Materials and methods

Sample collection of rainfall

Three rainwater samples were collected during the three typhoons. When it rained, a glass beaker with a stainless-steel funnel was placed to collect rainwater. All bottles were washed with Milli-Q water and local water more than thrice before filling. The sampling bottles were completely filled and inverted, without air bubbles. Dichloromethane (DCM) was added to the samples where PAHs were detected to prevent bacterial degradation.

Quantitative and qualitative ions in GC-MS analysis

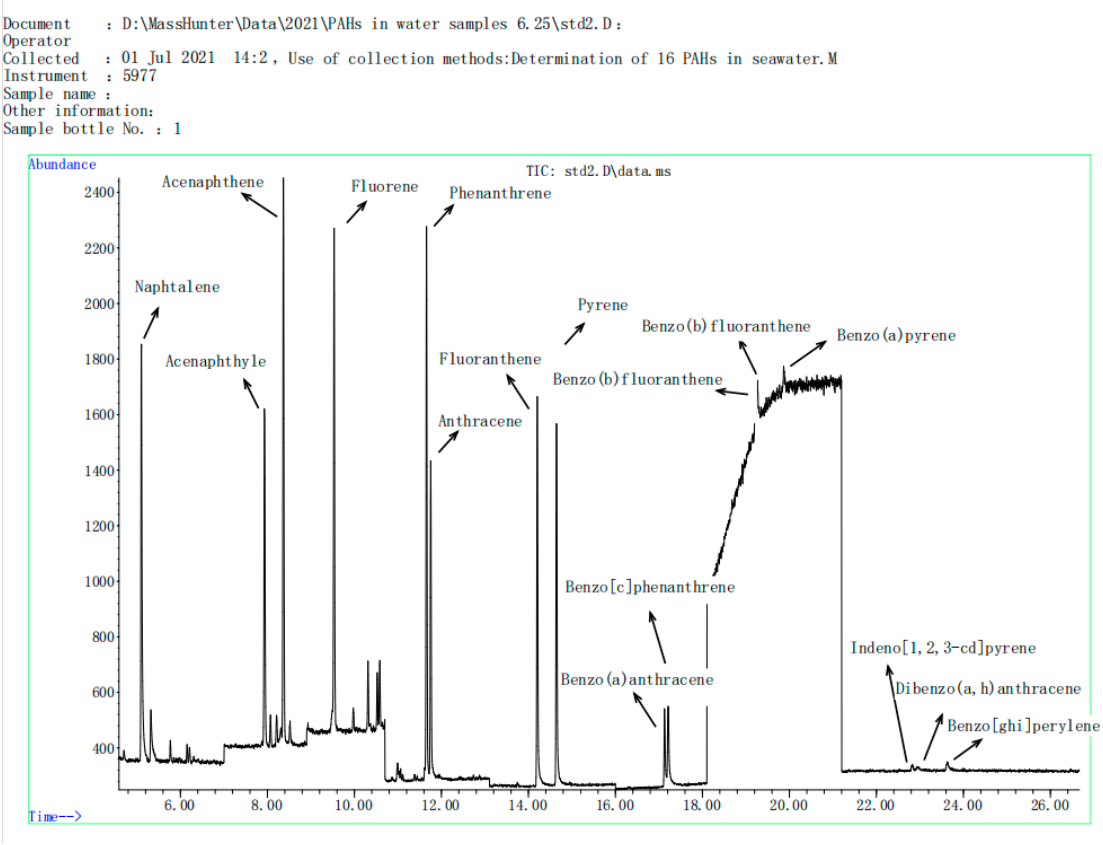
The information on target ions (quantitative and qualitative ions) in GC-MS analysis

Serial No.	Testing items	Quantitative ions	Qualitative ions
1	Nap	128	127,129
2	Acy	152	151,153
3	Ace	153	152,154
4	Flu	166	165,167
5	Phe	178	179
6	Ant	178	176,179
7	Fla	202	100,101
8	Pyr	202	200,203
9	BAa	228	226
10	Chr	228	226,229
11	BbF	252	125,253
12	BkF	252	250,253
13	BaP	252	250,253
14	DahA	276	138,277
15	InP	278	139,276
16	BghiP	276	138,274

Quality control

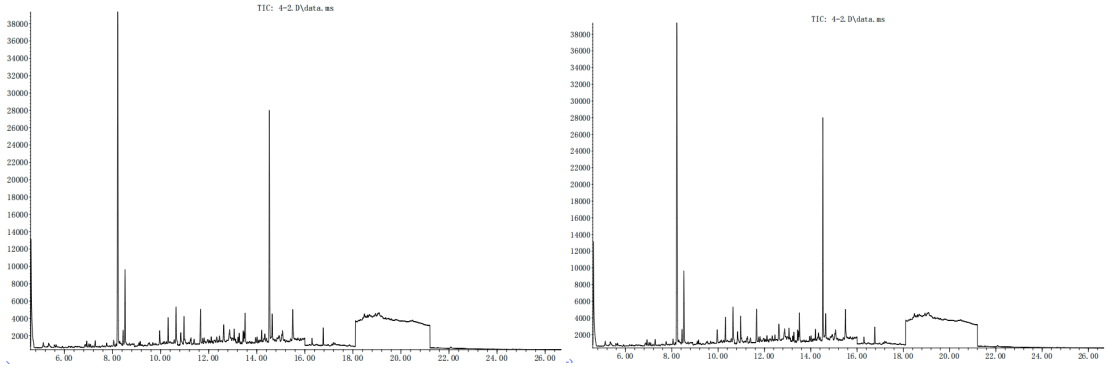
PAHs	Average	Standard deviation	Accuracy RSD(%)	Recycling rate(%)	Detection limit
Ant	2.1223	0.0890	4.19	70.7	1ng/L
Acy	3.2116	0.1498	4.66	107.1	1ng/L
Ace	2.6519	0.1041	3.93	88.4	1ng/L
Flu	2.7325	0.1284	4.70	91.1	1ng/L
Phe	2.2556	0.0941	4.17	75.2	1ng/L
Ant	3.3639	0.1780	5.29	112.1	1ng/L
Fla	2.6337	0.1119	4.25	87.8	1ng/L
Pyr	2.5992	0.0572	2.20	86.6	1ng/L
BaA	4.1934	0.2451	5.85	139.8	1ng/L
Chr	3.3432	0.2386	7.14	111.4	1ng/L
BbF	3.7315	0.2016	5.40	124.4	2ng/L
BkF	3.1169	0.3273	10.50	103.9	2ng/L
BaP	3.8347	0.2724	7.10	127.8	2ng/L
InP	3.4138	0.5954	17.44	113.8	2ng/L
DahA	5.7757	0.7434	12.87	192.5	2ng/L

