Supporting Online Material for

A 150-yr record of polycyclic aromatic compounds in the Sihailongwan Maar Lake, northeast China: impacts of the socio-economic developments and pollution control

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S1. Location and site conditions

Sihailongwan Maar Lake (SHLW) is situated about 20 km southwest of Jingyu County in the southeastern Jilin Province. It is a closed, nearly circular lake (Fig. S1) in the Cenozoic Longgang volcanic area of northeast China. It has a diameter of approximately 750 m, a surface area of around 0.5 km^2, and a catchment area of about 0.7 km^2 [1]. SHLW has a bowl-shaped basin with a flat lake bottom and a maximum water depth of approximately 50 meters [2]. The lake sits at an elevation of 791 meters above sea level and is encircled by pyroclastic rock walls that rise to a height of roughly 20 meters [2]. The East Asian monsoon climate has a significant impact on the weather patterns throughout the year in SHLW and Jingyu County. Spring is characterized by dry and windy conditions, while summer is hot and rainy. Autumn brings rain and a rapid cooling of temperatures, and winter is long, dry, and cold [3]. The annual temperature and precipitation at Jingyu County meteorological station is 3.25 ℃ and ~767 mm [4].

Table S1. Blank (ng), method detection limit (LOD) (pg), and reporting limit (RL) of target polycyclic aromatic compounds in this study. Note: n.d. is not detected.

No.	Analytes	MW[a] (g mol^{-1})	MI[a]	QI[a]	LOD[b] (pg)	Blank (ng)
1	Naphthalene	128.18	128	127	0.18	3.25
2	2-Methylnaphthalene	142.2	142	141	0.09	1.24
3	1-Methylnaphthalene	142.2	142	141	0.53	3.20
4	1,3-dimethylnaphthalene	156.23	156	141	1.36	6.59
5	Acenaphthylene	152.2	152	151	0.25	0.43
6	Acenaphthene	154.21	153	154	0.89	0.01
7	Fluorene	166.22	166	165	0.98	1.56
8	Phenanthrene	178.24	178	179	1.98	3.56
9	Anthracene	178.24	178	179	1.10	3.36
10	2-Methylphenanthrene	192.26	192	191	0.67	2.01
11	3,6-Dimethylphenanthrene	206.29	206	205	1.25	0.35
12	Fluoranthene	202.26	202	200	0.12	0.56
13	Pyrene	202.26	202	200	0.18	1.89
14	Retene	234.34	219	234	1.7	0.98
15	Benzo[a]anthracene	228.3	228	229	0.38	0.76
16	Chrysene	228.3	228	229	0.22	0.34
17	Benzo[b+j]fluoranthene	252.32	252	253	0.64	0.18
18	Benzo[e]pyrene	252.32	252	253	0.26	0.19
19	Benzo[a]pyrene	252.32	252	253	0.20	0.12
20	Perylene	252.32	252	253	0.47	0.43
21	Indeno[1,2,3-cd]pyrene	276.34	276	274	0.54	0.15
22	Dibenzo[a,h]anthracene	278.36	278	279	0.79	0.97
23	Benzo[g,h,i]perylene	276.34	276	277	0.43	1.26
24	Quinoline	129.16	129	102	5.08	0.75
25	Benzo[h]quinoline	179.22	179	151	3.45	0.10.
26	Acridine	179.22	179	151	4.03	0.12
27	Carbazole	167.21	167	139	4.01	0.06
28	1-Naphthaldehyde	156.19	156	128	10.00	0.98
29	9-Fluorenone	180.21	180	152	4.26	6.54
30	9,10-Anthraquinone	208.22	208	180	6.53	5.23
31	Benzo[a]florenone	230.27	230	202	3.67	1.79
32	7H-Benzo[d,e]anthracene-7-one	230.27	230	202	6.79	2.23
33	Benzo[a]anthracene-7,12-dione	258.28	258	230	6.58	0.18
34	5,12-naphthacenequinone	258.28	258	230	5.61	1.58

Table S2. Minimum, maximum, average concentrations (ng g^{-1}) of polycyclic aromatic compounds (PACs) in sediments of the Sihailongwan Maar Lake (SHLW), northeast China.

