1	Supporting Online Materials for
2 3	Elemental carbon and polycyclic aromatic compounds in a 150-yr
	sediment core from Lake Qinghai, Tibetan Plateau, China:
4	
5	Influence of regional and local sources and transport paths
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7	Y.M. Han, C. Wei, B.A.M. Bandowe, W. Wilcke, J.J. Cao,
8	B.Q. Xu, S.P. Gao, X.X. Tie, G.H. Li, Z.D. Jin, Z.S. An
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13	This file is 26 pages in total, including materials listed below:
14	S1 Materials and methods;
15	Figures S1 to S10;
16	Tables S1 to S2.
17	
18	S1 Materials and methods
19	S1.1. Study area and sample collection
20	Lake Qinghai (36°32'-37°15' N, 99°36'-100°47' E), the largest lake in China,
21	has a water surface altitude of 3,194 m above sea level, a surface water area of
22	approximately 4,260 km ² , and a catchment of more than 29,660 km ² 1 . Under the
23	influence of ASM and westerly climate, the precipitation occurs mainly in summer,
24	contributing most of the annual average values (337 mm a ⁻¹), while in winter the area

is covered by ice. The lake is divided into two sub-basins by a NNW-trending piggy-back horst ¹, and five major rivers surrounding the lake feed the detrital input into the lake, especially the northern sub-basin (Fig. S1B). The population in the catchment is concentrated in the cities of Gangcha, Tianjun, and Haiyan, which are also located in the northern part. Human activities are therefore expected to have greater influence in the northern part of the lake.

In September 2011, a 33-cm long sediment core QH11-1 was collected from the 31 southern part of Lake Qinghai (in the same place as core QH03-02 from Jin et al.¹) 32 with a water depth of 28 m using a gravity corer (Uwitec, Austria; Fig. S1B). A 33 distinct water-sediment interface and discernible laminations were observed, 34 indicating a lack of bioturbation and post-depositional disturbance. There was a 35 remarkable litho-stratification between the upper and lower part of the core. 36 Sediments of the upper 6 cm were grey clay, while blackish clay sediments occurred 37 below, which is consistent with previous reports by Xu et al. 2 and Jin et al. 1 . The 38 core was sectioned continuously at 0.5 cm intervals, and then the sediments were 39 freeze-dried, agate mortar ground, and frozen at -20°C until further analysis. The 40 water content and dry density data were estimated from the mass of samples before 41 and after freeze-drying. 42

Another sediment core QH03-14, located in the northern sub-basin (Fig. S1), and collected in July 2003 and of which the chronology and metal deposition has been reported before ¹, was used to compare EC, char, and soot deposition between the southern and northern sub-basins.

S1.2. Chronology reconstruction

The activities of ¹³⁷Cs, ²¹⁰Pb, and ²²⁶Ra in the depth increments of QH11-1 48 were analyzed for sediment dating by direct gamma counting of 3-6 g of dried 49 sediments using a multi-channel γ -ray spectrometer (PerkinElmer, GWL-120-15)^{1,3}. 50 The measured ²¹⁰Pb and ¹³⁷Cs activities against depth are presented in Fig. S2. Excess 51 ^{210}Pb ($^{210}\text{Pb}_{ex})$ activities were calculated from $^{210}\text{Pb}_{tot}$ and ^{226}Ra activities with the 52 level-by-level method. The activity of excess 210 Pb_{ex} in the uppermost 1-cm section of 53 core QH11-1 in our study (about 478.25 Bq kg⁻¹) is similar to that reported by Jin et al 54 ¹ (474.3 Bq kg⁻¹), who collected the sediment core from the same site as we did. 55 However, our activitives are higher than those reported by others in the southern basin 56 4,5 , which may be associated with sediment focusing ⁶ as our cores (collected in 2003) 57 58 and in 2011, respectively) were obtained in deeper water than most previous cores. However, the ²¹⁰Pb_{ex} activity in the surface sediment of core QH11-1 is much lower 59 than that in the northern sub-basin ^{1, 2} because of more detrital inputs in the north 60 sub-basin of the lake. ¹³⁷Cs exhibited a peak, which is similar with other cores 61 collected in the Lake Qinghai^{1, 2, 5} and this suggests that substantial mixing of the 62 sediments did not occur. Similarly, higher ¹³⁷Cs concentrations were found in the 63 cores of northern sub-basin^{1, 2, 5}. 64