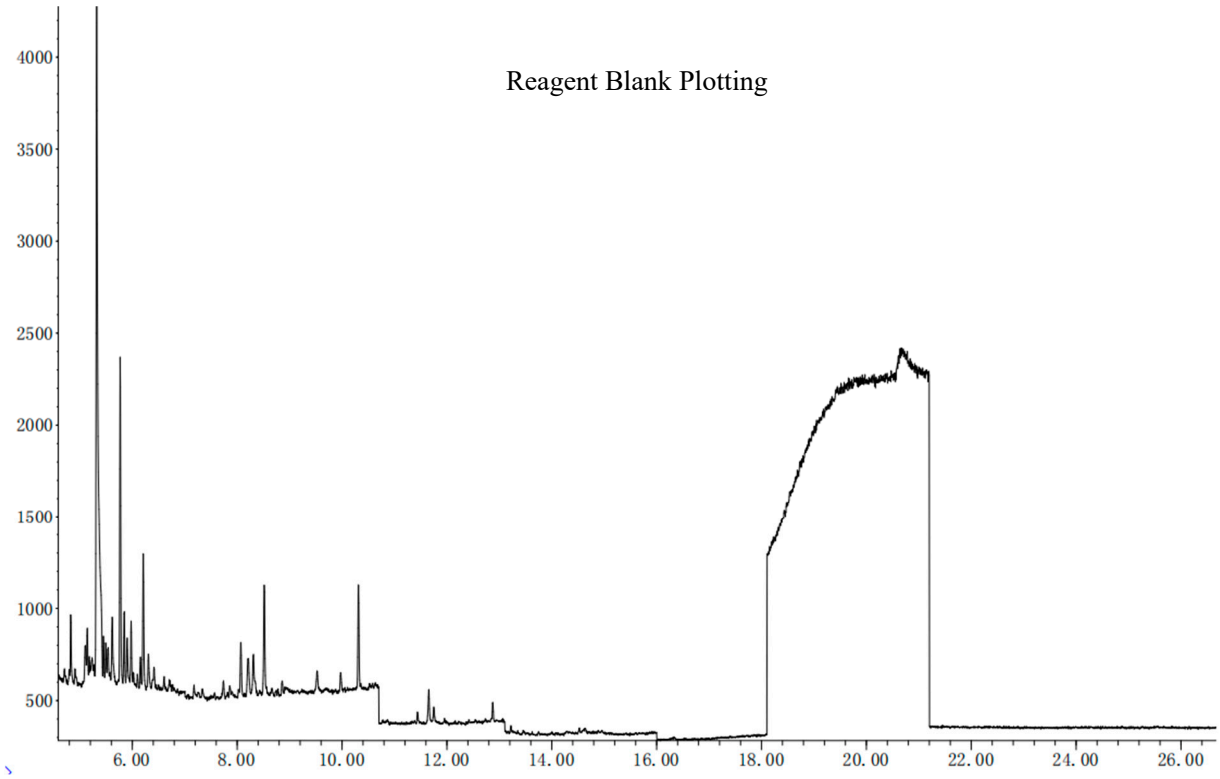
The GC-MS analyte peaks are as follows:



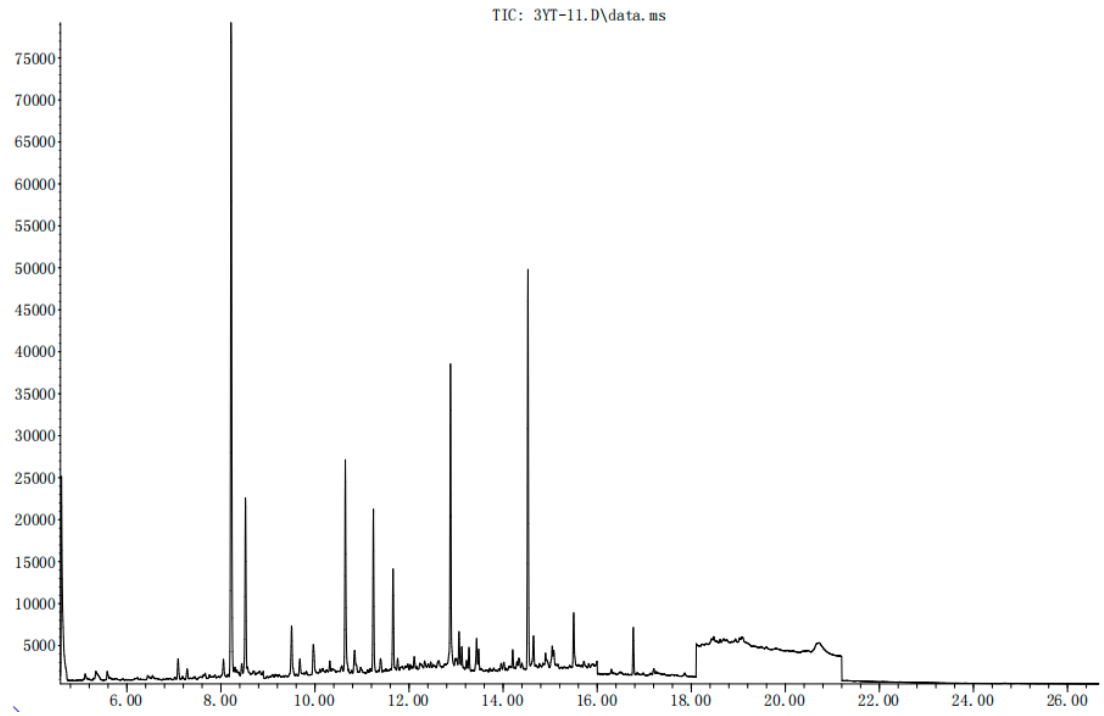
Plots of typical riverine samples and rain samples generated by GC-MS analysis