Species	Abbreviation	Minimum (Max)	Maximum (Min)	Average	Max/Min
Polycyclic aromatic hydrocarbons	PAHs				
Naphthalene	Naph	47.6	416	142	8.7
2-Methylnaphthalene	2-MNaph	35.9	289	115	8.0
1-Methylnaphthalene	1-MNaph	28.1	220	72.1	7.8
1,3-Dimethylnaphthalene	1,3-DMNaph	2.8	11.2	6.29	3.9
Acenaphthylene	Acey	1.7	55.0	11.4	33.2
Acenaphthene	Acen	4.1	38.2	13.9	9.4
Fluorene	Fluo	21.5	190	60.1	8.9
Phenanthrene	Phe	47.5	456	151	9.6
Anthracene	Ant	2.4	44.6	11.8	18.3
2-Methylphenanthrene	2-MPhe	10.0	73.5	26.5	7.4
3,6-Dimethylphenanthrene	3,6-DMPhe	0.4	27.9	5.86	78.2
Fluoranthene	Flua	19.9	655	159	32.9
Pyrene	Pyr	13.1	482	111	36.9
Retene	Ret	13.3	131	40.7	9.9
Benzo[a]anthracene	BaA	2.6	157	39.2	60.5
Chrysene	Chr	14.8	416	106	28.1
Benzo[b+j]fluoranthene	B(bj)F	26.0	684	191	26.3
Benzo[e]pyrene	BeP	19.5	527	148	27.0
Benzo[a]pyrene	BaP	2.0	281	47.3	139
Perylene	PER	36.7	228	93.9	6.2
Indeno [1,2,3-cd]pyrene	IcdP	1.9	370	78.6	199
Dibenzo[a,h]anthracene	DahA	0.3	78.2	15.6	244
Benzo[g,h,i]perylene	BghiP	1.4	313	67.2	220
Oxygenated PAHs	OPAHs				
1-Naphthaldehyde	1-NALD	0.3	26.6	4.6	80.7
9-Fluorenone	9-Fluo	8.5	248	63.7	29.2
9,10-Anthraquinone	9,10-AQ	1.0	108	22.7	105
Benzo[a]fluorenone	BaFLU	0.3	178	25.6	661
7H-Benzo[d,e]anthracene-7-one	BdeAQ	1.2	73.1	10.0	59.0
Benzo[a]anthracene-7,12-dione	BaAQ	3.3	59.7	17.5	18.2
5,12-Naphthacenequinone	5,12-NQ	0.2	22.4	4.1	107
Azaarenes	AZAs				
Benzo[h]quinoline	BhQ	1.3	201	16.4	155
Acridine	ACR	0.8	34.2	8.2	42.2
Quinoline	QUI	0.1	315	30.9	2628
Carbazole	CAR	0.6	125	21.9	212
Sum of azaarenes	∑4AZAs	4.7	500	77.3	106
Sum of oxygenated PAHs	∑7OPAHs	35.8	639	148	17.9
Sum of 25PAHs	∑25PAHs	527	5240	1714	9.9
∑PAHs without perylene	∑24PAHs	381	5182	1620	13.6
∑7OPAHs/∑24PAHs		0.05	0.43	0.13	8.6
Low molecular weight parent PAHs	LMW-PAHs	131	1062	390	8.1
High molecular weight parent PAHs without perylene	HMW-PAHs	117	3797	964	32.4
LMW-PAHs/HMW-PAHs		0.23	2.05	0.71	8.9
∑COM-PAHs [1]		122	3212	849	26.2
∑COM-PAHs/∑24PAHs		0.22	0.64	0.45	2.95

[1] ∑COM-PAHs: Combustion-derived PAH = sum of Flua, Pyr, BaA, Chr, BeP, BaP, IcdP, BghiP, B(bj)F, and DahA.

Table S3. The toxic equivalency quotients (TEQs, ng g^{-1}) of PAHs and OPAHs in SHLW sediment core.

