

# The ancient Chinese civilization left remarkable signals in the marine environment since the Bronze Age

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## Article

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2                                   **environment since the Bronze Age**

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16

17    **Abstract**

18    To understand the onset of human civilization evolution recorded in the marine  
19    environment, a 6000-year record of black carbon (BC), including char and soot, was  
20    examined in a sediment core from the central South Yellow Sea. The results showed  
21    that the colder and drier climate dominated the variation in fire activity in northern

22 China after mid-Holocene via decreased precipitation and vegetation cover. The char  
23 released from the fire activity, can barely retrieve traces preserved in the marine  
24 sediments during the civilization evolution in China since ~1 ka BP. Fortunately, the  
25 soot-BC signal demonstrated that anthropogenic forces have overwhelmed the natural  
26 causes of soot emission since ~4 ka BP (Bronze Age) in northern China. The variation  
27 in soot closely matched periods when there was large-scale use of coal or charcoal after  
28 ~2 ka BP and when indigenous coking technology was promoted after ~1.3 ka BP, and  
29 times with low soot abundance coincided with periods of social unrest. This work  
30 provides evidence that the soot signal could be a robust tracer for tracking the  
31 civilization evolution, and the ancient Chinese civilization left remarkable soot signals  
32 in the marine environment since the Bronze Age.

33 **Key words:** Civilization evolution in China, black carbon, soot signal, sedimentary  
34 record, Bronze Age

35

## 36 **1. Introduction**

37 Industrialization is a revolutionary stage in the civilization evolution because it  
38 has changed the lifestyle of human beings and brought great conveniences to human life  
39 (1). However, the scale of carbon emissions since the industrial revolution have led to a  
40 rise in atmospheric greenhouse gases at an unprecedented rate due to the large-scale use  
41 of fossil fuels (*e.g.*, petroleum and coal) (2), which has significantly affected the surface

42 ecosystems (3), the atmospheric chemical composition (4), and even the regional and  
43 global climate (5). The energy structures always link to the civilization evolution since  
44 humans could use fire by biomass fuels as early as the middle Pleistocene (6, 7).  
45 However, the visible trace for the onset of the ancient human civilization in the  
46 environment is poorly constrained, which varies spatially over the earth and represents a  
47 key piece of information to the concept of the Anthropocene (2).

48         Black carbon (BC) is a highly aromatic, recalcitrant carbon species released both  
49 from the wildfire and anthropogenic fire for the main energy utilization in ancient  
50 human civilization. It has two subtypes, char and soot (8, 9), which can be distinguished  
51 by thermal optical reflectance due to their different burning temperatures (10, 11). BC is  
52 widely distributed in lake and ocean sediments via transportation by atmospheric  
53 circulation and surface runoff (12). Char particles (1~100  $\mu\text{m}$ ) are residues of  
54 combustion at temperatures of 300~600°C that retain some of the morphological and  
55 structural features of their precursors, and are a function of biomass burning. Soot  
56 particles (< 1  $\mu\text{m}$ ) are produced during high-temperature combustion (> 600°C), which  
57 is strongly related to anthropogenic fuels (*e.g.*, petroleum, coal, and charcoal) or high-  
58 intensity biomass burning (8, 9, 10, 13, 14, 15). The sedimentary records of char and  
59 soot can provide information for tracking past changes in fire activity forced by climate  
60 change or human activities on multi-decadal to millennial scales (16). For example, the  
61 char/soot ratio in sediment cores from the eastern China marginal seas was used to

62 reconstruct the variation in the energy structures in China over the last 200 years (9),  
63 and char has been used to track fire activities during the Holocene in Africa (12), the  
64 Pacific Islands (17), Europe (18), and North America (19).