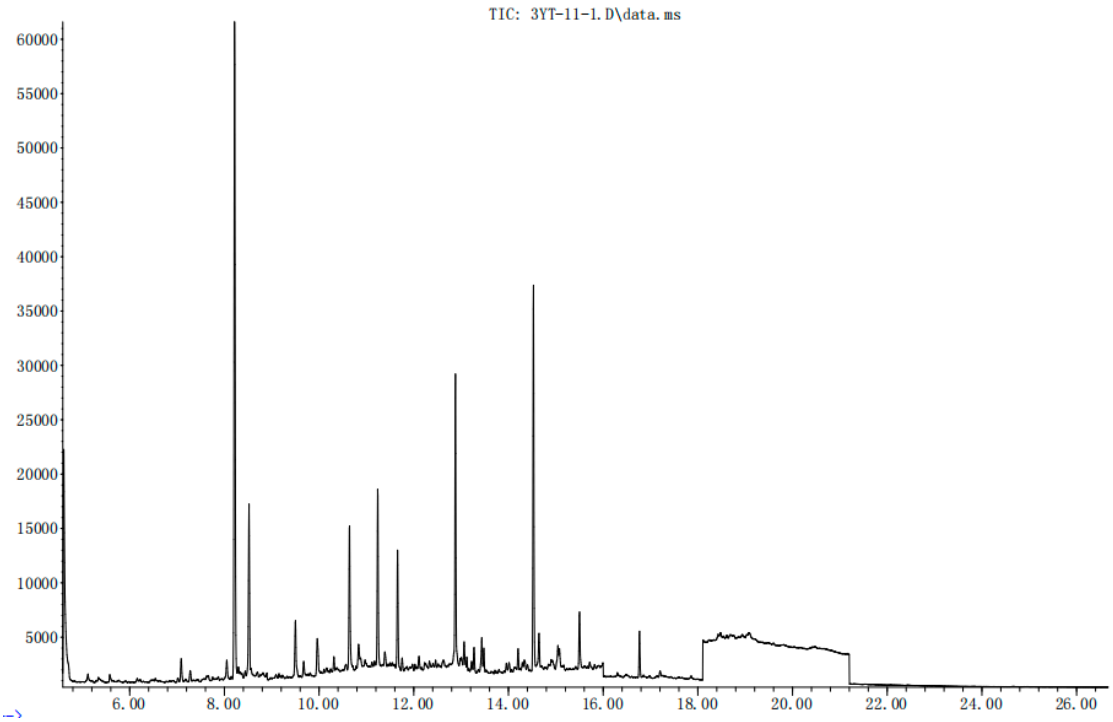
The figures below showed the plots from rain sample (left figure, collected in "Bavet") and YT2 riverine sample collected before "Bavet".



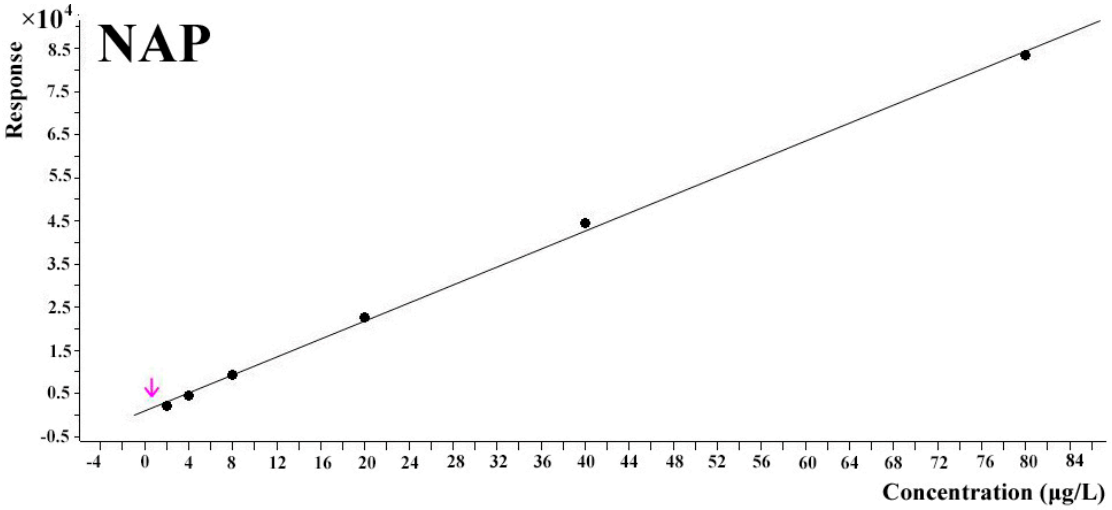


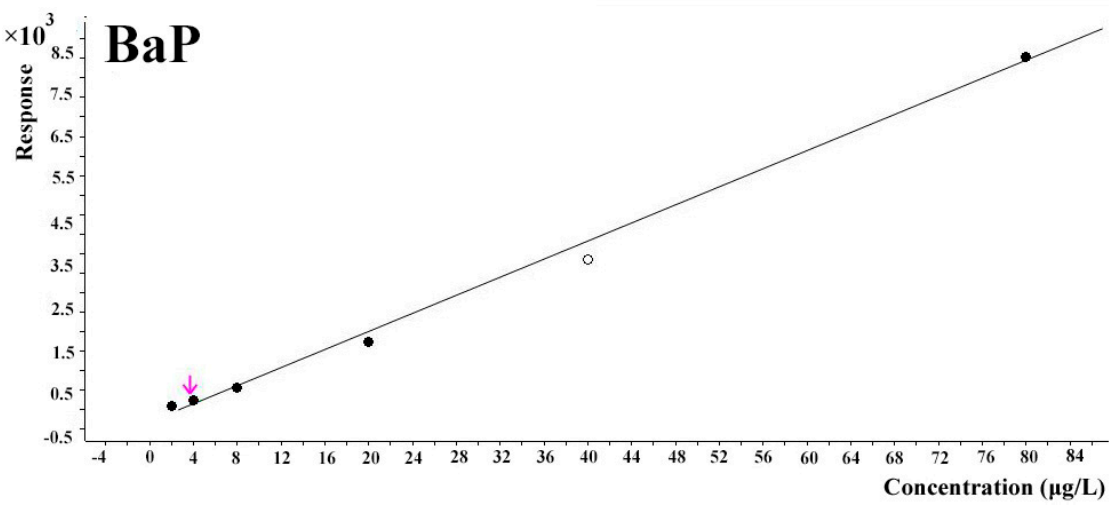
The figures below showed plots from riverine sample YT11 (up) collected before-"Poseidon" and after "Mesak", and its parallel sample (down).