Year	Naph	2-MNaph	1-MNaph	Acey	Acen	Fluo	Phe	Ant	Flua	Pyr	BaA	Chr	B(bj)F	BeP	BaP	PER	IcdP	DahA	BghiP	ΣPAHs	9,10-AQ	BdeAQ	ΣOPAHs
TEF[1]	0.001	0.001	0.0025	0.001	0.001	0.0005	0.0005	0.0005	0.5	0.001	0.005	0.03	0.1	0.002	1	0.0001	0.1	1.1	0.02	□	0.018	0.0039	□
2020.50	0.26	0.18	0.29	0.04	0.03	0.08	0.12	0.01	145.19	0.20	0.27	4.46	21.30	0.33	54.22	0.01	15.85	30.72	3.25	276.81			
2019.76	0.17	0.13	0.21	0.01	0.01	0.05	0.12	0.01	177.87	0.24	0.36	5.61	25.90	0.40	68.62	0.01	18.32	34.46	3.23	335.72	0.77	0.02	0.80
2019.00	0.19	0.14	0.24	0.01	0.02	0.05	0.12	0.01	135.23	0.19	0.26	4.82	30.17	0.50	71.64	0.01	6.57	15.89	1.17	267.20	0.84	0.10	0.94
2018.28	0.20	0.12	0.22	0.02	0.02	0.05	0.09	0.01	98.08	0.13	0.18	3.27	17.33	0.28	39.47	0.00	9.04	16.22	1.73	186.46	1.03	0.14	1.18
2017.21	0.16	0.13	0.24	0.01	0.02	0.04	0.12	0.01	147.22	0.21	0.32	5.55	29.69	0.46	74.41	0.01	10.34	20.62	2.54	292.09	0.87	0.04	0.91
2015.35	0.20	0.14	0.24	0.02	0.02	0.05	0.13	0.01	129.18	0.17	0.31	5.08	31.85	0.48	69.06	0.01	6.68	21.05	1.27	265.94	0.56	0.01	0.57
2012.81	0.42	0.23	0.35	0.03	0.02	0.10	0.09	0.01	104.84	0.14	0.32	5.46	29.50	0.45	79.92	0.01	13.58	37.42	3.15	276.00	0.84	0.04	0.88
2011.22	0.29	0.25	0.38	0.01	0.02	0.04	0.14	0.01	137.32	0.17	0.45	7.00	45.39	0.68	102.18	0.01	15.36	27.80	2.62	340.11	1.14	0.29	1.43
2009.52	0.39	0.26	0.55	0.03	0.03	0.07	0.22	0.02	249.40	0.32	0.55	9.89	60.13	0.83	281.25	0.01	18.52	53.25	3.99	679.70	1.37	0.01	1.39
2007.43	0.39	0.29	0.44	0.02	0.03	0.06	0.20	0.02	241.74	0.33	0.61	9.98	52.58	0.92	137.52	0.01	22.41	85.97	4.65	558.15	1.53	0.05	1.58
2005.32	0.22	0.15	0.24	0.02	0.03	0.05	0.13	0.01	171.98	0.22	0.34	8.02	38.56	0.65	77.79	0.00	10.24	17.58	2.21	328.45	0.68	0.02	0.70
2003.45	0.19	0.16	0.25	0.03	0.02	0.06	0.14	0.01	170.21	0.22	0.45	8.00	41.08	0.68	103.55	0.00	24.09	55.84	4.00	408.98	0.65	0.01	0.66
2001.41	0.28	0.21	0.33	0.04	0.03	0.07	0.17	0.02	234.14	0.33	0.46	9.13	43.46	0.70	104.17	0.00	28.42	59.23	5.06	486.26	0.97	0.06	1.04
1998.24	0.18	0.16	0.23	0.05	0.04	0.06	0.17	0.02	240.53	0.34	0.61	9.51	56.51	0.82	266.32	0.01	20.38	49.12	3.65	648.73	1.03	0.21	1.23
1995.67	0.21	0.16	0.25	0.03	0.04	0.07	0.23	0.02	327.71	0.48	0.78	12.48	64.47	1.05	171.21	0.01	37.00	67.88	6.25	690.31	1.94	0.08	2.01
1993.21	0.13	0.12	0.19	0.01	0.03	0.05	0.17	0.01	215.60	0.30	0.66	8.99	57.58	0.84	126.98	0.00	14.82	29.48	2.24	458.19	0.93	0.07	1.00
1989.44	0.13	0.13	0.23	0.02	0.03	0.05	0.16	0.01	217.84	0.31	0.73	8.89	68.38	0.97	143.85	0.01	8.36	22.46	1.47	474.00	0.69	0.01	0.70
1982.72	0.13	0.15	0.21	0.01	0.01	0.02	0.09	0.01	123.08	0.21	0.44	4.84	35.49	0.51	85.47	0.01	8.80	19.81	1.37	280.65	0.37	0.03	0.40
1980.02	0.11	0.14	0.19	0.01	0.01	0.02	0.07	0.01	88.08	0.14	0.29	3.59	24.77	0.38	62.95	0.01	6.97	15.84	1.11	204.68	0.37	0.04	0.41
1978.31	0.16	0.11	0.15	0.01	0.01	0.02	0.06	0.00	74.86	0.11	0.16	2.48	15.08	0.23	34.22	0.01	4.96	9.33	0.81	142.79	0.46	0.08	0.54
1977.13	0.11	0.10	0.15	0.01	0.01	0.03	0.08	0.01	105.54	0.16	0.25	4.28	25.50	0.38	60.10	0.01	17.55	29.78	2.60	246.66			
1975.46	0.10	0.09	0.13	0.01	0.01	0.02	0.07	0.01	93.56	0.15	0.22	3.66	21.41	0.33	51.25	0.00	13.93	23.52	2.11	210.60	0.21	0.04	0.25
1973.43	0.11	0.11	0.14	0.01	0.01	0.03	0.08	0.01	106.22	0.16	0.24	3.84	22.62	0.33	51.44	0.01	15.04	21.59	2.38	224.35	0.13	0.01	0.15
1972.00	0.18	0.15	0.22	0.01	0.02	0.03	0.09	0.01	93.88	0.14	0.26	3.97	26.52	0.40	57.58	0.01	16.80	33.55	2.33	236.15	0.17	0.01	0.19
1970.57	0.13	0.11	0.18	0.02	0.02	0.04	0.10	0.01	105.36	0.15	0.29	4.45	31.21	0.44	60.39	0.01	9.43	18.12	1.46	231.89	0.13	0.01	0.15
1968.90	0.08	0.06	0.10	0.01	0.01	0.02	0.07	0.01	76.15	0.11	0.21	3.19	22.04	0.31	44.97	0.01	9.24	15.67	1.32	173.57	0.15	0.01	0.16
1966.57	0.10	0.10	0.14	0.01	0.01	0.02	0.06	0.00	49.41	0.07	0.14	2.07	14.34	0.19	25.10	0.01	5.63	8.70	0.84	106.94	0.07	0.03	0.10
1963.88	0.13	0.11	0.16	0.01	0.01	0.03	0.06	0.00	45.99	0.06	0.13	2.11	18.85	0.49	27.36	0.01	5.80	11.37	0.91	113.57	0.42	0.03	0.45
