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Supplementary Information

Contrasting temporal trends and relationships of total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in rural low-altitude and remote high-altitude lakes

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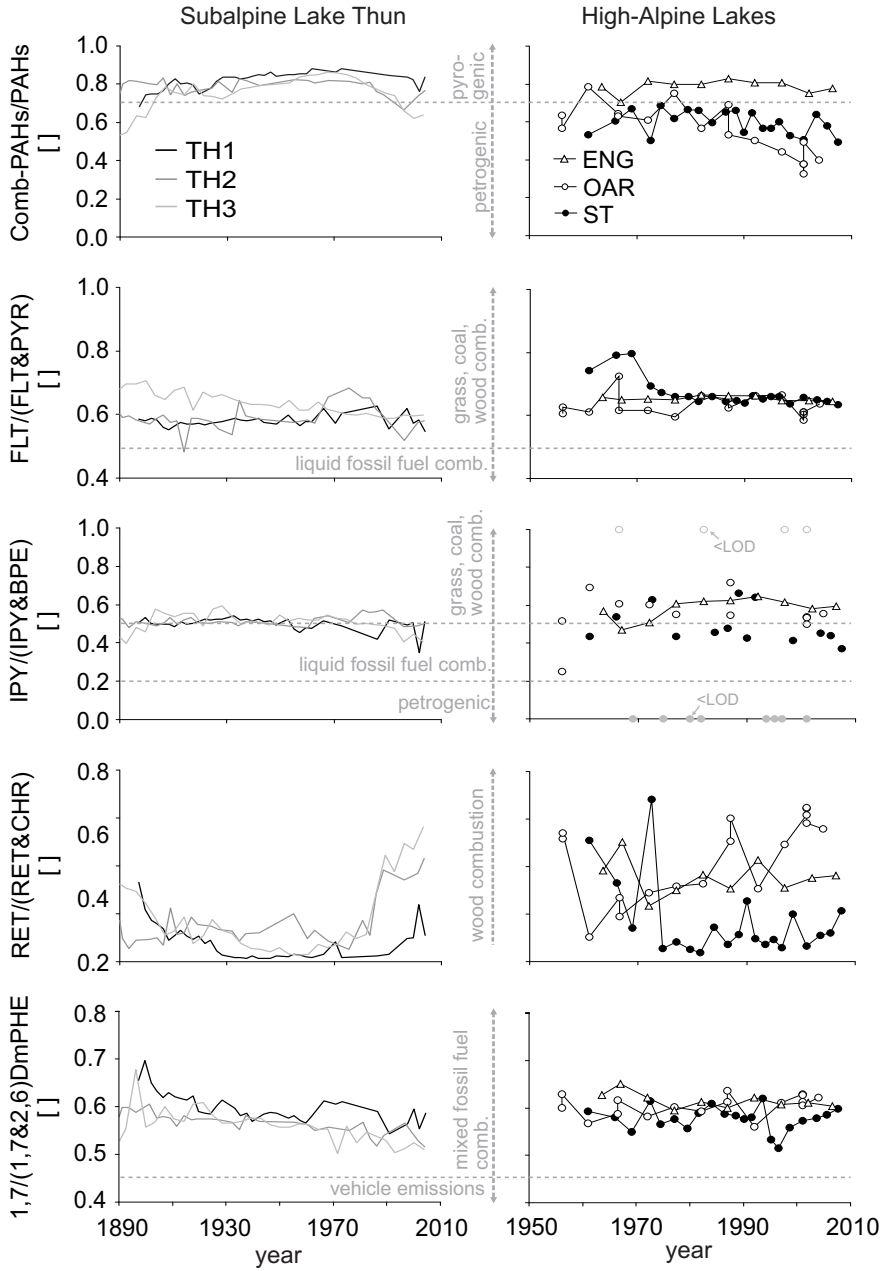

Figure S1. Source-characteristic PAH ratios and molecular markers in three dated sediment cores (TH1, TH2, and TH3) from Lake Thun and sediment cores from mountain lakes (Lake Engstlen ENG, Lake Oberaar OAR, Lake Stein ST).

Rationale

FLT, PYR, BaA, CHR, BbF, BkF, BaP, IPY, and BPE (denoted Comb-PAHs) were stated to be markers for pyrogenic origin[1]. The ratio Comb-PAHs / PAHs, where PAHs denotes the 16 EPA PAHs, higher than 0.7 indicates combustion dominated source, whereas a lower ratio points towards petrogenic origin[1].

Other frequently applied ratios are FLT / (FLT&PYR) and IPY / (IPY&BPE). FLT / (FLT&PYR) and IPY / (IPY&BPE) are possible markers for the distinction between grass, coal, or wood combustion, and fossil fuel combustion[2-4].

As the formation of PAHs by combustion of wood may be identified by RET[5], the ratio RET / (RET&CHR) was calculated.

1,7 / (1,7&2,6)DmPHE may reveal vehicle emissions (ratio <0.45) and mixed combustion of fossil fuel (0.45-0.7)[2].