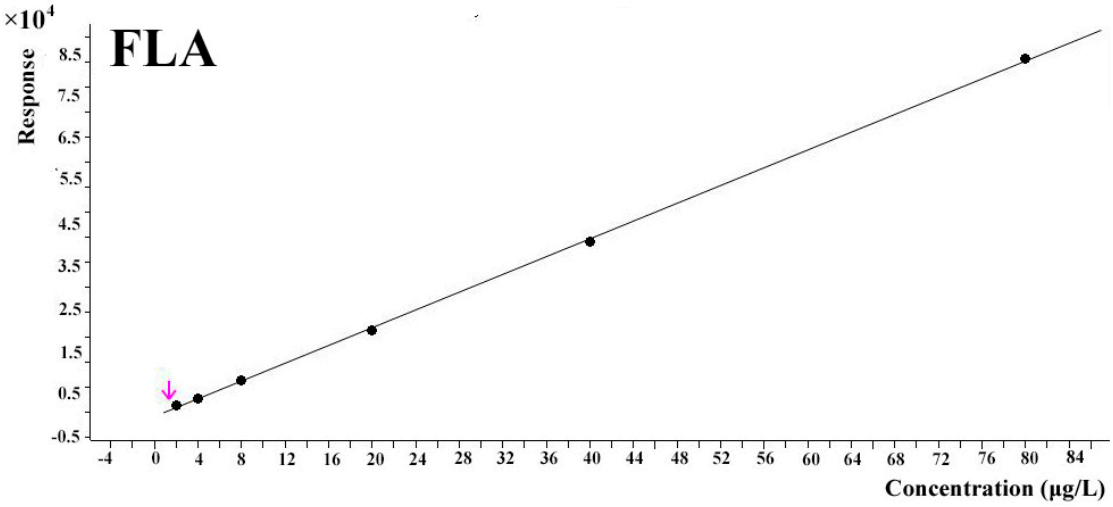


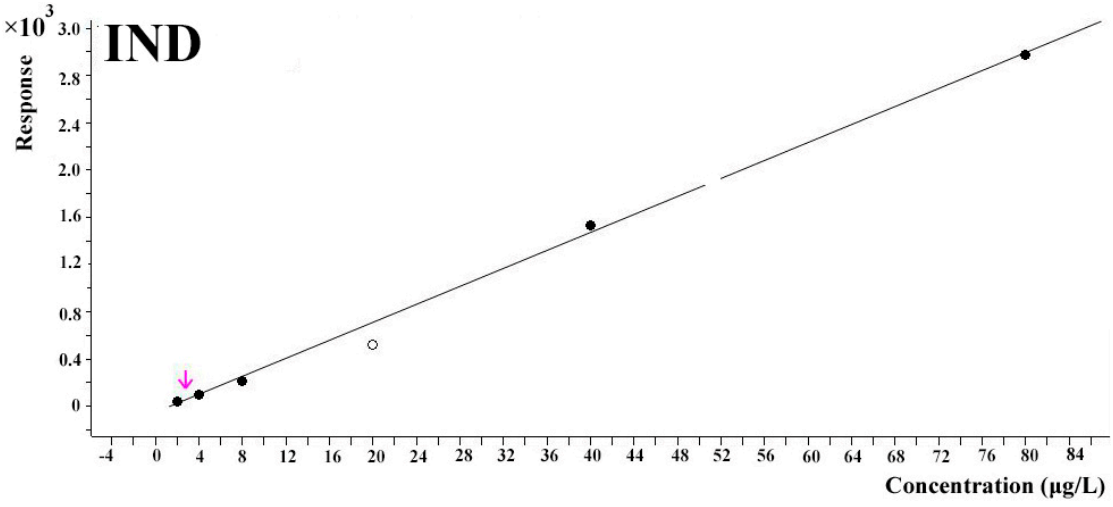


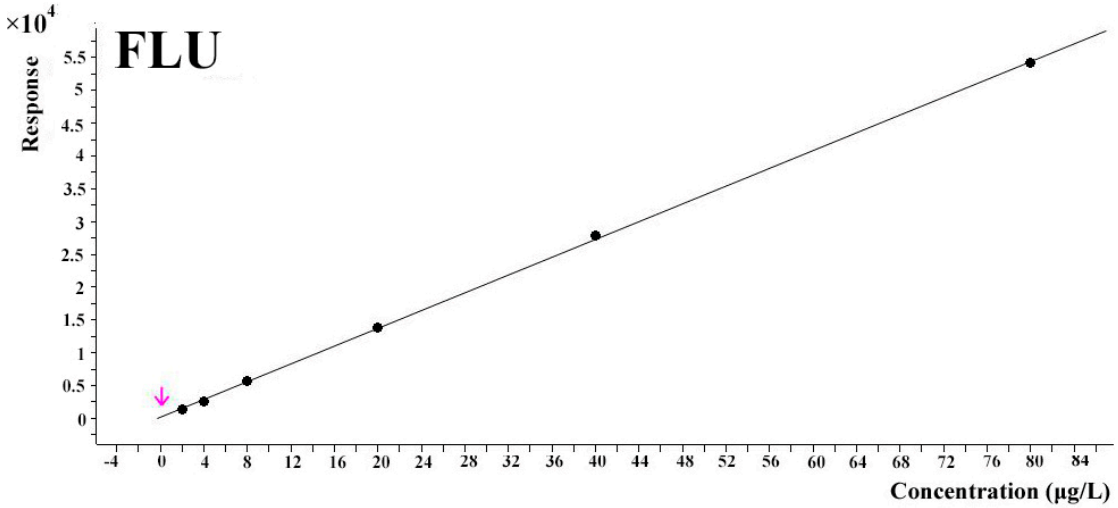
The Standard curve of PAHs in GC-MS analysis