1961.88	0.11	0.09	0.16	0.01	0.01	0.02	0.05	0.00	37.50	0.02	0.09	1.43	12.23	0.15	21.03	0.01	2.84	9.77	0.31	85.81	0.13	0.03	0.16
1959.82	0.05	0.04	0.07	0.01	0.01	0.01	0.03	0.00	31.72	0.05	0.07	1.12	6.85	0.10	15.29	0.01	4.04	7.63	0.51	67.60	0.11	0.07	0.19
1956.88	0.10	0.09	0.12	0.01	0.01	0.02	0.05	0.00	41.44	0.07	0.11	1.62	10.72	0.15	19.88	0.01	5.47	9.66	0.91	90.42	0.03	0.02	0.05
1953.77	0.07	0.08	0.10	0.02	0.01	0.02	0.04	0.01	37.53	0.06	0.11	1.36	9.79	0.14	22.65	0.01	6.11	13.89	0.99	92.98	0.04	0.01	0.05
1950.63	0.07	0.06	0.12	0.01	0.01	0.02	0.03	0.00	21.23	0.03	0.06	0.84	6.48	0.09	12.85	0.01	3.43	11.31	0.45	57.10	0.03	0.03	0.06
1947.60	0.10	0.08	0.11	0.00	0.01	0.01	0.04	0.00	20.97	0.03	0.04	1.00	7.07	0.10	11.85	0.01	3.23	5.13	0.46	50.25	0.04	0.03	0.07
1944.49	0.09	0.07	0.10	0.01	0.01	0.01	0.03	0.00	15.34	0.02	0.04	0.73	5.70	0.08	8.27	0.01	3.31	1.91	0.30	36.05	0.04	0.01	0.05
1941.56	0.12	0.09	0.14	0.00	0.01	0.01	0.03	0.00	15.18	0.02	0.04	0.65	4.91	0.07	8.02	0.01	2.66	2.83	0.37	35.13	0.08	0.02	0.10
1940.14	0.09	0.09	0.12	0.00	0.01	0.02	0.03	0.00	18.51	0.02	0.04	0.85	5.54	0.09	10.30	0.01	3.24	4.40	0.46	43.83	0.02	0.01	0.04
1936.66	0.12	0.13	0.20	0.00	0.01	0.02	0.03	0.00	20.57	0.02	0.04	0.72	4.75	0.06	6.87	0.01	4.11	6.38	0.50	44.55	0.03	0.01	0.04
1933.59	0.09	0.10	0.11	0.00	0.01	0.02	0.04	0.00	20.34	0.02	0.04	0.76	5.49	0.07	7.76	0.01	2.59	3.59	0.36	41.42	0.03	0.01	0.04
1931.53	0.14	0.08	0.15	0.01	0.01	0.02	0.04	0.00	16.52	0.02	0.03	0.57	4.13	0.06	9.18	0.01	1.84	2.13	0.26	35.20	0.05	0.01	0.06
1928.94	0.13	0.10	0.17	0.00	0.01	0.02	0.03	0.00	15.63	0.02	0.03	0.55	3.61	0.06	6.44	0.01	2.04	5.86	0.26	34.98	0.09	0.01	0.10
1925.15	0.12	0.08	0.15	0.00	0.00	0.02	0.05	0.00	36.78	0.05	0.06	1.16	6.22	0.10	8.34	0.01	2.70	16.60	0.32	72.77	0.19	0.02	0.21
1921.00	0.08	0.07	0.11	0.00	0.02	0.02	0.04	0.00	17.90	0.02	0.03	0.71	4.39	0.07	9.87	0.01	1.28	0.35	0.19	35.18			
1918.65	0.10	0.07	0.12	0.01	0.01	0.02	0.04	0.00	20.02	0.03	0.04	0.82	5.42	0.07	5.74	0.01	2.82	4.08	0.35	39.77			
1916.16	0.09	0.07	0.12	0.00	0.01	0.02	0.04	0.00	18.22	0.02	0.03	0.68	3.96	0.06	5.02	0.02	1.80	4.91	0.20	35.27	0.09	0.02	0.11
1913.13	0.16	0.10	0.16	0.00	0.01	0.01	0.04	0.00	16.97	0.02	0.04	0.67	3.76	0.07	3.85	0.02	1.10	8.21	0.13	35.33	0.07	0.02	0.10
1910.58	0.05	0.04	0.07	0.00	0.01	0.01	0.02	0.00	10.74	0.01	0.02	0.55	3.24	0.05	4.27	0.01	1.49	0.64	0.18	21.43	0.04	0.01	0.05
1906.95	0.10	0.12	0.18	0.00	0.01	0.02	0.03	0.00	14.32	0.02	0.02	0.47	2.92	0.04	3.04	0.01	1.64	4.85	0.24	28.04			
1904.36	0.11	0.12	0.17	0.00	0.01	0.02	0.03	0.00	12.09	0.02	0.02	0.48	2.93	0.04	5.06	0.01	2.06	3.36	0.21	26.73			
1900.44	0.08	0.08	0.13	0.01	0.01	0.02	0.04	0.00	13.95	0.02	0.03	0.65	4.43	0.18	7.20	0.01	1.07	2.05	0.14	30.09	0.87	0.02	0.89
1897.07	0.06	0.06	0.10	0.00	0.01	0.01	0.04	0.00	11.30	0.01	0.03	0.44	2.66	0.04	2.03	0.01	0.63	1.80	0.04	19.29	0.20	0.01	0.21
1894.07	0.07	0.07	0.09	0.00	0.01	0.02	0.03	0.00	9.97	0.01	0.02	0.59	3.74	0.06	4.31	0.01	0.41	2.43	0.18	22.01	0.12	0.02	0.14
1891.29	0.11	0.09	0.14	0.01	0.01	0.02	0.03	0.00	14.31	0.02	0.05	0.69	4.19	0.09	4.56	0.02	1.43	5.11	0.18	31.04	0.02	0.02	0.04
1888.80	0.08	0.07	0.10	0.00	0.01	0.01	0.03	0.00	13.89	0.02	0.03	0.60	2.60	0.05	4.42	0.02	1.23	2.24	0.18	25.60			
1886.90	0.08	0.06	0.09	0.01	0.01	0.01	0.03	0.00	11.75	0.01	0.01	0.55	2.70	0.07	5.92	0.01	0.19	4.54	0.03	26.06	0.03	0.03	0.06
1884.99	0.16	0.16	0.22	0.00	0.01	0.02	0.04	0.00	12.30	0.02	0.02	0.54	3.79	0.05	3.28	0.02	0.37	1.49	0.15	22.64	0.17	0.04	0.21
1883.08	0.09	0.08	0.11	0.00	0.01	0.02	0.04	0.00	18.57	0.02	0.03	0.73	4.14	0.07	7.53	0.02	1.63	1.15	0.25	34.51	0.12	0.02	0.13
1881.17	0.08	0.09	0.13	0.01	0.01	0.02	0.04	0.00	14.06	0.02	0.02	0.64	4.61	0.07	6.37	0.02	0.57	10.47	0.11	37.33	0.20	0.00	0.20
1879.26	0.11	0.09	0.13	0.00	0.00	0.01	0.03	0.00	17.89	0.02	0.03	0.68	4.13	0.05	4.57	0.02	2.28	2.36	0.34	32.76	0.10	0.03	0.13