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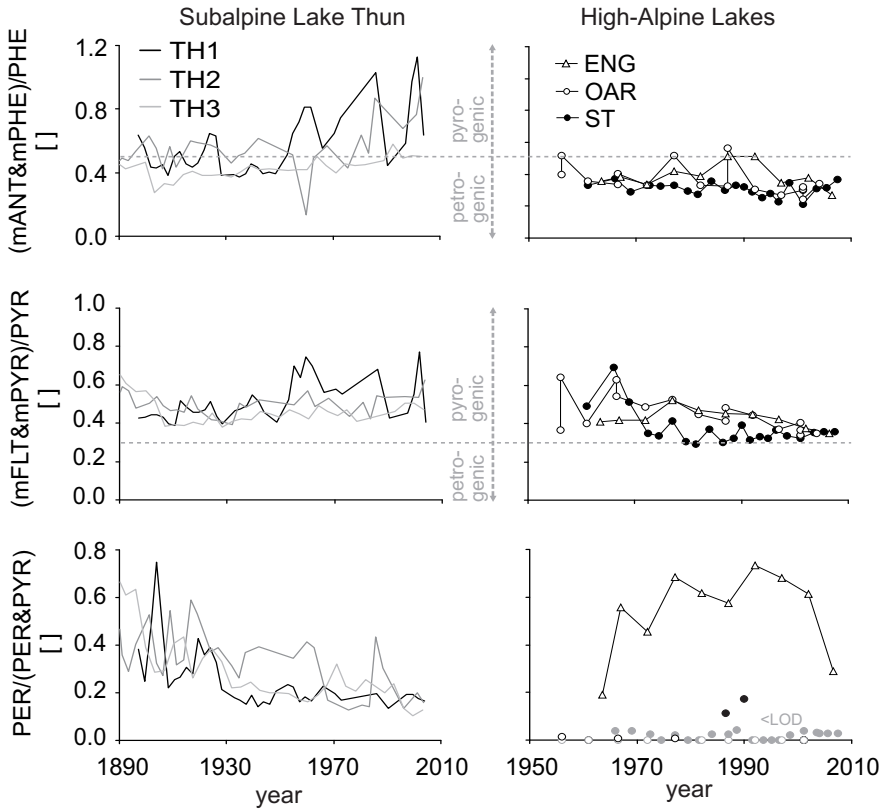

Figure S2. Source-characteristic PAH ratios and molecular markers in three dated sediment cores (TH1, TH2, and TH3) from Lake Thun and sediment cores from mountain lakes (Lake Engstlen ENG, Lake Oberaar OAR, Lake Stein ST).

Rationale

(mANT&mPHE)/PHE and (mFLT&mPYR)/PYR are ratios indicating pyrogenic sources of PAHs, when they are above 0.5 and 0.3, respectively[1]. (Brändli et al., 2008).

PER can be formed *in situ* from non-specific precursor materials by biotic and abiotic diagenetic processes under anoxic conditions in sediment[2-3].

References

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Table S1. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core TH1 from Lake Thun.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2004	3184	21.7	1.3	43	4	7	13	110	18	241
2001	3492	20.3	1.1	19	1	4	7	70	9	78
2000	2570	22.5	1.5	23	8	5	11	108	18	157
1997	2282	24.1	1.2	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1993	2232	26.4	1.4	20	3	5	10	82	7	175
1991	2215	24.6	1.4	20	3	9	18	152	13	283
1987	2409	26.1	2.3	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1978	1936	32.4	1.7	22	3	6	15	151	42	331
1973	1887	28.0	1.7	31	7	10	21	191	44	549
1971	2014	29.5	1.5	30	5	11	21	173	31	360
1966	1650	23.5	1.5	37	12	18	32	285	62	764
1962	1895	25.2	2.2	35	8	11	28	233	66	625
1960	2009	22.6	2.3	35	7	11	29	236	56	462
1957	2860	20.3	2.8	41	12	19	34	399	69	672
1955	3012	16.9	2.0	29	6	12	26	214	39	383
1953	2831	22.9	2.0	22	5	12	21	182	25	360
1948	2315	19.7	2.0	59	51	13	50	539	263	1124
1946	2884	20.1	1.5	66	32	43	69	846	112	1775
1944	2684	28.7	1.9	64	35	25	86	950	131	1773
1939	2730	n.a.	2.2	78	57	28	106	1266	150	2475

n.a. = not available

Table S1. continued.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
1939	3744	20.5	3.1	63	33	25	81	833	111	1503
1939	2835	20.0	2.4	64	45	27	90	1047	107	1718
1937	2755	17.2	1.4	44	35	15	54	593	62	974
1935	3168	16.6	1.2	44	35	13	42	471	50	828
1930	3688	22.3	0.9	36	13	10	28	311	49	536
1930	3496	14.9	1.0	35	10	11	24	238	23	354
1928	2806	19.0	1.0	40	5	12	23	188	24	240
1926	3257	16.9	1.1	41	5	12	26	211	54	250
1923	3023	20.7	1.2	38	3	10	25	182	27	197
1921	3304	15.0	1.9	55	6	10	21	184	30	270
1919	3228	16.2	1.4	44	6	11	20	174	27	262
1917	3059	13.7	1.3	45	6	11	23	177	29	266
1914	2987	14.1	1.2	46	8	13	26	234	44	401
1912	3340	12.9	1.3	43	6	16	30	283	51	389
1910	3327	12.5	1.4	31	3	8	16	124	20	157
1908	3552	12.6	1.1	24	2	5	11	76	10	85
1905	3496	11.3	1.0	35	2	8	17	140	17	148
1903	3654	11.2	0.9	42	4	9	21	155	20	172
1901	3588	11.4	0.9	36	4	5	19	116	15	103
1899	4168	12.4	0.8	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