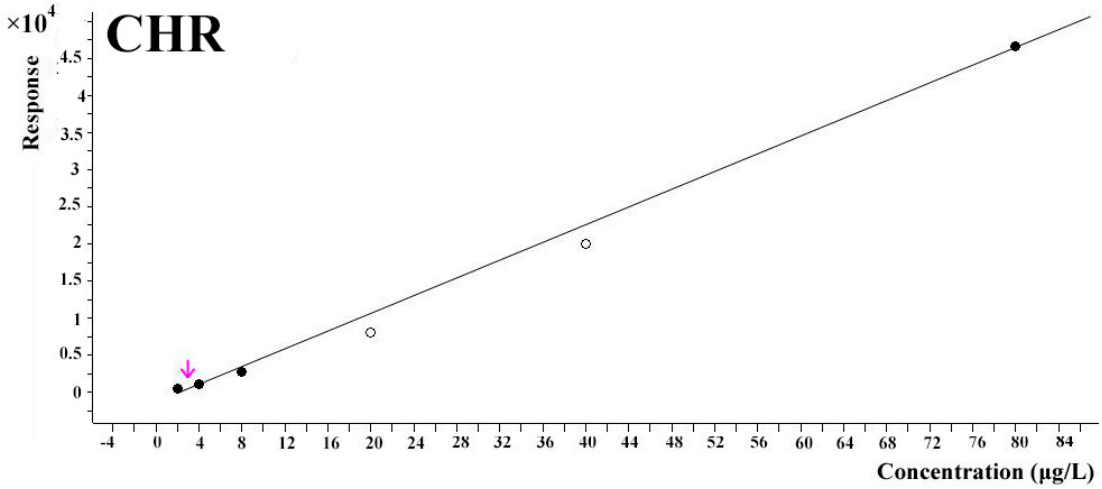


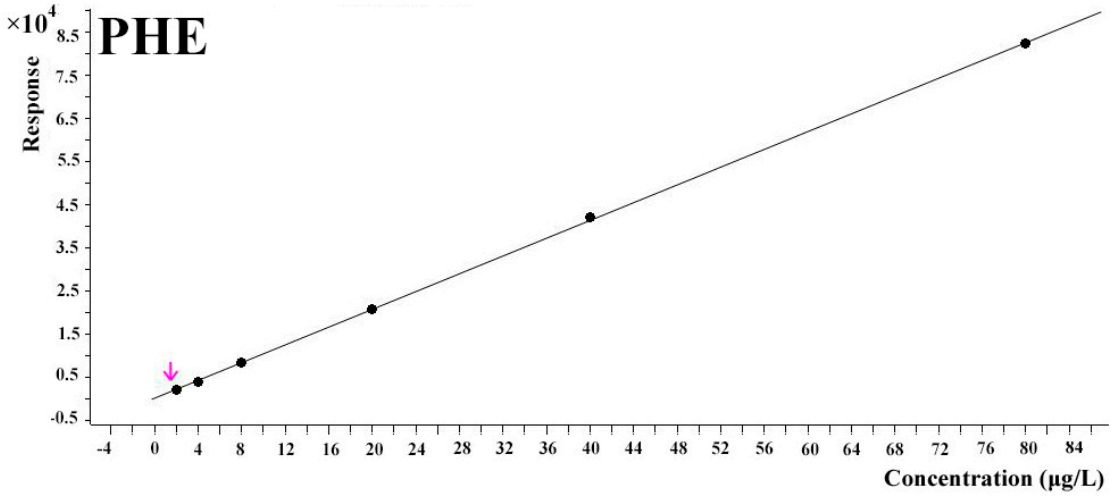

BaP
×10^3
Response
8.5
7.5
6.5
5.5
4.5
3.5
2.5
1.5
0.5
-0.5
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

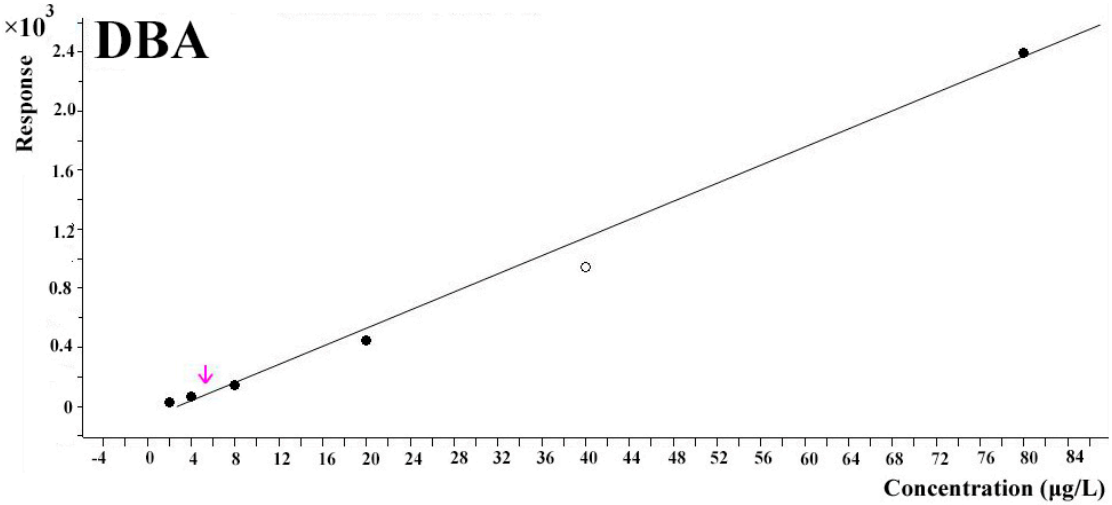

FLA
×10^4
Response
8.5
7.5
6.5
5.5
4.5
3.5
2.5
1.5
0.5
-0.5
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

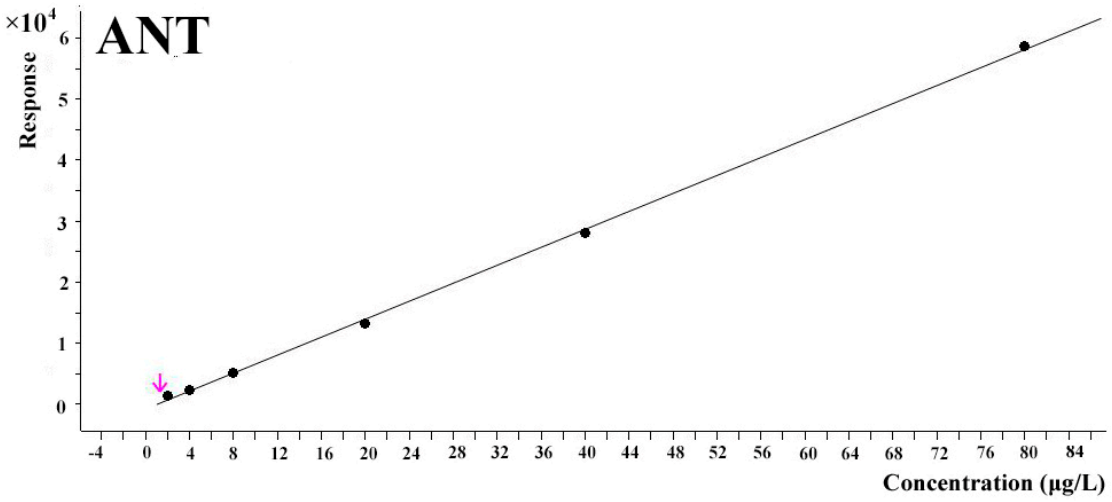

IND
×10^3
Response
3.0
2.8
2.4
2.0
1.6
1.2
0.8
0.4
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