65 From the time when human beings use fire by biomass fuels as early as the  
66 middle Pleistocene to the period of the large-scale use of fossil fuels in the industrial  
67 revolution, there have been several stages in the evolution of human civilization,  
68 Neolithic Age, Bronze Age, and Iron Age. Sediment cores provides striking evidence of  
69 past contamination linked to the civilization evolution in various continents during  
70 human history (9, 20). Northern China is the birthplace of Chinese civilization, which is  
71 one of the oldest human civilizations. Therefore, this is a unique area for tracing  
72 civilization evolution in the environment. Studies on the historical records in the  
73 sedimentary strata for China have concentrated on fire activity in the large river basins  
74 in southern China (20) or examined local scales (15, 21, 22) associated with climate  
75 change and human activity, while seldom on the time node of civilization evolution in  
76 China.

77 The South Yellow Sea (SYS) lies in the path of the East Asian continental  
78 outflow driven by the East Asian winter monsoon (EAWM). It can receive and preserve  
79 large amounts of land-based materials from northern China as a natural archive via  
80 atmospheric deposition and ocean current transportation (23). Therefore, this is a unique  
81 and ideal place for tracing the civilization evolution in northern China. This work

82 establishes a 6000-year high-resolution BC (char and soot) record from sediment core  
83 ZY2, taken from the central SYS, to reveal the fire history of a large-scale northern  
84 China since the mid-Holocene by exploring the relative importance of climate change  
85 and human activity, and trace the human civilization evolution in China. This reveals  
86 that the evolution of human civilization in China left remarkable signal in the marine  
87 sediment since the Bronze Age.

88

## 89 **2. Materials and Methods**

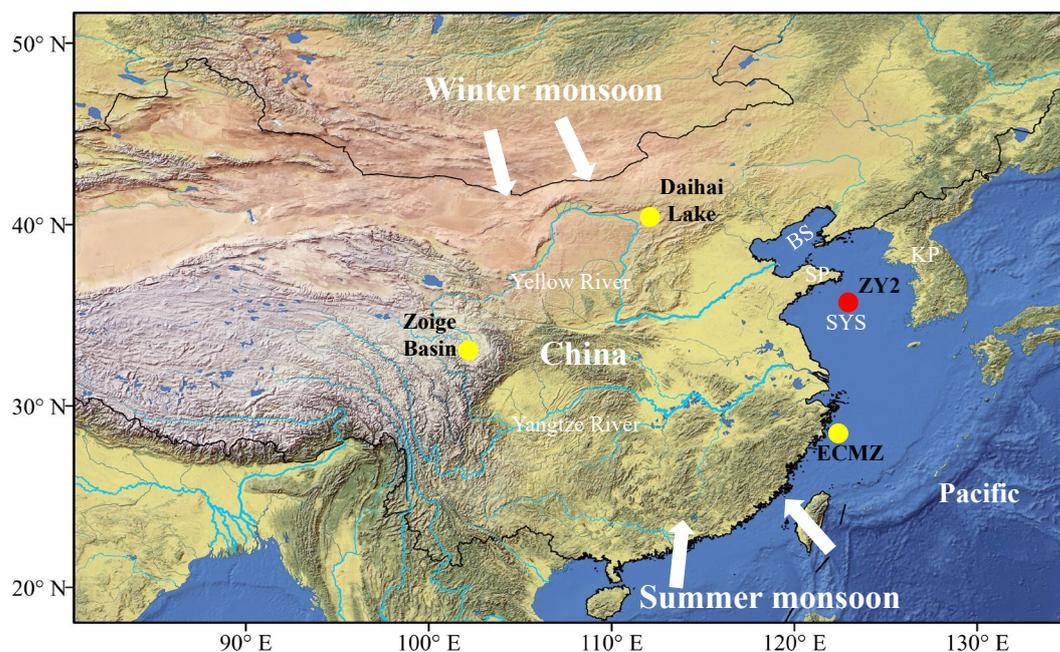
### 90 *2.1. Study area and sampling*

91 The SYS is a semi-closed marginal sea in the western Pacific located between  
92 China and the Korean Peninsula (Fig. 1). It is indirectly influenced by the Yellow and  
93 Yangtze Rivers (24). Under the influence of the EAWM, the SYS is downwind of the  
94 Asian continental outflow in spring and winter (25), and large amounts of land-based  
95 materials are deposited in the marginal seas (26–28). Sediment core ZY2 (35°31'N,  
96 122°39'E) was collected in the SYS at a water depth of 68.5 m from the R/V Dong-  
97 Fang-Hong 2 in 2006. It is 342 cm long. The core was sampled at 1 cm intervals and  
98 kept at –20°C. The lithology and chronology of the core were reported previously (24).  
99 The age at the bottom of the core is ~6.2 ka BP, which is roughly when the modern  
100 current circulation of the Yellow Sea developed (29). The homogeneous gray, dark gray