n.a. = not available

Table S2. Polycyclic aromatic hydrocarbons in dated sediment core TH1 from Lake Thun.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2004	200	102	109	87	46	109	85	14	81	1270
2001	55	35	62	42	16	28	26	11	47	511
2000	117	74	100	90	47	101	85	12	83	1037
1997	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1993	108	54	82	65	34	66	57	9	59	836
1991	227	109	154	119	63	119	96	12	88	1483
1987	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1978	197	159	242	218	84	181	139	53	191	2036
1973	359	258	358	323	149	279	232	64	242	3117
1971	268	174	235	247	120	207	219	52	219	2373
1966	470	325	415	403	215	397	342	82	320	4176
1962	457	349	467	473	215	362	345	108	381	4162
1960	340	249	364	316	145	260	247	75	266	3098
1957	516	345	508	522	204	378	382	100	458	4657
1955	278	182	295	271	125	221	241	71	271	2664
1953	243	146	208	204	96	171	190	45	172	2102
1948	852	491	658	607	302	613	546	126	564	6858
1946	1321	660	1006	838	451	785	654	128	619	9403
1944	1307	633	894	709	400	741	563	106	535	8954
1939	1733	890	1262	1013	556	987	769	147	702	12219

n.a. = not available

Table S2. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
1939	1071	598	822	646	374	681	512	108	484	7945
1939	1236	656	901	662	395	717	528	104	482	8778
1937	694	354	510	409	227	411	328	67	301	5078
1935	578	320	428	362	214	382	338	64	284	4455
1930	394	217	269	236	133	248	223	35	203	2942
1930	261	140	187	149	83	155	135	16	123	1945
1928	180	96	134	109	52	95	103	16	95	1411
1926	190	103	144	117	56	108	110	16	100	1542
1923	147	79	108	77	39	78	77	11	74	1171
1921	204	117	150	125	60	124	117	16	115	1602
1919	197	106	141	120	59	120	116	18	118	1541
1917	196	109	139	121	60	111	114	16	109	1531
1914	310	173	222	193	91	189	163	28	174	2314
1912	315	172	208	171	82	176	151	23	153	2269
1910	124	63	76	64	30	67	58	10	58	907
1908	61	33	48	40	19	40	35	5	35	528
1905	103	56	76	69	32	61	70	10	69	913
1903	125	68	86	65	30	67	76	7	66	1012
1901	73	34	49	36	17	33	41	3	40	626
1899	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

n.a. = not available

Table S3. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core TH2 from Lake Thun.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2004	1884	13.2	1.0	n.a.	2	8	10	58	3	55
2002	1572	12.6	1.1	30	2	4	11	65	5	72
2000	1904	14.1	1.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1996	1311	13.1	1.1	84	13	6	27	69	15	86
1993	1766	13.4	1.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1989	1575	n.a.	0.8	29	2	5	11	75	5	83
1987	1585	13.7	1.1	32	2	8	13	91	5	114
1983	1183	15.8	1.1	29	3	7	14	101	10	194
1979	1260	18.2	1.4	33	3	9	16	118	9	204
1972	881	15.7	1.2	30	3	8	15	109	11	207
1961	1013	16.0	1.1	29	4	6	14	93	10	161
1954	1308	17.6	0.6	47	6	14	25	261	38	357
1951	1817	15.1	0.7	34	5	11	23	210	33	333
1946	2032	16.5	0.7	21	5	4	11	88	14	155
1943	2565	10.1	1.0	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1939	1906	10.5	1.2	21	4	4	11	71	9	93
1935	1475	10.5	0.9	25	6	5	16	125	15	183
1931	1276	12.7	1.2	32	6	7	19	143	17	260
1931	1438	11.3	0.9	38	88	3	26	154	177	275

n.a. = not available

Table S3. continued.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
1926	1703	10.4	0.7	35	8	4	11	79	27	136
1923	1955	7.8	1.2	28	4	4	9	67	11	96
1919	2405	8.1	1.2	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1915	1661	8.8	0.8	21	3	3	8	66	5	78
1912	2305	8.3	0.8	19	3	3	8	54	4	63
1908	2119	9.7	0.8	93	65	5	25	208	388	351
1908	2266	8.7	0.6	35	9	6	18	148	19	220
1906	2659	10.1	0.5	29	7	3	13	98	10	106
1903	2169	10.7	0.7	32	17	5	19	209	18	344
1899	2160	8.9	0.4	28	10	5	17	156	14	207
1895	2697	9.7	0.5	22	8	5	15	137	16	186
1888	2343	8.7	0.4	27	10	5	15	135	15	198
1884	1935	9.9	0.6	31	19	4	20	184	24	272
1881	2556	7.9	0.9	22	12	3	13	111	16	160
1877	2525	8.4	0.7	22	2	2	7	40	3	39
1873	2751	9.4	0.6	31	3	6	8	40	4	43
1870	2382	9.2	0.7	19	3	2	8	47	5	43
1866	2841	9.1	0.6	29	27	3	17	161	29	221
1862	2923	10.6	0.5	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

n.a. = not available

Table S4. Polycyclic aromatic hydrocarbons in dated sediment core TH2 from Lake Thun.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2004	40	18	36	26	12	30	28	3	28	356
2002	53	24	44	33	15	35	32	4	33	466
2000	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1996	80	34	54	36	22	52	30		32	641
1993	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1989	56	24	46	33	18	37	39		29	492
1987	71	36	59	46	24	55	49	4	38	646
1983	104	49	80	70	37	71	69	7	54	898
1979	109	54	87	76	40	77	83	6	62	986
1972	96	51	93	86	53	92	67	7	65	993
1961	85	48	93	89	48	74	76	7	64	902
1954	266	168	193	154	85	219	134	15	119	2101
1951	239	159	180	155	73	199	101	30	110	1894
1946	115	77	80	68	32	93	45	11	48	866
1943	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1939	66	37	62	63	25	61	46	9	45	626
1935	124	70	116	120	52	108	92	16	88	1161
1931	143	86	154	158	66	106	101	21	106	1426
1931	239	126	174	179	82	244	145	24	140	2115