Fig. S2 presents the chronology reconstruction for core QH11-1 using the constant rate of ²¹⁰Pb supply (CRS) model⁷⁻⁹ and the corresponding accumulation rates. The unconstrained CRS model predicts the ¹³⁷Cs peak (58.1 Bq kg⁻¹) at 3.25 cm sediment depth to AD 1977 \pm 3.4, which is not in agreement with the global nuclear

69 bomb testing period of ~1963. However, the 3.75 cm sediment depth corresponds to AD 1966 \pm 4.4, which is very close to the global nuclear fallout (Fig. S2C). 70 Calculating the ¹³⁷Cs fluxes using mass accumulation rates (MARs) as derived from 71 the CRS model^{7, 8} (Fig. S2F) demonstrates that the shape of the ¹³⁷Cs peak is really 72 broad and it is difficult to define when the maximum flux occurred. This points to 73 post-deposition diffusional redistribution of ¹³⁷Cs in the sediment profile which is also 74 supported by the ¹³⁷Cs activity data that appeared to be moved downward the core 75 (Figs S2E). The constrained CRS model⁹ that forces the chronology through the ¹³⁷Cs 76 fix-point (3.75 cm = AD 1963) yields a chronology (Fig. S2D) very close to that 77 reconstructed using the CRS model. Thus, the "free-shape CRS model"⁷⁻⁹ was used in 78 our study for chronology reconstruction. 79

80

S1.3. EC, char, and soot measurement

The IMPROVE (Interagency Monitoring of Protected Visual Environments) 81 method was used to quantify EC, char, and soot concentrations following Han et al.^{10,} 82 ¹¹. Briefly, ~ 100 mg sediment for each sample was pretreated with hydrochloric and 83 hydrofluoric acids to remove carbonate, metal oxides, and silicates. The pretreated 84 residues were then filtered through pre-baked (850°C for 3 hours) quartz-fiber filters 85 (0.4 µm pore size, Whatman) using deionized water and air dried in an oven (35°C for 86 87 8 hours). A DRI Thermal/Optical Carbon Analyzer (Atmoslytic Inc. Calabasas, CA) was used to implement the IMPROVE protocol. It reports four OC fractions (OC1 to 88 OC4 at 120, 250, 450 and 550°C in a pure helium atmosphere), three EC fractions 89 90 (EC1 to EC3 at 550, 700 and 800°C in 2% oxygen/98% helium atmosphere), and one

91 pyrolyzed organic carbon (POC) fraction. The POC produced in the inert atmosphere
92 is monitored by a laser to both the reflectance and transmittance during thermal
93 analysis. The IMPROVE protocol defined EC as the sum of the three EC fractions
94 minus POC. Han et al. ¹² defined char as EC1 minus POC, while soot as the sum of
95 EC2 and EC3.

We are aware of the fact that the methodology for BC measurement is disputed 96 ¹³. The comparison of the different methods traditionally used for geological samples 97 showed that the BC concentrations determined with different methods can vary by a 98 factor of up to 571¹⁴. For aerosols, the most commonly used method is the thermal 99 optical method with different protocols, and the concentrations measured by the 100 different protocols for given samples on average, only varies by a factor of 2¹⁵, which 101 is much narrower than the wide range of measured concentrations caused by the 102 different methods used for geological samples. To ensure quality control and 103 validation of the analytical method, standard reference materials (SRMs) need to be 104 included into analytical protocols, which is mostly not the case. Furthermore, possible 105 106 matrix effects have to be accounted for.

For several decades, the thermal optical method was mainly used to quantify aerosol EC concentrations ¹⁶. Just since 2007, it was extended to measure EC in sediments and soils ^{10, 17}. Based on the finding that char and soot in standard reference materials (SRMs) can be stepwise oxidized in the EC1 and EC2+3 steps, respectively, it was proposed that the IMPROVE method can be used to differentiate between char and soot ¹². The adaptation, validation and application of this method have been tested in many previous studies. Positive and negative EC SRMs and environmental EC matrices ¹³ were tested in many previous studies ^{10, 11, 18}. In addition, Han et al. ¹⁹ demonstrated that the soot concentrations determined with the IMPROVE method (i.e. the sum of the EC2+3 steps) compared well with soot concentrations determined with the chemothermal (CTO-375) method designed to specifically measure soot carbon $^{20-22}$.