101 clayey silt of the core promised a stable sedimentary environment with sufficient

102 sediment supply (24).

103



104 **Fig. 1.** A map showing the winter and summer monsoons, Bohai Sea (BS), South  
105 Yellow Sea (SYS), Yellow and Yangtze Rivers, Pacific Ocean, Korean (KP) and  
106 Shandong (SP) Peninsulas, ZY2 sediment core (red circle), and locations of fire-record  
107 sediment core studied previously (yellow circles) (20, 21, 22).

## 108 2.2. BC analysis

109 The abundance of BC was analyzed in 300 samples, at 1-cm intervals in the first

110 258 cm of the core and 2-cm intervals thereafter. The pretreatment method followed

111 Fang *et al.* (2018) (9). BC was measured at Lanzhou Institute of Arid Meteorology,

112 China Meteorological Administration using a thermal/optical carbon analyzer (DRI

113 Model 2001A; American Desert Research Institute, Reno, NV, USA), which adopted

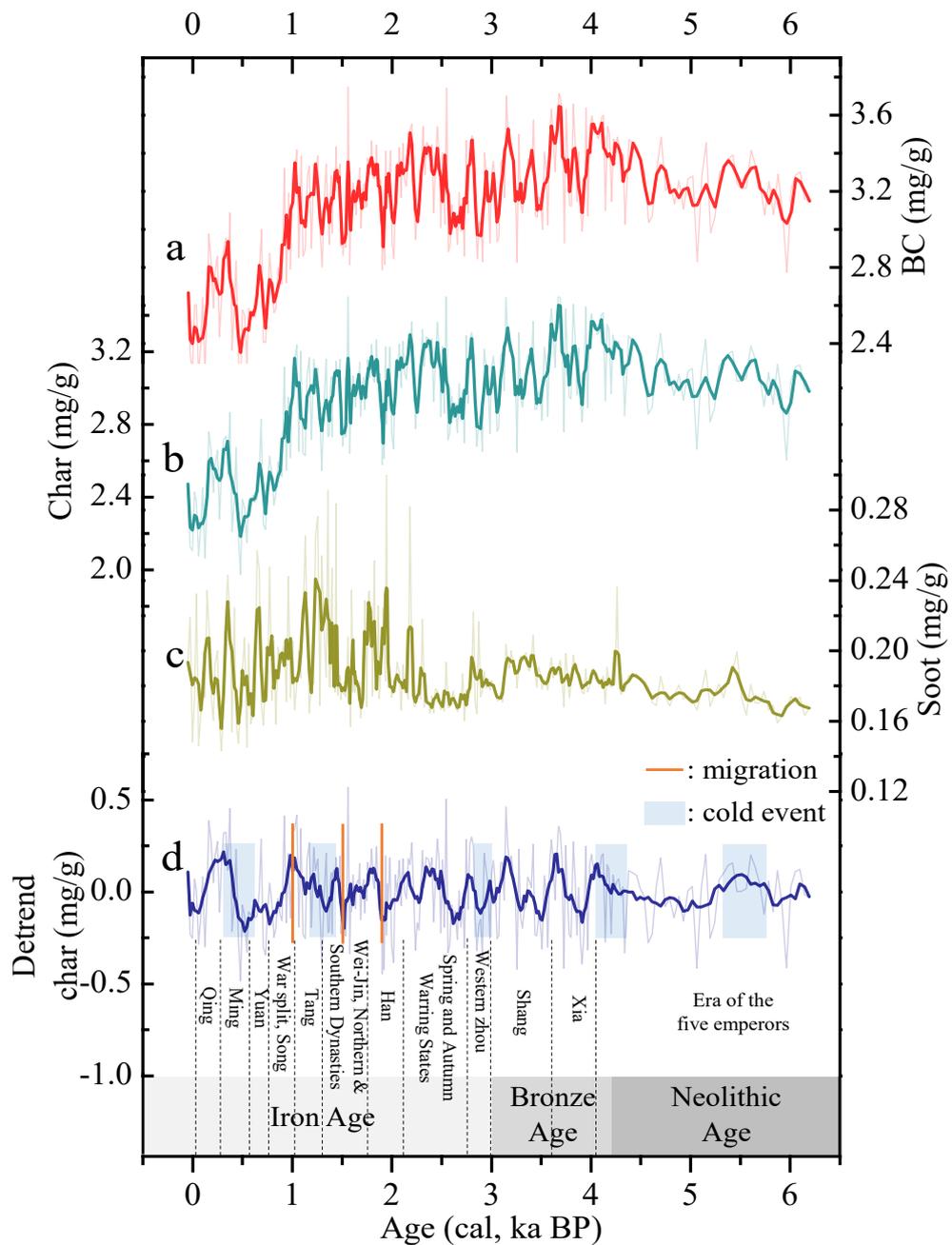
114 the interagency monitoring of protected visual environment (IMPROVE) protocol. To  
115 begin, the analyzer oven was heated to 550°C in 100% He. This was switched to an  
116 O<sub>2</sub>/He mixture (2%/98%) and the oven was heated from 550°C to 700°C and then to  
117 800°C. The gas generated with these temperature gradients was catalytically oxidized to  
118 CO<sub>2</sub> in an oxidation furnace (MnO<sub>2</sub>), and then converted into CH<sub>4</sub> in a reducing  
119 environment. The carbon content was measured with a flame ion detector, which has  
120 three peaks corresponding to organic carbon, char, and soot at three temperature stages  
121 (550, 700, and 800°C). BC equals the sum of char and soot (10) and is expressed in  
122 mg/g dry sediment. Replicate analyses showed that the relative error was within 5%,  
123 and the total carbon content of the blank filter sample was below 0.05 mg.