n.a. = not available

Table S4. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
1926	109	55	83	84	37	126	76	12	73	955
1923	71	31	52	49	21	48	40	5	39	575
1919	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1915	54	26	45	41	17	40	29	3	28	469
1912	44	20	37	33	15	34	28	3	24	391
1908	378	164	205	187	95	568	186	29	173	3122
1908	158	78	125	109	47	115	69	9	70	1236
1906	79	31	57	50	23	49	45	5	39	645
1903	232	109	204	199	89	177	118	22	118	1913
1899	144	66	127	119	49	103	70	14	73	1204
1895	140	74	104	90	39	110	62	8	62	1078
1888	142	77	117	114	49	110	79	11	75	1179
1884	184	103	173	173	77	147	107	19	116	1653
1881	111	70	96	87	40	95	57	10	52	956
1877	23	10	19	17	6	15	15		13	233
1873	28	12	19	17	7	20	14		12	264
1870	28	14	18	14	7	15	10	1	14	248
1866	165	98	108	79	43	102	78	10	69	1237
1862	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.

n.a. = not available

Table S5. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core TH3 from Lake Thun.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2005	1954	11.7	n.a.	27.53	1.20	5.06	8.87	35.73	1.88	36.51
2000	1917	n.a.	n.a.	43.60	1.42	7.84	10.67	41.90	2.28	46.86
1996	2236	11.1	n.a.	31.43	1.32	5.53	9.57	41.47	2.16	50.39
1993	2304	10.8	n.a.	28.29	1.84	3.08	8.16	43.03	3.97	64.96
1989	1921	11.9	n.a.	32.24	1.82	8.85	11.71	60.42	4.23	95.21
1985	2123	12.3	0.8	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1982	2006	12.2	0.8	22.35	1.90	5.73	11.03	51.25	3.17	86.22
1978	2028	12.2	0.8	21.69	3.01	3.97	10.13	59.95	6.51	116.40
1975	2096	12.0	0.8	24.20	4.64	4.39	10.92	70.53	8.54	146.53
1971	2417	11.6	0.8	23.30	4.38	4.67	12.32	70.52	11.27	157.57
1968	2252	11.4	0.9	20.75	4.08	4.91	11.42	78.01	8.75	168.52
1964	2069	10.6	0.7	27.49	8.69	4.52	16.03	119.56	15.73	286.75
1960	2378	8.5	0.6	30.23	9.37	6.28	18.26	150.61	17.01	306.66
1957	2296	7.2	0.7	26.22	10.91	4.44	16.44	141.05	16.51	261.16
1953	2602	5.6	0.5	24.29	10.94	3.43	16.38	125.26	13.21	211.80
1950	2595	7.3	0.6	27.11	14.37	2.63	16.84	132.56	13.39	215.72
1946	2372	7.1	0.7	29.41	12.25	5.24	16.58	110.90	12.01	186.11
1943	2622	6.4	0.7	27.87	8.04	4.29	15.37	106.73	10.51	169.87
1939	2106	6.3	0.8	33.14	9.03	3.48	14.05	89.25	15.69	150.95

n.a. = not available

Table S5. continued.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
1936	3047	6.5	0.5	27.77	3.55	3.76	11.43	77.92	8.89	122.38
1932	2570	6.5	0.5	27.42	2.95	3.43	9.70	64.72	5.41	94.65
1928	2430	6.1	0.6	24.30	2.13	2.93	8.59	51.45	3.45	69.82
1925	2876	6.1	0.6	23.69	3.89	2.54	10.15	55.85	6.17	76.30
1921	2344	5.3	0.7	27.28	2.66	3.12	10.18	63.70	6.31	93.05
1918	2180	6.8	0.7	25.08	3.29	4.33	11.17	85.59	13.53	150.92
1914	3280	6.2	0.6	20.33	1.92	2.48	7.98	56.81	5.14	78.51
1911	2596	6.3	0.6	20.46	1.55	3.16	8.20	60.04	4.90	86.83
1907	2376	6.2	0.8	24.20	1.87	4.16	9.19	80.80	6.78	116.22
1904	2951	5.8	0.7	21.24	2.08	3.65	8.61	59.65	4.52	75.44
1900	2546	6.1	0.6	19.79	1.83	2.48	7.25	43.46	2.45	44.40
1896	2305	6.3	0.8	19.38	1.54	2.93	7.73	42.39	2.39	37.13
1893	2302	5.9	0.8	22.66	1.72	3.26	8.31	40.58	1.93	29.62
1889	3208	6.7	0.7	18.18	1.68	2.46	6.86	33.58	1.95	21.91
1886	2583	5.7	0.7	20.97	2.07	2.68	7.38	33.44	2.05	19.15
1882	3148	6.0	0.6	21.73	2.33	2.74	7.56	35.30	1.89	17.49
1879	3334	4.6	0.4	23.18	2.05	3.01	7.22	31.09	1.51	13.37
1875	3139	4.3	0.6	19.47	1.87	2.73	6.44	28.35	1.04	13.28
1871	3935	4.2	0.7	18.19	1.34	2.57	5.09	24.70	0.62	10.81
1868	3763	4.0	0.5	17.45	1.26	2.25	4.49	21.88	0.41	8.24