[1] TEF: Toxic equivalency factor, the data was acquired from [5-7]

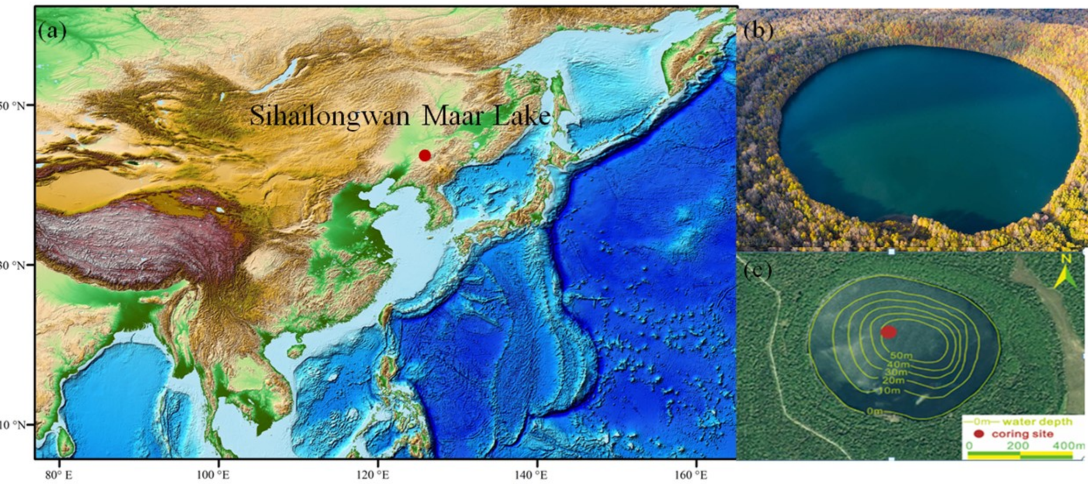

Fig. S1. Location of Sihailongwan Maar Lake (SHLW) in northeast China (a). Aerial view of SHLW and its forested catchment (b). The sampling location in the lake (c) according to Han et al., [8].