124

### 125 **3. Results**

126 The BC in sediment core ZY2 ranged from 2.1 to 3.7 mg/g (mean  $3.1 \pm 0.31$   
127 mg/g;  $1\sigma$ ;  $n = 300$ ), and increased slightly from ~6.2 to 4.0 ka BP and subsequently  
128 decreased. This showed a series of centennial to multi-centennial fluctuations, with an  
129 obvious decrease at ~1 ka BP (Fig. 2a). The char (range 2.0–3.5 mg/g, mean  $3.0 \pm 0.31$   
130 mg/g;  $1\sigma$ ;  $n = 300$ ) showed a similar trend to BC (Fig. 2b). The detrend char result (Fig.  
131 2d) obtained by 50% loess regression (to present the char variation on a centennial  
132 timescale) showed no significant periodicity in a spectral analysis with a multi-taper  
133 method and wavelet transform analysis. The strong correlation between char and BC ( $R^2$

134 = 0.99,  $P < 0.01$ ,  $n = 300$ ) and the lack of a correlation ( $R^2 = 0.020$ ,  $P = 0.014$ ,  $n = 300$ )  
135 between soot and BC suggested that char is the main component of BC. The soot  
136 abundance (range 0.14–0.31 mg/g, mean  $0.19 \pm 0.025$  mg/g;  $1\sigma$ ;  $n = 300$ ) increased  
137 slightly at mid-Holocene (Fig. 2c), similar to the char variation at that time. The soot  
138 abundance variation was distinct from that of char after  $\sim 4$  ka BP, with a pronounced  
139 increasing trend at  $\sim 2$  ka BP and a downtrend at  $\sim 1.3$  ka BP (Fig. 2c).