Table S5.continued.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
1864	3421	4.2	0.5	21.08	1.40	2.68	5.24	22.68	0.44	8.39
1861	3405	4.3	0.4	19.79	1.64	2.36	4.92	21.28	0.53	9.73
1857	3783	4.3	0.6	19.18	1.44	2.05	4.00	17.57	0.44	8.63
1854	3551	4.7	0.7	18.85	1.46	2.17	4.84	21.50	0.47	9.91
1850	3492	4.7	0.6	21.37	1.44	1.81	4.95	18.24	0.51	7.23
1847	3659	4.3	0.6	19.66	1.22	2.03	4.79	21.35	0.45	10.14
1843	2690	4.7	0.5	20.77	1.29	1.93	5.08	24.85	0.58	13.60
1839	3370	4.7	0.5	18.49	1.27	2.06	5.05	24.95	0.66	14.72
1836	3099	4.9	0.4	34.00	1.23	3.03	4.27	20.86	0.49	11.47
1832	3066	4.2	0.6	19.43	1.10	2.04	4.17	18.63	0.44	9.63
1829	3134	5.8	0.5	16.76	1.09	2.22	5.20	22.51	0.55	12.70

Table S6. Polycyclic aromatic hydrocarbons in dated sediment core TH3 from Lake Thun.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2005	24.47	9.56	16.14	16.99	6.11	10.67	9.91	0.94	14.45	226.03
2000	31.95	11.76	20.58	20.49	7.68	12.99	12.67	1.68	15.86	290.21
1996	33.69	12.07	20.62	19.40	7.49	11.72	10.37	1.04	15.09	273.38
1993	45.31	17.62	28.04	29.13	10.99	22.86	20.78	3.03	22.91	353.99
1989	66.38	25.61	41.21	41.62	16.93	31.95	28.00	2.97	29.24	498.37
1985	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.
1982	56.90	27.73	43.77	49.65	23.40	31.50	39.72	5.04	39.10	498.46
1978	78.16	40.78	62.62	71.36	33.88	47.78	51.48	4.76	52.00	664.46
1975	97.56	51.98	79.53	100.49	48.05	69.65	78.95	6.96	71.79	874.71
1971	101.09	55.26	85.73	115.42	57.27	82.01	88.83	10.66	79.27	959.55
1968	105.41	59.40	94.08	117.46	55.76	73.60	94.73	8.10	78.53	983.50
1964	161.89	86.15	131.64	153.61	68.97	100.33	103.93	15.34	95.34	1395.98
1960	192.88	105.12	157.18	176.67	86.75	127.80	130.07	23.85	119.91	1658.64
1957	164.44	90.09	135.24	142.40	67.19	108.93	104.43	14.92	92.99	1397.38
1953	116.26	58.50	103.54	102.91	48.76	68.62	76.03	11.23	71.41	1062.56
1950	132.04	61.18	100.05	105.33	46.91	81.92	82.25	13.46	68.77	1114.53
1946	109.56	54.26	86.42	92.98	42.18	64.25	58.83	7.88	60.24	949.09
1943	98.81	49.35	75.13	79.84	38.13	56.44	56.05	6.52	54.53	857.47
1939	87.62	44.38	67.99	73.11	32.90	54.30	56.67	6.45	53.49	792.50

n.a. = not available

Table S6. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
1936	69.27	34.14	52.38	55.16	24.86	29.71	37.60	2.57	38.06	599.45
1932	51.19	22.82	39.00	40.40	22.46	20.47	28.31	1.72	24.59	459.23
1928	35.42	15.97	28.94	29.31	15.89	12.49	20.66	0.00	14.14	335.49
1925	40.10	18.69	31.37	33.96	17.78	16.50	29.02	0.00	21.36	387.38
1921	45.89	25.01	39.19	39.16	17.32	18.87	24.27	1.79	25.49	443.28
1918	94.50	52.52	62.58	65.22	33.38	43.25	48.03	3.82	38.26	735.45
1914	36.17	17.62	30.10	32.87	15.24	17.15	30.01	1.51	24.29	378.14
1911	41.52	20.57	32.04	36.50	16.49	18.77	31.40	2.24	27.07	411.74
1907	63.09	36.14	47.37	49.29	53.50	27.82	40.34	2.77	32.25	595.80
1904	39.00	18.94	29.00	30.15	15.76	15.03	25.69	1.40	18.75	368.91
1900	18.48	7.71	16.54	15.62	6.26	4.66	7.41	0.45	8.81	207.60
1896	16.24	6.80	14.83	13.39	17.31	5.47	11.64	0.93	12.51	212.61
1893	12.99	5.39	12.21	10.95	4.95	2.72	6.39	n.a.	9.69	173.38
1889	10.68	4.03	9.52	7.67	3.05	3.83	5.59	n.a.	6.90	137.89
1886	10.56	2.97	7.72	5.57	2.23	2.97	3.90	n.a.	5.76	129.42
1882	10.51	3.08	7.83	5.12	2.86	2.00	n.a.	n.a.	4.34	124.77
1879	8.63	2.50	7.25	5.36	1.40	2.55	n.a.	n.a.	6.35	115.45
1875	7.05	1.94	6.02	4.10	1.33	1.66	n.a.	n.a.	4.31	99.59
1871	5.78	1.73	5.33	3.96	1.12	1.36	n.a.	n.a.	3.76	86.36
1868	5.11	1.16	4.08	2.69	0.81	1.10	n.a.	n.a.	2.84	73.75

