water exchange directions were different among different site, which may be influenced by atmospheric concentration and other nature parameters, such as wind speed [18]. Additionally, a significant correlation ($R^2 = 0.80$, $p < 0.01$) was determined between *ff* and log $K_{OW}$ of PAHs (Fig. S6). This suggested that air–water exchanges of PAH depend on their molecular weight, vapor pressure, evaporation enthalpy and hydrophobic properties. Furthermore, this significant correlation also revealed the occurrences of PAHs in Yangtze River come from direct pollution source (LMW PAHs) and atmospheric transportation (HMW PAHs).

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