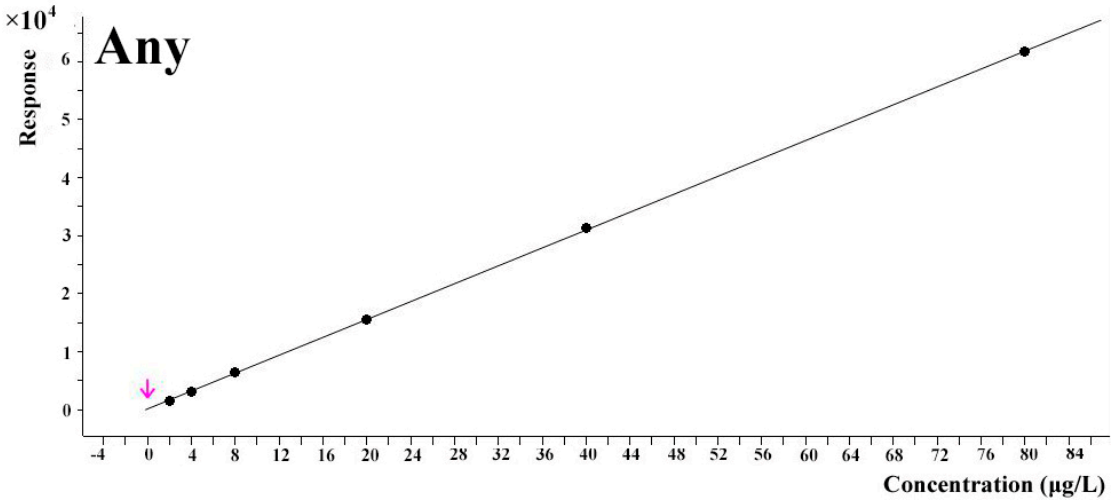

FLU
×10^4
Response
5.5
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

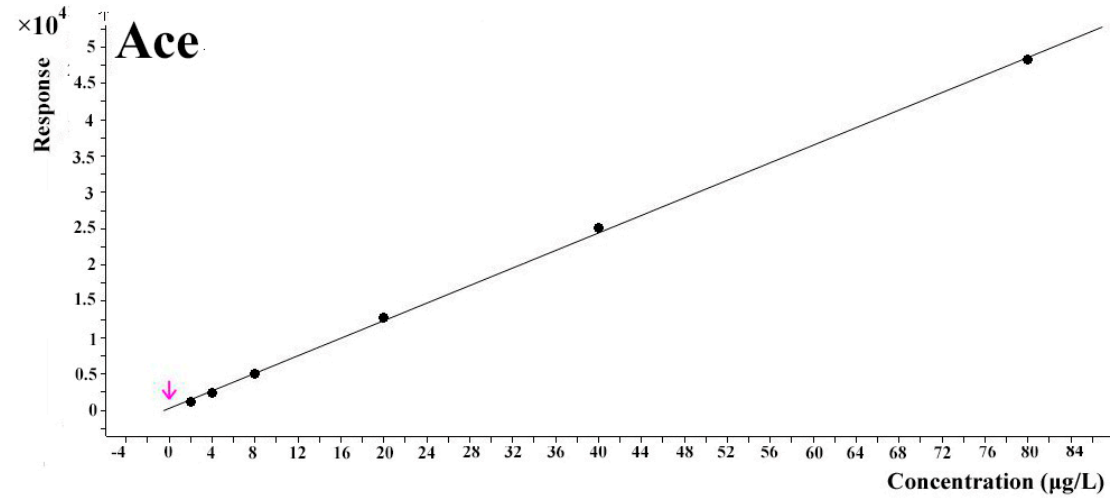

CHR
×10^4
Response
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

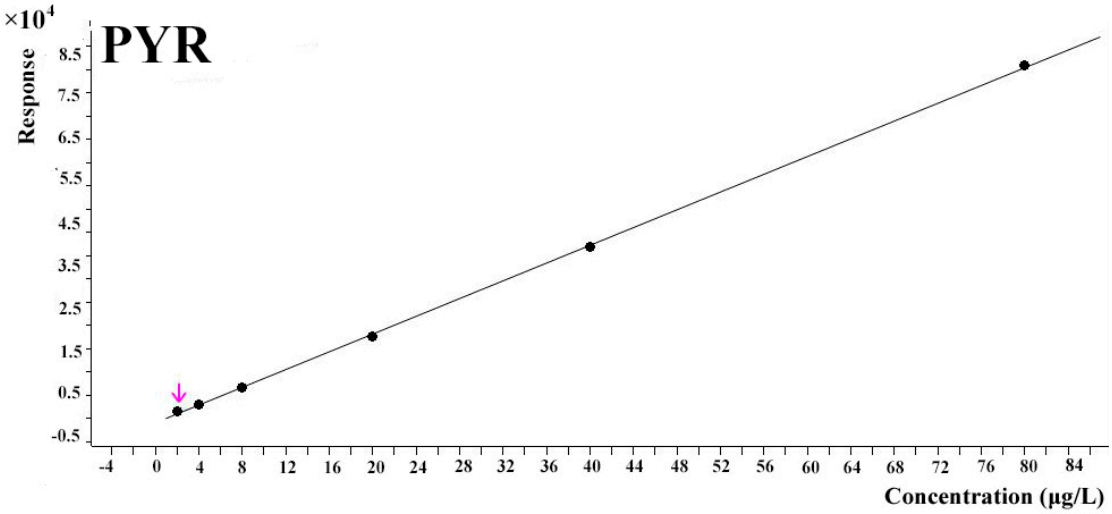

PHE
×10^4
Response
8.5
7.5
6.5
5.5
4.5
3.5
2.5
1.5
0.5
-0.5
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

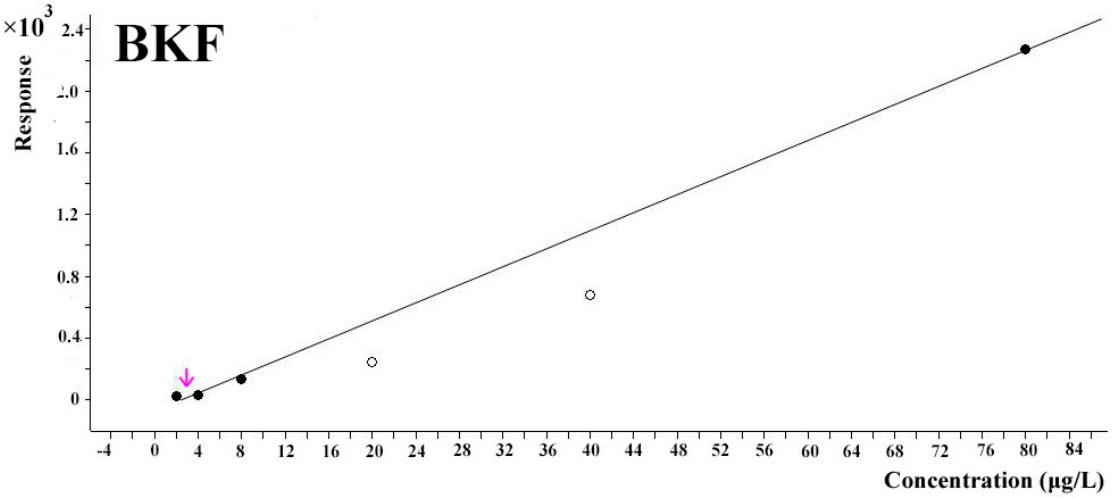

DBA
×10^3
Response
2.4
2.0
1.6
1.2
0.8
0.4
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