n.a. = not available

Table S6. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
1864	5.07	1.47	4.27	3.07	1.09	1.28	n.a.	n.a.	2.94	81.10
1861	5.11	1.31	4.07	2.59	1.00	0.84	n.a.	n.a.	2.40	77.57
1857	4.64	1.37	3.95	2.90	0.84	1.34	n.a.	n.a.	2.59	70.95
1854	6.23	1.46	5.27	3.81	1.41	0.81	n.a.	n.a.	3.77	81.96
1850	4.45	1.00	3.96	2.38	1.09	0.57	n.a.	n.a.	1.84	70.85
1847	5.34	1.37	5.11	3.23	1.12	0.69	n.a.	n.a.	2.49	78.99
1843	6.74	1.90	5.97	4.69	1.60	1.33	n.a.	n.a.	4.15	94.45
1839	6.94	2.11	6.15	4.72	2.36	1.08	n.a.	n.a.	4.21	94.78
1836	5.90	1.41	4.68	3.07	1.25	0.57	n.a.	n.a.	2.24	94.48
1832	5.26	1.27	4.47	3.20	1.28	0.67	n.a.	n.a.	3.39	74.98
1829	6.51	1.77	5.41	3.85	1.28	0.85	n.a.	n.a.	3.11	83.82

n.a. = not available

Table S7. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core from Lake Engstlen.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2007	1239	15.2	1.5	10.20	0.48	1.44	2.85	22.08	1.06	25.52
2002	1042	13.2	1.8	9.09	0.36	1.16	2.57	19.53	0.80	19.29
1997	1093	24.5	2.6	8.73	0.56	1.06	2.56	19.96	1.00	27.13
1992	976	29.5	2.2	8.09	0.42	1.16	2.59	20.04	1.09	27.65
1987	1338	24.6	2.7	6.10	0.55	0.75	2.03	16.50	0.97	27.36
1982	1540	16.8	3.0	8.09	0.55	1.20	2.60	21.32	1.08	30.38
1977	1988	13.1	3.3	7.58	0.38	1.10	2.44	20.78	1.42	27.99
1972	2182	12.2	3.1	11.24	0.59	1.41	2.52	18.47	1.07	31.87
1967	1567	10.0	3.2	26.70	0.66	2.01	3.05	17.38	1.00	23.33
1964	2579	21.0	2.9	17.79	0.95	2.16	3.47	24.44	1.38	37.69

Table S7. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2007	14.11	6.85	21.65	28.84	9.51	5.01	15.51	1.77	10.58	177.45
2002	10.54	5.14	14.60	21.30	7.14	4.82	12.86	1.34	9.28	139.82
1997	14.82	7.14	19.38	29.57	10.05	8.01	20.68	2.35	12.96	185.98
1992	14.05	6.97	17.59	29.49	10.33	8.54	25.08	2.91	13.83	189.82
1987	13.85	7.22	19.27	29.27	10.19	6.72	17.15	2.04	10.20	170.19
1982	15.25	7.50	18.13	27.13	9.61	7.95	21.13	2.80	12.88	187.60
1977	15.02	8.73	18.00	25.96	9.60	8.01	19.51	2.98	12.53	182.05
1972	17.02	7.89	23.46	33.64	10.84	6.60	17.18	1.52	16.52	201.86
1967	12.54	5.90	15.79	24.84	8.27	4.95	12.72	1.18	14.31	174.63
1964	19.52	9.18	24.51	40.92	13.37	5.44	24.01	2.08	18.06	244.96

Table S8. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core from Lake Oberaar.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2004	30020	1.5	0.3	9.80	0.15	0.96	0.68	4.71	0.08	2.94
2001	28380	1.3	0.2	9.77	0.13	1.02	0.95	6.09	0.11	2.46
2001	28381	n.a.	n.a.	8.65	0.10	1.07	1.15	8.03	0.18	2.83
2001	28382	n.a.	n.a.	12.27	0.11	0.86	0.77	4.48	0.08	2.32
2001	28383	n.a.	n.a.	7.15	0.10	0.73	0.90	6.76	0.11	3.08
1997	34507	1.5	0.3	5.21	0.09	0.53	0.52	3.38	0.05	2.09
1992	11841	1.4	0.3	5.46	0.16	0.56	0.66	4.90	0.09	3.08
1987	12071	1.1	0.3	5.79	0.25	0.65	0.87	8.99	0.22	7.41
1987	12072	n.a.	n.a.	8.34	0.15	0.32	0.41	2.40	0.05	2.93
1982	11291	1.3	0.3	5.78	0.23	0.54	0.48	3.71	0.10	3.70
1977	11344	1.1	0.3	8.20	0.30	0.37	0.45	3.92	0.11	9.11
1972	16460	1.1	0.3	5.76	0.21	0.57	0.65	4.87	0.16	4.68
1967	16460	1.1	0.3	5.45	0.25	0.56	0.55	4.19	0.33	4.64
1967	16461	n.a.	n.a.	7.81	0.18	0.31	0.42	2.46	0.08	3.71
1961	19797	0.9	0.3	4.90	0.28	0.62	0.75	5.75	0.00	7.30
1956	14825	0.8	0.3	7.96	0.28	0.70	0.65	4.74	0.26	5.56