In the meantime, the application of the IMPROVE method to differentiate 119 between char and soot has been extended to aerosols ²³⁻²⁸, sediments and soils ^{19, 29, 30}. 120 All these studies have shown that the IMPROVE protocol delivers results which 121 reasonably reflect the char and soot contributions to EC in the samples. For example, 122 the char/soot ratio was successfully used to distinguish different sources in aerosol 123 studies ^{28, 31}. A study of the spatial distribution of char and soot in urban, rural, and 124 remote areas ²⁷ revealed that char has local sources and is not far transported, while 125 soot is regionally dispersed, which is in line with our knowledge about the larger 126 particle size of char than of soot ³². The 150 yr historic reconstruction of char and soot 127 deposition from several sediment cores in eastern China¹⁹ showed similar historical 128 profiles for soot but different ones for char, which further confirms the different 129 transport modes of char and soot. The sedimentary soot profiles ¹⁹ well reflect the 130 131 industrialization history of China, showing an abrupt increase in the late 1970s, the timing of opening and economic reform of China. The stronger association of PAHs 132 with soot than with char in soils and sediments in the Guanzhong Basin in China³³ is 133 134 consistent with the results of the previous SRM test that showed a higher absorption capacity of PAHs by soot than by char ³⁴. The Holocene char and soot records
reconstructed from Lake Daihai reflected the paleo-wildfire occurrence and they
showed good correlations with climate changes and human activities ²⁹.

In recent years, brown carbon (BrC) has received increasing attention as a type 138 of carbonaceous aerosol which also absorbs light ³⁵. The light-absorbing 139 characteristics of BrC ³⁵ and char ²⁴ both are specific for certain wavelengths of the 140 light spectrum, and their light absorption is weaker than that of soot. BrC and char are 141 142 distinctively different. Char (or charcoal) particles are combustion residues that are inert and can exist in the environment millions of years ³⁶, whereas BrC is water 143 soluble and considerably more labile. Quantification methods of BrC and char are 144 different. Char is operationally measured as the carbon that remains after chemical or 145 thermal pretreatment ^{12, 37-39}, whereas BrC is measured as the carbon extractable by 146 water or organic solvents ^{35, 40, 41}. 147

148

S1.4. PAHs, OPAHs and AZAs analyses

Concentration of 29 parent- and alkyl-PAHs, 15 OPAHs and 4 azaarenes 149 (AZAs) were determined. A mixture of 7 deuterated PAHs and 2 deuterated OPAHs 150 were used as internal standard to quantify parent-/alkyl-PAHs and carbonyl-OPAHs, 151 152 respectively. A detailed description of the analytical procedure for the measurement is given by Bandowe and Wilcke ^{42, 43}. Briefly, about 2 g of sediment sample, 25 µL of 153 each of the deuterated OPAHs and PAHs mixtures were added as internal standards to 154 the accelerated solvent extractor (ASE) cell. Samples were extracted using ASE 200 155 156 (Dionex, Sunnyvale, CA, USA) with dichloromethane in a first extraction cycle, and

157	with $CH_3COCH_3/CH_2C_{12}/CF_3COOH$ (1%) (250:125:1 v/v/v) in a second extraction
158	cycle. The two extracts of each sample were combined, dried on Na ₂ SO ₄ , and cleaned
159	up/fractionated on silica gel (10% deactivated) columns. The PAHs fraction was
160	eluted with 15 mL hexane/dichloromethane (5:1 v/v), followed by 8 mL
161	dichloromethane and 5 mL acetone to elute OPAHs and AZAs. The PAHs and OPAHs
162	+ AZAs fractions were rotary evaporated to about 0.5 mL, transferred to a 2 mL GC
163	vial, spiked with 25 μL (22 $\mu g/mL)$ fluoranthene- D_{10} as recovery standard, put in the
164	refrigerator and followed by GC/MS measurement.
165	An Agilent 7890A gas chromatograph coupled to an Agilent 5975C mass
166	spectrometer in EI and SIM mode was used for measurements. The GC Oven program
167	for PAHs, OPAHs, and AZAs was the same as in previous papers ^{43, 44} . All data

recording and processing was done with the Agilent MSD ChemStation software 168 169 package.

Target compounds were quantified by the internal standard technique using 170 seven calibration standards prepared from target compound standards each spiked 171 172 with a constant concentration of internal standard. IBM SPSS 19.0 for Windows and 173 Origin 8.5 were used for statistical analysis and graphics.