140 **Fig. 2.** Sediment BC data for core ZY2 since ~6.2 ka BP: the variation in (a) BC,  
 141 (b) char, and (c) soot; (d) the char detrend with 50% loess regression (Fig. S1). Signal  
 142 smoothing averaging three adjacent points for (a) to (c), and seven points for (d).

143 **4. Discussion**

144 *4.1. The climate change dominated the variations of char*

145 The similar production processes shared by BC (including char and soot) and  
146 polycyclic aromatic hydrocarbons (PAHs) could result in inference with BC sources by  
147 the signatures of PAHs (30-32). The source pattern of pyrogenic high-ring PAHs in the  
148 Yellow Sea sediments compared well with that of aerosol PM<sub>2.5</sub> (*i.e.*, particles with an  
149 aerodynamic diameter < 2.5 μm) collected upwind from northern Chinese cities, Beijing  
150 and Qingdao, demonstrating the significant influence of atmospheric deposition on the  
151 input of combustion-derived substances into the marginal sea off northern China (25).  
152 Moreover, the atmospheric deposition of BC contributed ~51% to the total input of BC  
153 in the Bohai Sea and more than that in the SYS (23). Thus, EAMW-forced atmospheric  
154 deposition played a dominant role in the BC input in the SYS. The history of the  
155 EAWM intensity has been reconstructed by analyzing the sensitive grain size (24). The  
156 lack of a correlation between the abundance of BC and grain size ( $R^2 = 0.015$ ,  $P < 0.05$ ,  
157  $n = 300$ ) suggests that the intensity of the EAWM was not the dominant factor causing  
158 the variations in BC (both char and soot) (char:  $R^2 = 0.011$ ,  $P = 0.058$ ; soot:  $R^2 = 0.068$ ,  
159  $P < 0.01$ ). Additionally, the absence of a correlation between the abundance of BC and  
160 the clay, silt, and sand contents suggests that the BC abundance was not influenced by  
161 the dilution effects of other sediment fractions. Therefore, the sedimentary records of  
162 BC in sediment core ZY2 closely reflect the emissions due to fires in northern China.

163           The general decline in fire activity recorded by the char in core ZY2 (Fig. 3a) is  
164 consistent with char or BC records from sediment cores from the inner shelf of the East  
165 China Sea (20), the Zoige Basin in the eastern Tibetan Plateau (22), and Lake Daihai in  
166 north-central China (15, 21) (Fig. 1, Fig. S2). This suggests that the gradual weakening  
167 in fire activity since the mid-Holocene is a common phenomenon across the monsoon  
168 region of China. In the mid-Holocene, in the Northern Hemisphere, the earth was near  
169 its perihelion in the summer, which would result in a stronger radiation (33) and a  
170 greater land–ocean thermal contrast, thus increase temperature gradients, and strengthen  
171 the Asian Summer Monsoon (34). With the precession cycle, the earth gradually  
172 approached its apogee in summer in the Northern Hemisphere in the late Holocene (33).  
173 The earth’s orbit decreased summer insolation in the Northern Hemisphere (Fig. 3b)  
174 (35) and induced the Asian Summer Monsoon. Oxygen isotopes are used to track past  
175 summer monsoons and the precipitation intensity (36). Stalagmite  $\delta^{18}\text{O}$  in Dongge and  
176 Sanbao Caves in China shows a gradual increase since ~7 ka BP (36, 37), indicating  
177 gradual weakening of the precipitation since the mid-Holocene. Simultaneously, the  
178 temperature (Fig. 3d) (38), moisture (Fig. 3c) (39), and vegetation cover (Fig. 3e) (40)  
179 in northern China showed downward trends.