n.a. = not available

Table S8. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2004	1.68	0.73	1.63	2.29	0.68	0.05	0.48	0.00	0.38	27.23
2001	1.74	0.70	1.54	2.24	0.68	0.13	0.77	0.00	0.65	28.99
2001	1.85	0.64	1.45	2.07	0.60	0.24	1.04	0.11	0.90	30.89
2001	1.48	0.53	1.38	2.04	0.66	0.00	0.56	0.00	0.00	27.54
2001	1.96	0.96	2.09	2.97	0.95	2.12	0.62	0.00	0.62	31.12
1997	1.06	0.47	1.36	1.93	0.56	0.00	0.31	0.00	0.00	17.57
1992	1.57	0.63	2.43	3.21	0.96	0.00	0.00	0.00	0.00	23.72
1987	3.94	1.69	5.27	7.59	2.42	5.34	3.26	0.34	1.27	55.30
1987	1.76	0.64	2.08	2.29	0.77	1.90	0.48	0.00	0.40	24.89
1982	1.89	0.87	2.51	3.75	1.17	0.00	0.34	0.00	0.00	25.07
1977	6.16	3.13	5.44	5.68	2.37	5.33	1.61	0.00	1.30	53.46
1972	2.91	1.38	3.34	4.44	1.52	0.00	0.43	0.00	0.29	31.21
1967	1.77	1.08	3.57	5.24	1.78	0.00	2.26	0.00	0.00	31.67
1967	2.30	1.01	3.02	3.38	1.26	3.25	0.83	0.00	0.54	30.57
1961	4.64	2.28	5.90	8.29	2.86	8.80	3.22	0.00	1.43	57.00
1956	3.62	2.17	3.42	4.65	1.87	0.00	1.08	0.00	3.23	40.19

Table S9. Total organic carbon, black carbon, and polycyclic aromatic hydrocarbons in dated sediment core from Lake Stein.

year	sediment flux [g/m^2/y]	total organic carbon [mg/g]	black carbon [mg/g]	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]
2008	70242	3.3	2.1	8.30	0.13	1.18	1.23	7.70	0.19	4.81
2006	64672	4.1	2.6	8.97	0.16	1.11	1.33	8.73	0.26	6.82
2004	71391	4.4	3.0	7.73	0.17	1.01	1.13	7.54	0.00	7.18
2001	31495	3.7	2.6	8.85	0.12	0.77	0.95	4.60	0.09	4.71
1999	75156	2.4	2.0	6.28	0.11	0.80	1.01	6.73	0.15	4.61
1997	94359	2.8	2.4	7.91	0.12	0.75	0.88	4.61	0.10	5.47
1995	82816	2.7	1.8	7.32	0.10	0.74	0.74	4.01	0.08	4.61
1994	54045	2.2	1.8	6.91	0.11	0.68	0.75	3.98	0.05	4.64
1992	107962	2.1	2.0	6.92	0.16	0.65	0.78	5.51	0.15	7.04
1990	74097	1.7	1.2	5.92	0.10	0.88	1.11	7.96	0.18	4.77
1989	97699	2.0	1.9	5.24	0.10	0.50	0.64	4.13	0.09	4.67
1987	106341	2.1	2.1	5.68	0.10	0.52	0.74	5.38	0.15	5.38
1984	55447	2.7	2.7	6.31	0.15	1.00	1.21	8.71	0.22	7.01
1982	88354	2.5	2.5	6.28	0.15	0.63	0.76	4.94	0.16	6.33
1980	85491	2.7	2.2	6.66	0.16	0.60	0.89	6.01	0.17	8.04
1977	55545	2.9	2.6	6.60	0.14	0.78	0.92	5.74	0.17	6.77
1975	71233	2.6	2.6	6.59	0.17	0.55	0.94	6.68	0.19	9.15
1973	61139	2.2	1.9	5.65	0.20	0.64	0.96	6.42	0.13	4.84
1969	55876	2.3	1.9	5.63	0.24	0.40	0.85	5.74	0.07	8.53
1966	33067	2.6	2.6	7.50	0.83	0.49	1.30	10.26	0.20	13.15
1961	52218	2.2	1.4	6.72	0.45	0.91	1.33	10.08	0.14	8.61

Table S9. continued.

year	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
2008	2.78	0.77	2.73	3.54	0.88	0.51	0.78	0.00	1.33	36.87
2006	3.74	1.16	4.33	6.01	1.46	0.72	1.71	0.00	2.18	48.68
2004	3.87	1.25	4.94	6.82	1.66	0.99	2.12	0.00	2.56	48.98
2001	2.45	0.74	3.39	2.90	1.33	2.17	0.00	0.00	0.46	31.07
1999	2.62	0.74	2.73	3.18	0.80	0.38	0.76	0.00	1.09	31.98
1997	2.81	1.18	4.94	4.30	1.92	0.25	0.00	0.00	0.61	35.87
1995	2.38	0.72	3.27	2.56	0.99	2.04	0.00	0.00	0.35	29.92
1994	2.46	0.82	3.62	3.06	1.19	0.16	0.00	0.00	0.31	28.74
1992	3.57	1.27	5.97	5.00	1.93	0.18	0.86	0.00	0.48	40.47
1990	2.68	0.83	3.07	3.93	1.00	0.51	1.17	0.17	1.56	35.84
1989	2.56	1.07	4.16	4.49	1.54	0.33	1.27	0.00	0.64	31.42
1987	2.96	1.08	4.63	5.18	1.75	0.38	1.11	0.00	1.21	36.27
1984	3.63	1.02	4.07	5.33	1.26	0.61	1.63	0.16	1.92	44.24
1982	3.47	1.32	5.60	5.49	1.97	0.15	0.00	0.00	0.79	38.03
1980	4.13	1.44	6.30	5.82	1.96	0.19	0.00	0.00	0.67	43.06
1977	3.50	1.03	4.21	4.53	1.20	0.38	0.85	0.17	1.11	38.10
1975	4.43	1.55	7.63	6.65	2.55	0.18	0.00	0.00	0.92	48.19
1973	2.13	0.57	2.70	2.79	0.66	0.08	0.26	0.00	0.15	28.18
1969	2.18	0.75	7.40	5.56	1.21	0.26	0.00	0.00	0.29	39.10
1966	3.49	0.60	7.13	6.14	0.88	0.08	0.14	0.00	0.12	52.33
1961	3.00	0.77	4.76	3.81	0.76	0.03	0.24	0.09	0.31	42.02