174

S1.5. Quality assurance and quality control

175 All glassware was rinsed with acetone, machine-washed, baked at 250 °C for 12 h, and rinsed with high purity solvents before use. All solvents used for extraction, 176 column chromatography, and standard preparation were high purity for pesticide 177 178 residue analysis. To check and correct for possible contaminations during the

179 analytical procedure, we processed blanks made of diatomaceous earth at the beginning of each batch of 24 samples. Most compounds were either not detected in 180 the blanks or were measured in negligible quantities, significantly below those found 181 182 in the samples. The measured compound concentrations in the samples were corrected by subtracting the mean blank concentrations. We checked the accuracy of PAH 183 measurements by including the certified reference material ERM-CC013a (BAM, 184 Berlin, Germany) into our analyses. The mean recovery of all PAHs for which 185 certified values were provided was 110%. We checked the recovery of all internal 186 standards. The mean recovery of deuterated PAHs and OPAHs was 80% (67-88%). 187 188 All concentrations are reported without recovery correction.

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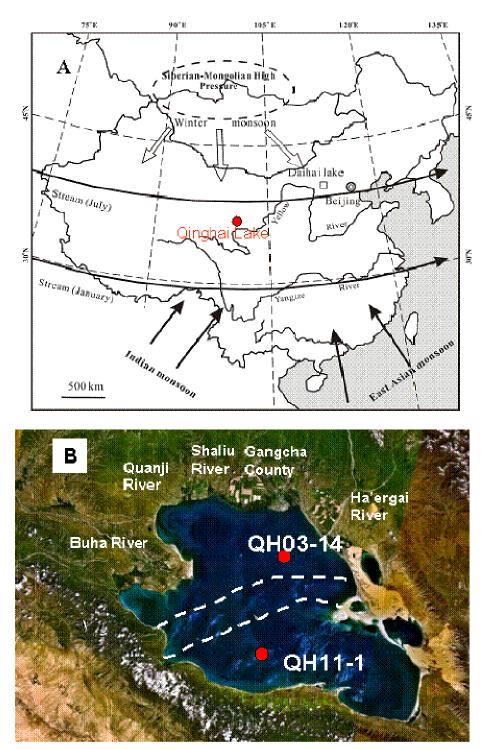
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320 Fig. S1. (A) Climatic systems of China, including the East Asian and Indian summer

- 321 monsoons, the Siberian–Mongolian High, and the Westerlies. (B) Location of the
- 322 sediment cores QH03-14 and QH11-1 obtained from different sub-basins of Lake
- 323 Qinghai.
- 324

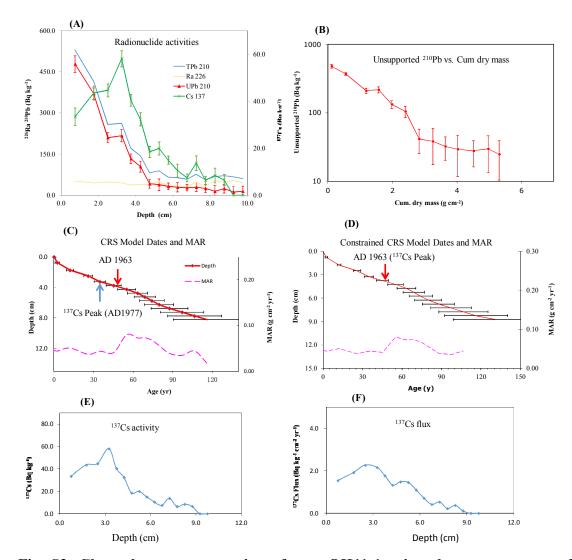


Fig. S2. Chronology reconstruction of core QH11-1 using the constant rate of 327 supply (CRS) ²¹⁰Pb model. (A) Measured Pb, Cs, and Ra activities, including total 328 ²¹⁰Pb (blue line), excess ²¹⁰Pb (red line with error bars), ²²⁶Ra (cvan line), and ¹³⁷Cs 329 (green line with error bars) concentrations along with depth variation; (B) Excess 330 ²¹⁰Pb against cumulative mass; (C) CSR model dates and corresponding mass 331 accumulation rate (MAR), which show that ¹³⁷Cs peak at 3.25 sediment depth occurs 332 at ~1977; (D) Constrained (3.75 cm through 1963) CRS model dates and 333 corresponding MAR; (E) ¹³⁷Cs peak vs. depth presenting one ¹³⁷Cs peak; (F) ¹³⁷Cs 334 flux calculated using the CRS model vs. depth presenting that the shape of the ¹³⁷Cs 335 peak is broad. 336

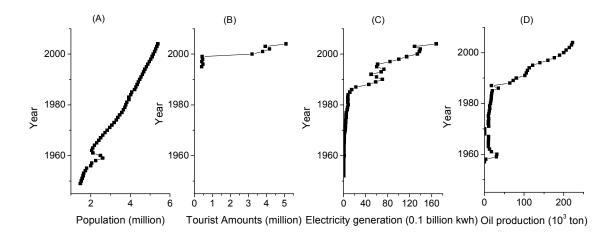


Fig. S3. History of the population numbers, number of tourists, extents of electricity
generation and oil production (or consumption) in Qinghai Province in the past 60
years. Tourism in Qinghai Province is mainly driven by in the attractions around Lake
Qinghai.