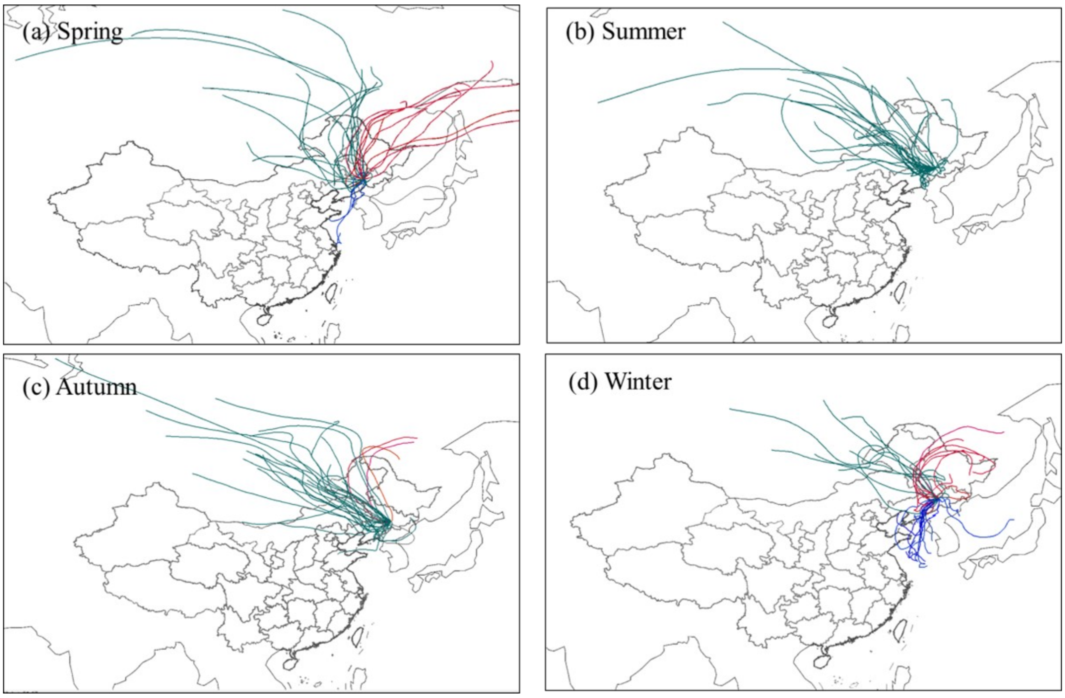

Fig. S2. 72-hour backward air mass trajectories passing over Sihailongwan Maar Lake (SHLW) in (a) spring (April); (b) summer (June); (c) autumn (October); and (d) winter (January) 2020 (produced using the NOAA ARL website: www.arl.noaa.gov/ready/). The trajectories are divided into northeasterly air mass inflow (red) from Russia's far East, southeasterly inflow (blue) from East Asia, and northwesterly inflow (green) from northern Mongolia.

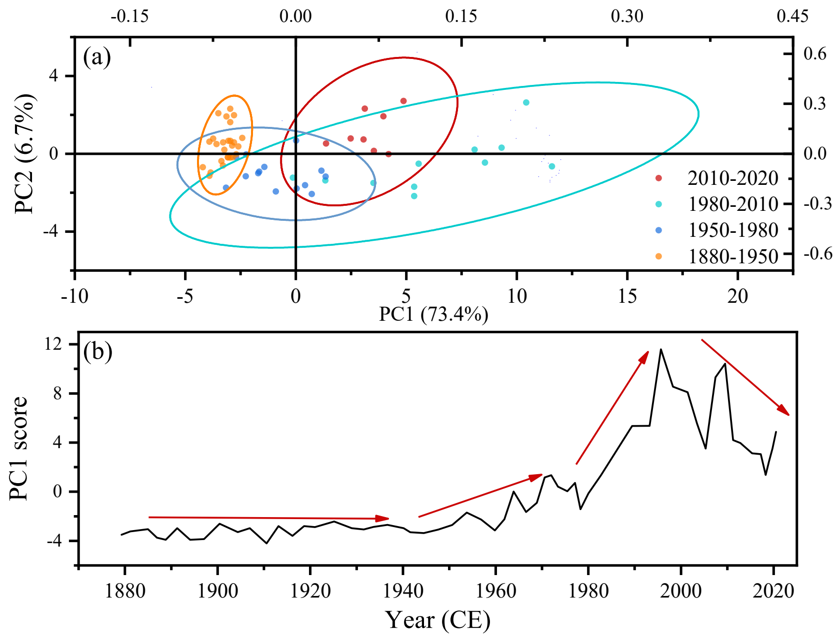

Fig. S3 PCA (Principal Component Analysis analyses) of PAHs (a) Classification of temporal variations based on the scores of PCA (principle component analysis) components PC1 (principal component 1, the dominant component) and PC2 (principal component 2); (b) temporal variations of PC1 in SHLW sediment core.