Table S10. Polycyclic aromatic hydrocarbons in blank samples based on a typical sample amount.

sample	naphthalene [µg/kg]	acenaphthylene [µg/kg]	acenaphthene [µg/kg]	fluorene [µg/kg]	phenanthrene [µg/kg]	anthracene [µg/kg]	fluoranthene [µg/kg]	pyrene [µg/kg]	benzo[*a*] anthracene [µg/kg]	chrysene [µg/kg]
blank TH1-1	2.18	<0.01	0.55	0.27	1.16	<0.01	1.07	2.25	<0.01	<0.01
blank TH1-2	2.33	<0.01	0.78	0.46	1.66	<0.01	0.62	0.45	<0.01	<0.01
blank TH1-3	2.18	0.11	0.59	0.30	1.06	<0.01	0.27	<0.01	<0.01	<0.01
blank TH1-4	6.30	0.26	1.20	0.96	2.59	<0.01	0.65	0.56	<0.01	<0.01
blank TH2-1	6.30	0.26	1.20	0.96	2.59	<0.01	0.65	0.56	<0.01	<0.01
blank TH2-2	3.45	0.13	0.41	0.32	1.49	<0.01	0.52	<0.01	<0.01	<0.01
blank TH2-3	2.84	0.09	0.36	0.27	1.48	<0.01	0.65	0.75	<0.01	<0.01
blank TH2-4	2.29	0.06	0.38	0.23	1.06	<0.01	0.55	0.73	0.39	<0.01
blank TH3-1	14.07	0.17	2.31	1.17	2.83	<0.01	0.65	<0.01	0.55	0.20
blank TH3-2	22.43	<0.01	2.42	1.70	3.45	<0.01	1.16	<0.01	<0.01	<0.01
blank TH3-3	7.06	<0.01	0.64	0.38	1.45	<0.01	0.45	<0.01	<0.01	<0.01
blank TH3-4	4.31	<0.01	0.87	0.63	1.82	<0.01	0.43	<0.01	<0.01	<0.01
blank TH3-5	4.05	0.14	1.18	0.70	2.23	<0.01	0.55	<0.01	<0.01	<0.01
blank TH3-6	4.57	0.13	0.76	0.46	2.13	<0.01	1.10	1.46	0.78	<0.01
blank ENG	9.22	0.05	0.30	0.30	0.47	0.01	0.17	0.15	<0.01	<0.01
blank OAR-1	4.59	0.04	0.41	0.60	4.40	0.10	0.70	0.65	<0.01	<0.01
blank OAR-2	4.48	<0.01	0.37	0.27	0.89	<0.01	0.19	0.10	0.12	0.17
blank ST	4.73	0.03	0.53	0.57	3.69	0.08	0.67	0.58	0.16	0.09

Table S11. Polycyclic aromatic hydrocarbons in blank samples based on a typical sample amount.

sample	benzo[*b*] fluoranthene [µg/kg]	benzo[*k*]fluoranthene [µg/kg]	benzo[*a*]pyrene [µg/kg]	indeno[1,2,3,*c*,*d*] pyrene [µg/kg]	dibenzo[*a*,*h*]anthracene [µg/kg]	Benzo[*g*,*h*,*i*]perylene [µg/kg]	sum 16 PAHs [µg/kg]
blank TH1-1	<0.01	0.23	<0.01	<0.01	<0.01	<0.01	7.70
blank TH1-2	0.18	0.59	<0.01	<0.01	<0.01	<0.01	7.07
blank TH1-3	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	4.51
blank TH1-4	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	12.52
blank TH2-1	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	12.52
blank TH2-2	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	6.31
blank TH2-3	<0.01	<0.01	<0.01	2.24	<0.01	1.88	10.58
blank TH2-4	1.21	<0.01	<0.01	<0.01	<0.01	<0.01	6.90
blank TH3-1	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	21.96
blank TH3-2	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	31.16
blank TH3-3	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	9.98
blank TH3-4	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	8.05
blank TH3-5	0.20	0.32	<0.01	<0.01	<0.01	<0.01	9.36
blank TH3-6	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	11.39
blank ENG	<0.01	<0.01	<0.01	<0.01	<0.01	0.11	10.78
blank OAR-1	<0.01	<0.01	<0.01	<0.01	<0.01	0.21	11.69
blank OAR-2	0.15	0.16	0.16	<0.01	<0.01	0.17	7.22
blank ST	0.16	0.03	0.12	<0.01	<0.01	<0.01	11.43