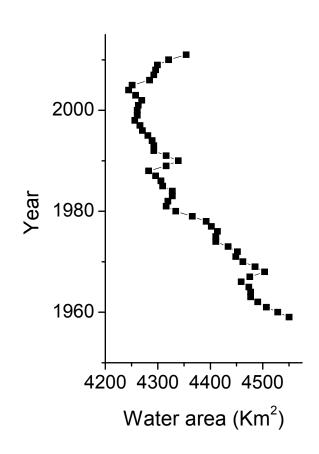


Fig. S4. History of the surface size of Lake Qinghai in the past 50 years.

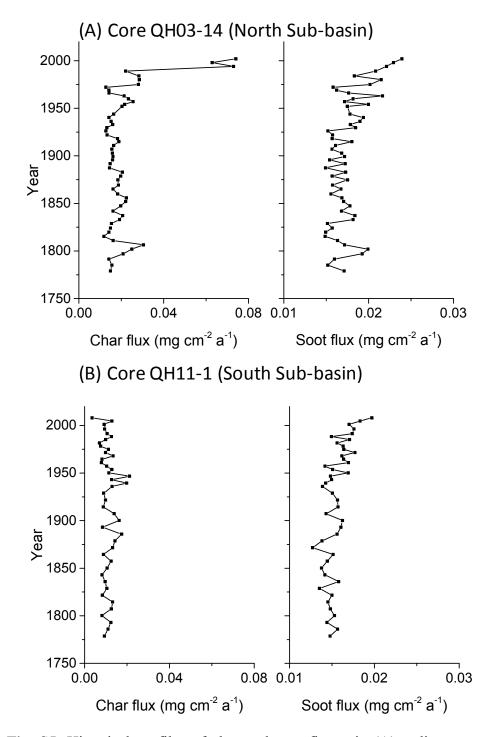


Fig. S5. Historical profiles of char and soot fluxes in (A) sediment core QH03-14 (northern sub-basin with more human inputs) and (B) sediment core QH11-1 (southern sub-basin).



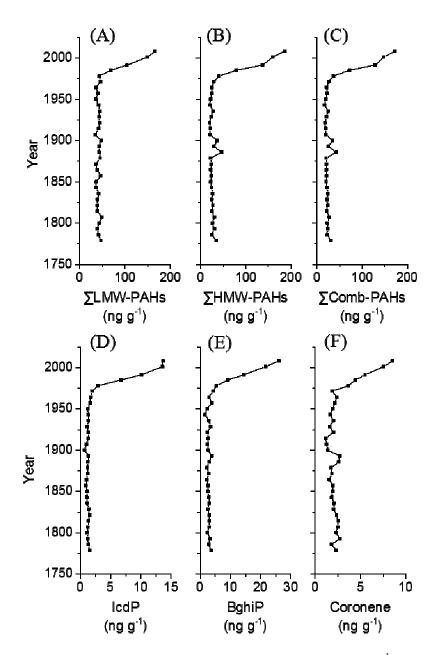


Fig. S6. Vertical distribution of the concentrations (ng g^{-1}) of (A) Σ LMW-PAHs; (B)

 \sum HMW-PAHs; (C) \sum COMB-PAHs; (D) IcdP (indeno[c,d]pyrene); (E) BghiP 359 (benzo[g,h,i]perylene; and (F) Coronene. See main text and Table S1 for the definition 360 of abbreviations.

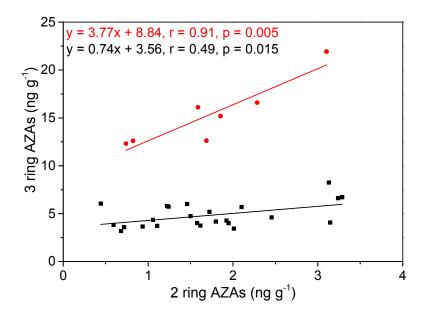


Fig. S7. Relationship between the concentrations of 2-ring and 3-ring azaarenes
(AZAs) in sediments of Lake Qinghai before (dark squares) and after (red
cycles) the 1960s. The considered 2-ring azaarene is quinoline, while 3-ring
azaarenes include benzo[h]quinoline, acridine and carbazole.