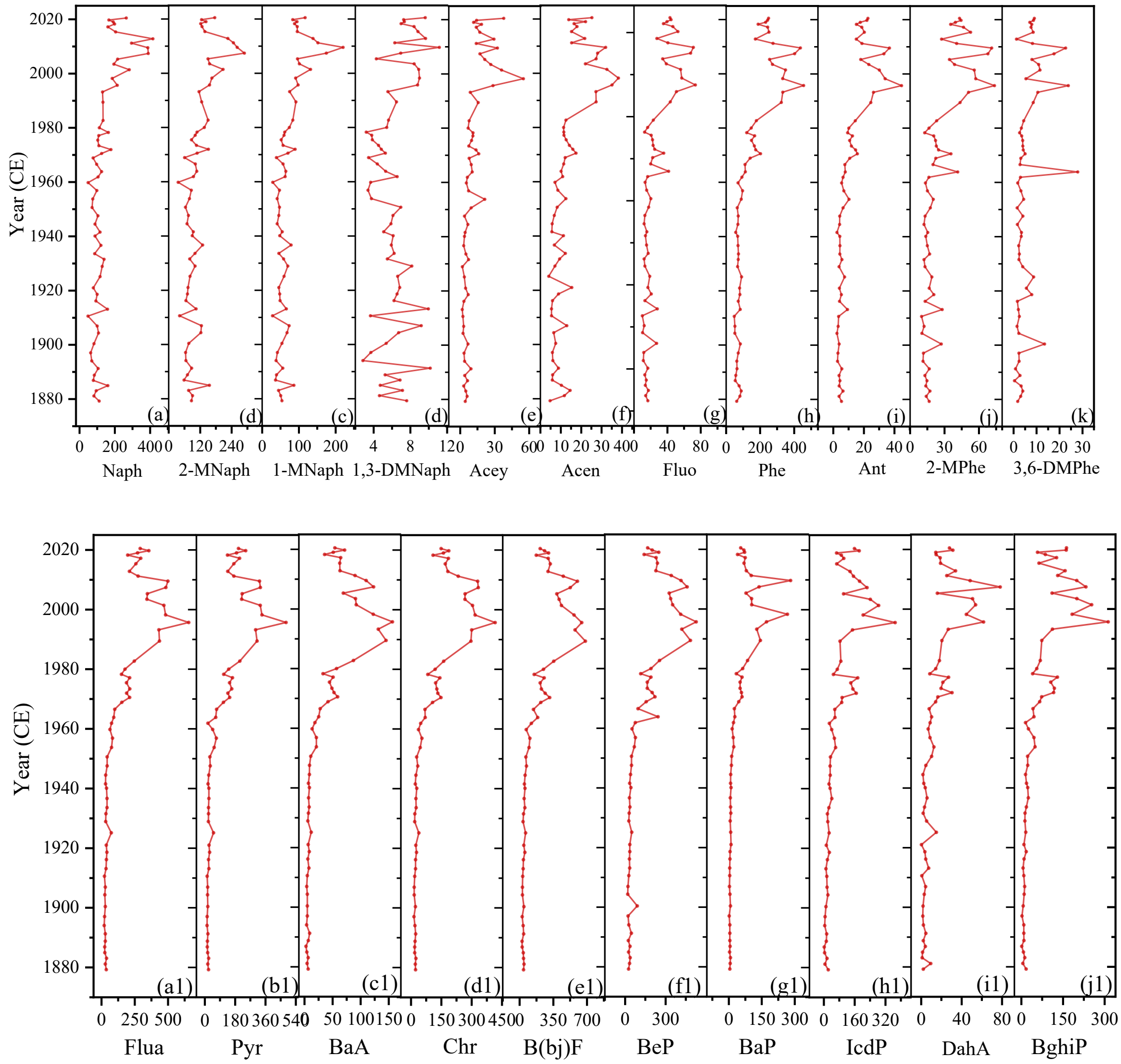

Fig. S4 Vertical distribution of PAH concentration (ng g^{-1}) in the SHLW sediment core. The abbreviations are explained in Table S2.

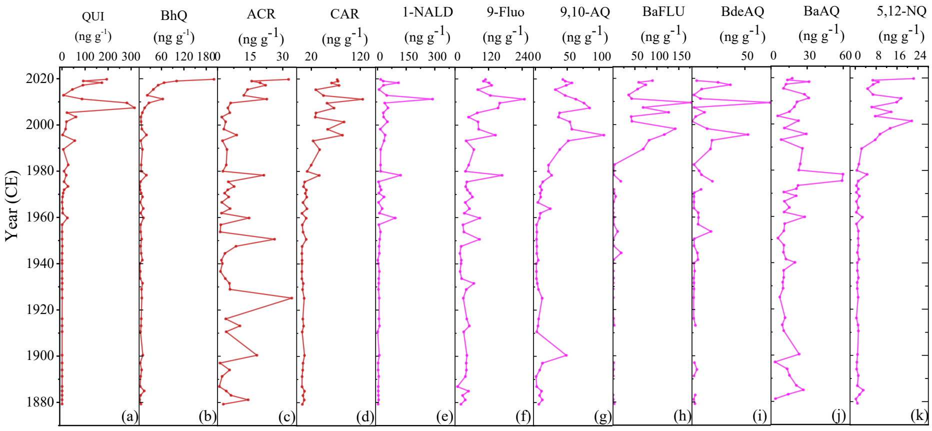

Fig. S5 Vertical distribution of the concentrations and fluxes of AZAs and OPAHs in the SHLW sediment core. The abbreviations are explained in Table S2.