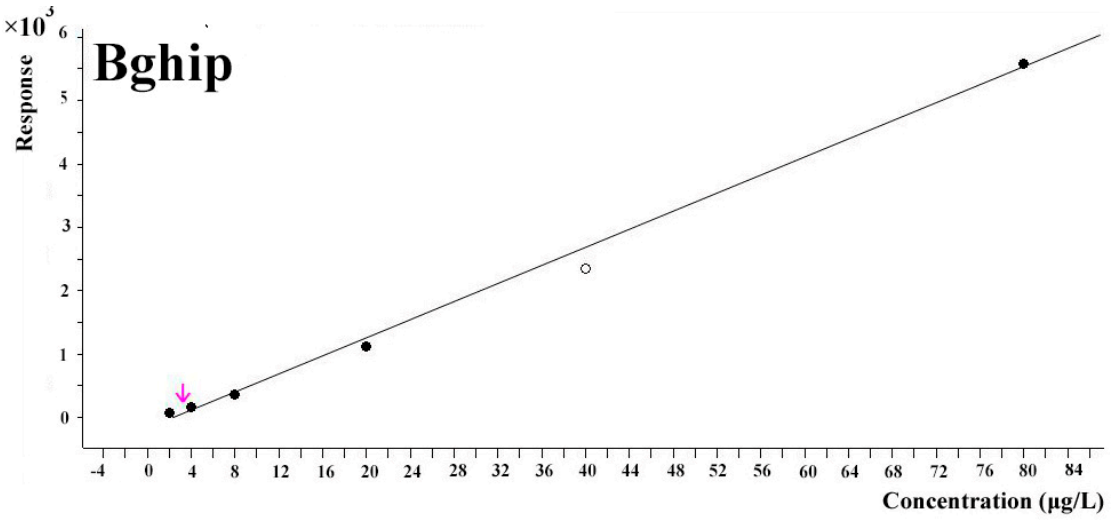

ANT
×10^4
Response
6
5
4
3
2
1
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

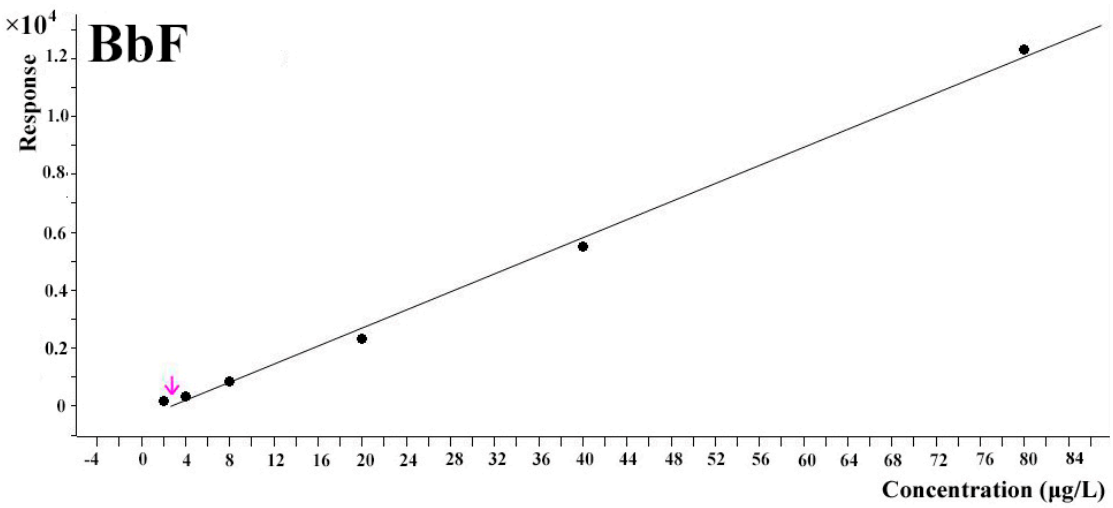

Any
×10^4
Response
6
5
4
3
2
1
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (µg/L)

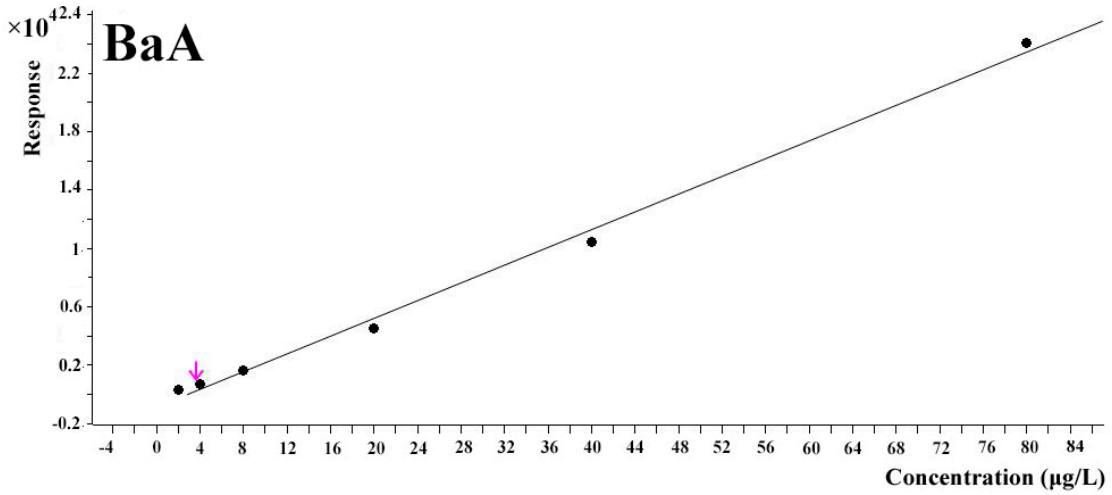

Ace
×10^4
Response
5
4.5
4
3.5
3
2.5
2
1.5
1
0.5
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (μg/L)


PYR
×10^4
Response
8.5
7.5
6.5
5.5
4.5
3.5
2.5
1.5
0.5
-0.5
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (μg/L)


BKF
×10^3
Response
2.4
2.0
1.6
1.2
0.8
0.4
0
-4 0 4 8 12 16 20 24 28 32 36 40 44 48 52 56 60 64 68 72 76 80 84
Concentration (μg/L)


Bghip
×10^5
Response
Concentration (μg/L)


BbF
×10^4
Response
Concentration (μg/L)


BaA
×10^4
Response
Concentration (μg/L)