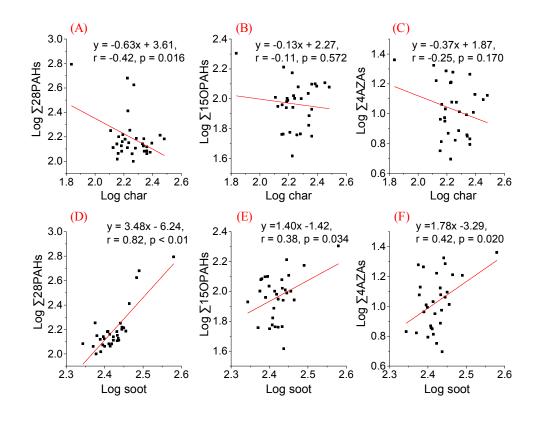
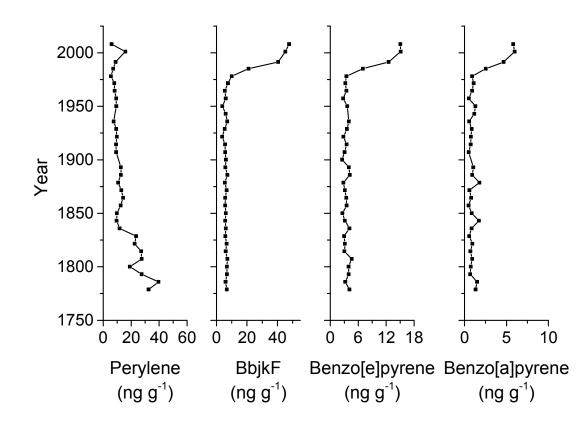


Fig. S8. Relationships between the concentrations of (A) $\sum 28$ PAHs (total measured PAHs without perylene) vs char, (B) $\sum 15$ OPAHs vs char, (C) $\sum 4$ AZAs vs char, (D) $\sum 28$ PAHs vs soot, (E) $\sum 15$ OPAHs vs soot, and (F) $\sum 4$ AZAs vs soot. Concentrations of PAHs, OPAHs, AZAs (ng g⁻¹) and char, soot (mg g⁻¹) are log-transferred.

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Fig S9. Vertical distribution of the concentrations of (A) perylene and its combustion-derived isomers (MW = 252) such as (B) benzo[b+j+k]fluoranthene (BbjkF), (C) benzo[e]pyrene and (D) benzo[a]pyrene in the sediment core QH11-1 of Lake Qinghai. benzo[b+j+k]fluoranthene is sum of benzo[b]fluoranthene, benzo[j]fluoranthene and benzo[k]fluoranthene.

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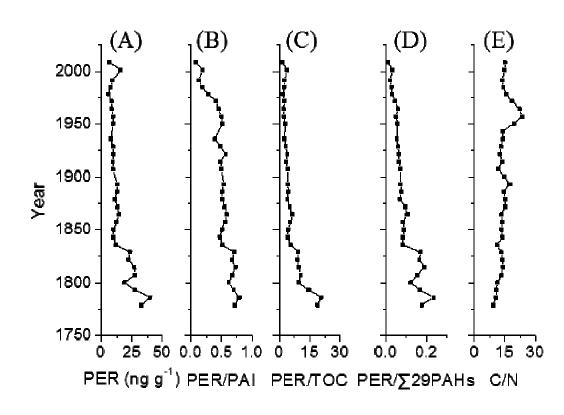


Fig. S10. (A) Perylene (PER) concentrations, and concentration ratios of (B) PER to its penta-aromatic isomers (PER/PAI), (C) concentration ratio of PER/total organic carbon (TOC), (D) concentration ratio of PER/sum of 29 PAHs ($\sum 29$ PAHs, including perylene), and of (E) TOC to total nitrogen concentrations (C/N) in Lake Qinghai.

Table S1. Statistical description concentrations of PACs, sum of concentrations of
 PACs and other sediment properties.