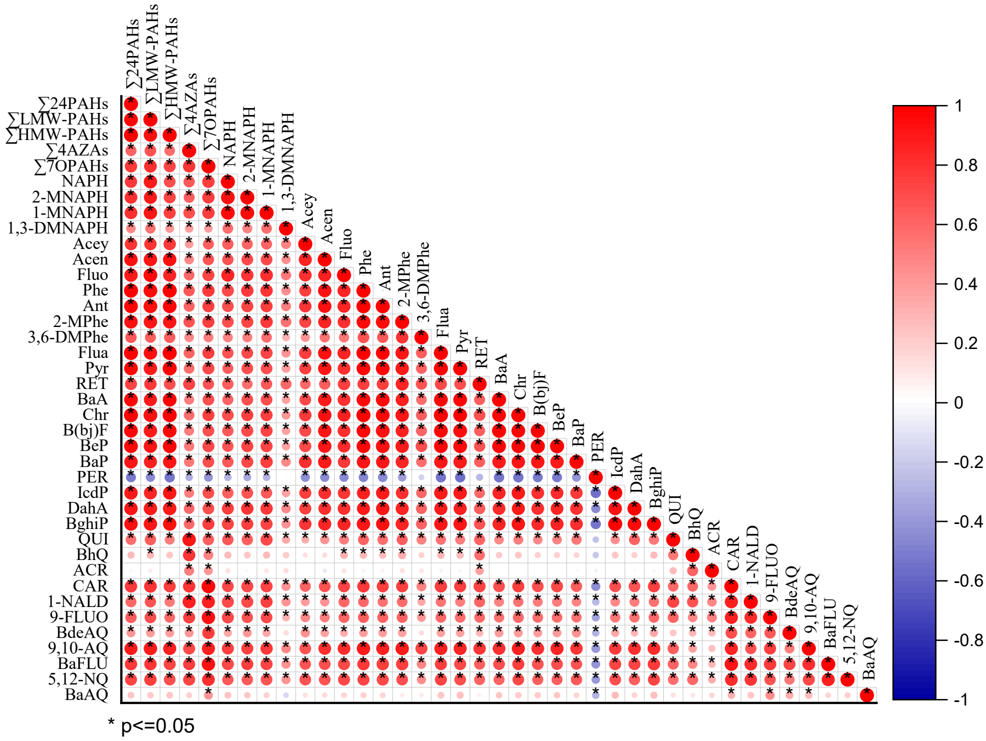

Fig. S6 Spearman correlation of PAHs, OPAHs, and AZAs in the Sihailongwan Maar Lake (SHLW). Color represents the correlation coefficient.

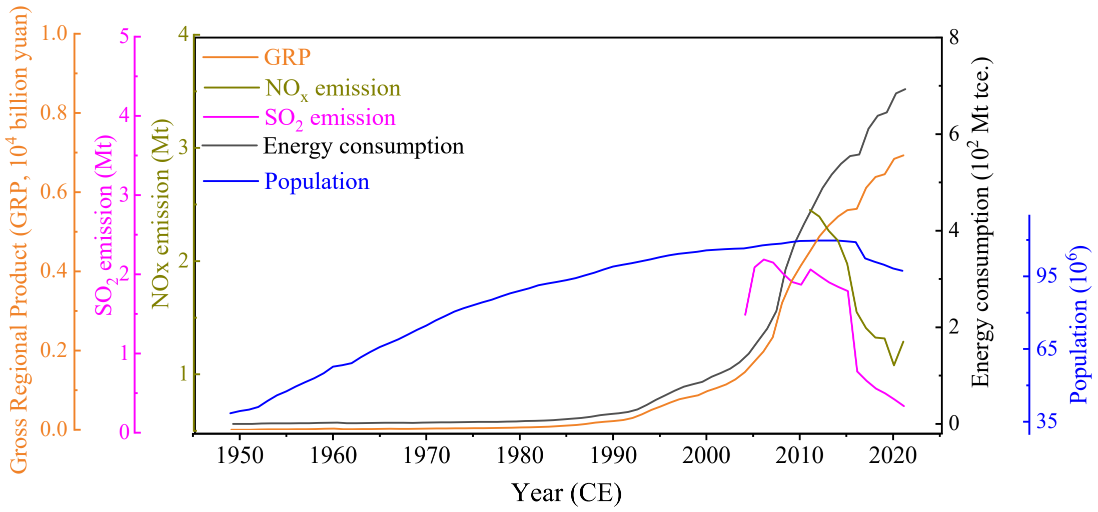

Fig. S7 Temporal curves of the SO_2 and NO_X emissions, energy consumption, Gross Regional Product (GRP), and population in northeast China. Data were obtained from National Data from National Bureau of Statistics (https://data.stats.gov.cn/index.htm).

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