Characteristics of 16 PAHs concentrations in samples of rainfall

The minimum, maximum and average concentrations of 16 polycyclic aromatic hydrocarbons in rainwater samples are shown in Table S1. The total concentration of polycyclic aromatic hydrocarbons (PAHs) in the first precipitation sample was 12.19 ng/L, the average concentration was 0.76 ng/L, the range was 0.64-3.64 ng/L. The 4-ring components made up 71.89 percent of the total concentration, while the 2-3-ring components made up 28.11 percent of the total concentration. The total concentration of polycyclic aromatic hydrocarbons (PAHs) collected in the second rainfall event was 28.28, with an average concentration of 1.77 ng/L and a range of 0.60 to 15.27 ng/L. Among them, components with two to three rings made up 64.1% of the overall concentration, while those with four rings made up 35.9%. The third precipitation sample has a total PAH concentration of 25.52 ng/L, an average concentration of 1.59 ng/L, and a range of 0.34-13.97 ng/L. The 2-3 ring component makes up more of the total concentration than the 4 ring component, which accounts for 56.05 percent of it. Phenanthrene concentrations were higher in the second and third sample collections.According to the overall concentrations of PAHs in the three rainy events, the average concentrations of 2-3 ring and 4 ring in rainwater were 11.96 and 10.01 ng/L, respectively. Moreover, 5-6 ring components were not detected in the three rainfall events, which may be because compared with the same group of polycyclic aromatic hydrocarbons, low-ring polycyclic aromatic hydrocarbons have higher water solubility and volatility, and are easier to enter the clouds and settle with rain [1]. The results show that this study is similar to the conclusion of Junesoo Park's previous study on atmospheric deposition in Galveston Bay, Texas, USA, that is, 2-4 rings are the main polycyclic aromatic hydrocarbons dissolved in rainwater samples, and phenanthrene is one of the most important polycyclic aromatic hydrocarbons in rainwater [2]. In addition, Wei et al. detected a high concentration of 36.9 mg/L PAHs in snow samples collected in Northeast China, which emphasized the low PAHs level found in this study. This may be due to differences in local pollution levels between the regions studied, as the PAHs emitted accumulate only near the source [3].

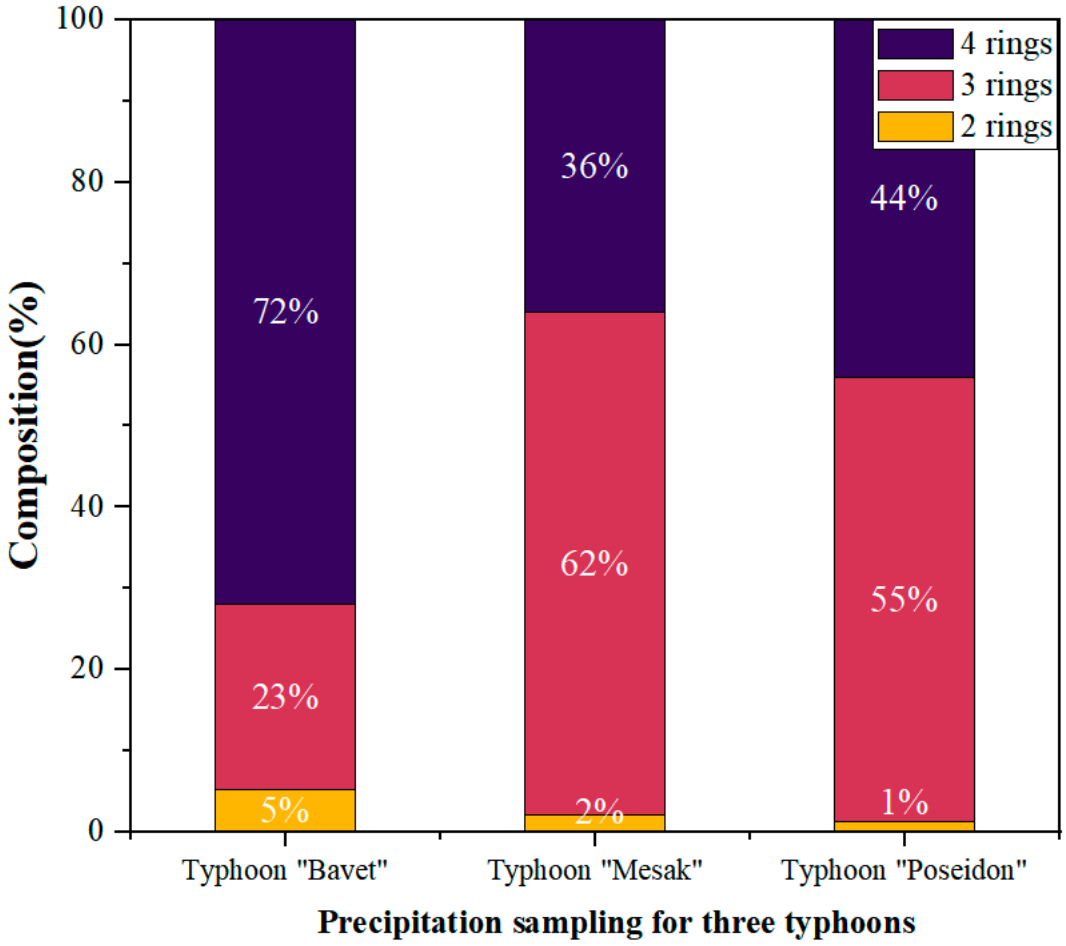


Figure S1. Distribution of polycyclic aromatic hydrocarbon concentrations in precipitation samples from three super typhoons, "Bavet", "Metsak" and "Poseidon".

Table S1. PAHs concentrations in rainwater samples from Typhoon "Bavet", "Metsak" and "Poseidon".

Component	PAHs concentrations in three consecutive typhoon samples		
	Typhoon "Bavet"	Typhoon "Maysak"	Typhoon "Poseidon"
NAP	0.64	0.60	0.34
FLU	n.d.	2.26	n.d.
PHE	1.48	**15.27**	**13.97**
ANT	1.31	n.d.	n.d.
PYR	2.17	3.45	4.21
BaA	**3.64**	3.57	3.70
CHR	2.96	3.15	3.31
$\sum_{16}$PAHs	12.20	28.30	25.53

n.d. indicates not detected.

1. Zhu, Y.; Duan, X.; Qin, N.; Lv, J.; Wu, G.; Wei, F. Health risk from dietary exposure to polycyclic aromatic hydrocarbons (PAHs) in a typical high cancer incidence area in southwest China. *Sci. Total Environ.* **2019**, *649*, 731–738.
2. Park, J.; Wade, T.L.; Sweet, S. Atmospheric Deposition of Polycyclic aromatic hydrocarbons (PAHs), Polychlorinated Biphenyls (PCBs) and Pesticides to Galveston Bay. 1999. Available online: https://tamug-ir.tdl.org/handle/1969.3/25859 (accessed on 23 December 2022).
3. Wei, S.H.I., Zhang, X.N., JIA, H.B., Feng, S.D., Yang, Z.X., Zhao, O.Y., Li, Y.L. Effective remediation of aged HMW-PAHs polluted agricultural soil by the combination of Fusarium sp. and smooth bromegrass (*Bromus inermis* Leyss). *J. Integr. Agric.* **2017**, *16*, 199–209.