	Abbreviation	Minimum	Maximum	Average	Max/Min
azaarenes	AZAs				
Quinoline	QUI	0.44	3.29	1.71	7.
Benzo[h]quinoline	BhQ	0.48	13.2	3.51	27.
Acridine	ACR	0.57	5.73	1.11	10.
Carbazole	CAR	1.81	3.84	2.57	2.
Oxygenated PAHs	OPAHs				
1-Indanone	1-IND	1.02	10.2	4.55	9.
1,4-Naphthoquinone	1,4-NQ	1.20	33.7	11.7	28.
1-Naphthaldehyde	1-NALD	0.67	3.42	1.81	5.
2-Biphenylcarboxaldehyde	2-BPCA	0.11	7.64	1.98	71.
9-Fluorenone	9-FLU	4.69	20.0	10.4	4.
1,2-Acenaphthenequinone	1,2-ACEQ	7.35	64.0	17.8	8.
9,10-Anthraquinone	9,10 - AQ	9.71	43.1	15.8	4.
1,8-Naphtalic anhydride	1,8-NA	1.06	38.5	7.69	36.
2-Meth-9,10-anthraquinone	2-MAQ	4.82	24.2	12.4	5.
Benzo[a]florenone	BAFLU	2.29	37.1	8.89	16
7H-Benzo[d,e]anthracen-7-one	BdeAQ	0.93	10.74	3.43	11.
Benzo[a]anthracene-7,12-dione	BaAQ	0.41	8.24	2.22	20.
5,12-Naphthacenequinone	5,12-NQ	0.01	1.86	0.77	32
6H-Benzo[c,d]pyren-6-one	BcdPQ	0.13	14.7	3.00	11
Polycyclic aromatic					
hydrocarbons	PAHs				
1,2,3,4-Tetrahydronapthalene	TH-NAPH	0.91	10.3	2.04	11.
Naphthalene	NAPH	10.1	72.7	17.9	7.
2-Methylnaphthalene	2-MNAPH	3.36	19.3	7.58	5.
1-Methylnaphthalene	1-MNAPH	5.22	31.7	14.0	6.
Biphenyl	BIPH	7.37	33.2	11.5	4.
1,3-Dimethylnaphthalene	1,3-DMNAPH	9.25	68.5	22.4	7.
Acenaphthylene	ACEY	0.32	2.29	0.65	7.
Acenaphthene	ACEN	0.33	6.02	1.57	18.
Fluorene	FLUO	2.18	17.5	3.78	8.
Phenanthrene	PHE	14.8	81.6	23.9	5.
Anthracene	ANT	1.82	14.9	3.48	8.
2-Methylphenanthrene	2-MPHE	2.24	40.7	10.8	18.
3,6-Dimethylphenanthrene	3,6-DMPHE	0.57	5.88	1.28	10.
Fluoranthene	FLUA	0.71	13.0	2.52	18.
Pyrene	PYR	2.09	25.9	4.95	12.
Retene	RET	2.07	60.7	9.34	29.
Benzo[a]anthracene	BaA	0.72	9.78	1.94	13.
Chrysene+Triphenylene	CHR	1.61	20.7	3.77	12.

Benzo[b+j+k]fluoranthene	BbjkF	3.76	47.7	10.2	12.7
Benzo[e]pyrene	BeP	2.55	15.1	4.56	5.9
Benzo[a]pyrene	BaP	0.49	14.6	1.80	30.1
Perylene	PER	5.55	39.5	14.5	7.1
Indeno [1,2,3-cd]pyrene	IcdP	0.73	13.7	2.50	18.7
Dibenzo[a,h]anthracene	DahA	0.47	4.84	1.35	10.4
Benzo[g,h,i]perylene	BghiP	1.58	26.0	4.79	16.5
Coronene	COR	1.17	8.53	2.62	7.3
Sum of azaarenes	∑4AZAs	4.97	23.0	11.6	4.6
Sum of oxygen-containing PAHs	∑150PAHs	41.4	201.5	98.0	4.9
Sum of parent polycyclic					
aromatic hydrocarbons	∑29PAHs	109.3	628.4	188.0	5.7
\sum PAHs without perylene	∑28PAHs	99.6	622.3	174.4	6.2
$\sum 150$ PAHs/ $\sum 28$ PAHs		0.31	1.21	0.65	3.9
Low molecular weight parent					
PAHs	LMW-PAHs	34.3	166.4	51.3	4.9
High molecular weight parent					
PAHs without perylene	HMW-PAHs	20.4	185.5	40.8	9.1
LMW-/HMW-PAHs		0.76	2.17	1.50	2.8
\sum COM-PAHs ¹		17.2	172.3	36.8	10.0
∑COM-PAHs/∑28PAHs		0.13	0.31	0.19	2.4
Total nitrogen	TN	1.64	2.84	1.93	1.7
Total carbon	TC	49.7	62.7	55.5	1.3
Inorganic carbon	IC	11.5	38.4	27.8	3.3
Total organic carbon	TOC	17.2	43.2	27.7	2.5
Total sulfur	TS	2.28	4.47	3.53	2.0
TOC/TN	C/N	9.12	23.4	14.4	2.6
Loss on Ignition	LOI	0.10	0.20	0.14	2.0

408 ¹ \sum COM-PAHs: Combustion derived PAH = sum of FLUA, PYR, BaA, CHR, BbjkF, BeP, BaP, IcdP

409 and BghiP.

	EC	Char	Soot	Char/ Soot	Σ OPAH	∑AZAs	Retene	Perylene	Σ PAHs	OPAH/ PAH	Σ LMW	Σ HMW	LMW/ HMW	ΣСОМ	$\Sigma \text{COM}/$ ΣPAHs	TOC	C/N
EC	1	0.809	0.161	0.645	0.269	0.031	0.149	0.043	0.145	0.117	0.152	0.19	-0.235	0.194	0.305	0.039	-0.089
Char	0.809	1	-0.45	0.962	-0.068	-0.251	-0.338	0.189	-0.371	<u>0.356</u>	<u>-0.35</u>	-0.316	0.115	-0.313	-0.051	-0.271	-0.122
Soot	0.161	-0.45	1	-0.635	0.534	0.483	0.794	-0.235	0.844	-0.433	0.818	0.819	-0.550	0.820	0.548	0.515	0.068
Char/Soot	0.645	0.962	-0.635	1	-0.137	-0.307	-0.419	0.202	-0.491	<u>0.42</u>	-0.464	-0.445	0.243	-0.442	-0.184	-0.341	-0.119
Σ OPAH	0.269	-0.068	0.534	-0.137	1	0.676	0.69	-0.56	0.592	<u>0.376</u>	0.59	0.59	-0.323	0.593	0.305	0.559	0.306
∑AZAs	0.031	-0.251	0.483	-0.307	0.676	1	0.503	-0.608	0.529	0.04	0.48	0.515	-0.452	0.516	0.293	0.762	0.565
Retene	0.149	-0.338	0.794	<u>-0.419</u>	0.690	0.503	1	-0.281	0.905	-0.233	0.88	0.885	-0.513	0.888	0.577	0.547	0.074
Perylene	0.043	0.189	-0.235	0.202	-0.560	-0.608	-0.281	1	-0.157	-0.388	-0.165	-0.186	0.073	-0.192	-0.044	-0.515	-0.499
Σ PAHs	0.145	<u>-0.371</u>	0.844	-0.491	0.592	0.529	0.905	-0.157	1	-0.485	0.982	0.984	-0.647	0.984	0.692	0.609	0.064
OPAH/PAH	0.117	<u>0.356</u>	-0.433	<u>0.42</u>	0.376	0.04	-0.233	<u>-0.388</u>	-0.485	1	<u>-0.445</u>	<u>-0.448</u>	0.347	<u>-0.444</u>	-0.32	-0.187	0.105
Σ LMW-PAH	0.152	<u>-0.35</u>	0.818	-0.464	0.590	0.48	0.88	-0.165	0.982	<u>-0.445</u>	1	0.979	-0.582	0.978	0.698	0.577	0.016
Σ HMW-PAH	0.19	-0.316	0.819	-0.445	0.590	0.515	0.885	-0.186	0.984	<u>-0.448</u>	0.979	1	-0.714	1.00	0.793	0.581	0.026
LMW/HMW	-0.235	0.115	-0.55	0.243	-0.323	-0.452	-0.513	0.073	-0.647	0.347	-0.582	-0.714	1	-0.711	-0.87	<u>-0.409</u>	-0.075
$\Sigma \operatorname{COM}$	0.194	-0.313	0.82	-0.442	0.593	0.516	0.888	-0.192	0.984	<u>-0.444</u>	0.978	1.00	-0.711	1	0.795	0.582	0.027
$\Sigma \operatorname{COM}/\Sigma$ PAHs	0.305	-0.051	0.548	-0.184	0.305	0.293	0.577	-0.044	0.692	-0.32	0.698	0.793	-0.87	0.795	1	0.29	-0.165
TOC	0.039	-0.271	0.515	-0.341	0.559	0.762	0.547	-0.515	0.609	-0.187	0.577	0.581	<u>-0.409</u>	0.582	0.29	1	0.78
C/N	-0.089	-0.122	0.068	-0.119	0.306	0.565	0.074	-0.499	0.064	0.105	0.016	0.026	-0.075	0.027	-0.165	0.78	1

Table S2. Pearson correlation coefficients between sediment properties used in this study.
 Bold indicates that correlation is significant at the 0.01 level, and <u>Underline</u> indicates that correlation is significant at the 0.05 level in a two-tailed test.