180           In general, lower temperatures are associated with decreased fire activity via  
181 vegetation productivity and the incidence of fire-promoting climate conditions (41).  
182 However, the char value of sediment core ZY2 unexpectedly increased slightly from

183 ~6.2 to 4.0 ka BP (Fig. 3a) when the climate became colder and drier. Note that fire  
184 activity is not only associated with temperature but also with vegetation cover and  
185 precipitation. Pollen analyses of boreholes from the Yellow River Delta showed that  
186 evergreen and broadleaved deciduous forest thrived in northern China (indicated by  
187 high *Quercus*, *Carpinus/Ostrya*, and *Ulmus/Zelkova* pollen counts) from 9.8 to 4.5 ka  
188 BP in a warmer, wetter climate, and a significant reduction in deciduous *Quercus* pollen  
189 and an increase in conifer *Pinus* pollen at ~4 ka BP corresponded to a drier climate (42).  
190 Moreover, the percentage of tree pollen in a profile from Lake Daihai (north-central  
191 China) showed a clear downward trend since the mid-Holocene (Fig. 3e) (39, 40).

192         Additionally, the decrease in precipitation not only affected the vegetative cover  
193 (fuel abundance) but also increased its flammability (41). Fire responds differently to  
194 precipitation and depends on whether vegetation cover (fuel abundance) is initially a  
195 factor limiting fire spread. In arid and semi-arid environments, increases in precipitation  
196 tend to increase fires (43), whereas increased precipitation reduces fire activity in humid  
197 environments (44). Therefore, the unexpected increase in fire activity from ~6.2 to 4.0  
198 ka BP could be attributed to a threshold effect of precipitation and vegetation cover on  
199 fire activity. Specifically, in the mid-Holocene, when northern China had a humid  
200 environment and vegetation cover (fuel abundance) was not a factor limiting fire spread,  
201 decreases in precipitation could enhance the flammability of vegetation and increase fire  
202 activity. Subsequently, as precipitation continued to decrease, the amount of vegetation

203 decreased and limited fire propagation. This induced the decrease in fire activity with  
204 the continuous reduction in precipitation and vegetation since ~4 ka BP (Fig. 3a).

205         Although the millennial-scale, large-area decrease in fire activity in northern  
206 China since ~4 ka BP can be explained by the natural climate dynamics (45), the  
207 evolution of human civilization should not be ignored in this process (46). The  
208 Zhoukoudian archaeological discovery illustrated that Beijingers had the ability to  
209 control fire behavior as early as the middle Pleistocene (7). Since then, fire has been  
210 used for heating, lighting, cooking, driving beasts, living in groups, and making and  
211 creating new living materials (47). The Yellow River Basin in northern China is the  
212 origin of the ancient Chinese civilization and millet agriculture. Millet agriculture was  
213 developed in the pre-Yangshao Period (8.5~7 ka BP) (48), and fire has been used to  
214 clear forest for cultivation and settlement since ~7.7 ka BP in China (47). Subsequently,  
215 millet agriculture developed rapidly in the Yellow River Basin (49). The number of  
216 archaeological sites in northern China increased substantially after ~7 ka BP and even  
217 more markedly between ~4.5 and 3 ka BP (Fig. 3f) (50). However, the number of  
218 archaeological sites in northern China declined sharply at ~3 ka BP, which may be  
219 related to climate change and the growth characteristics of millet, which is a drought-  
220 tolerant crop that is sensitive to temperature change (48). The gradually weakened  
221 insolation and decreased temperature in the Northern Hemisphere is not always  
222 conducive to millet cultivation. The decrease in archaeological sites after ~3 ka BP (Fig.

223 3f) corresponded to a major shift in the subsistence economy from predominantly  
224 animal husbandry and plant cultivation to mobile pastoralism across present-day  
225 semiarid northern China, which left fewer traces in the archaeological records than  
226 sedentary communities (51). This variation in human activity (number of archaeological  
227 sites) was not recorded by the char in core ZY2, suggesting that climate change was the  
228 dominant factor affecting fires on a millennial-scale over a large area, rather than human  
229 activity. However, there has a more pronounced decrease in fire activity at ~1 ka BP  
230 (Fig. 3a), when a semi-pastoral, semi-agricultural economy was established in northern  
231 China to accommodate the cold-dry climate (52), and fire management was used to  
232 prevent uncontrolled wildfires to limit damage to settlements and agricultural resources  
233 (53), which might have led to the obvious decrease in fire activity at ~1 ka BP.

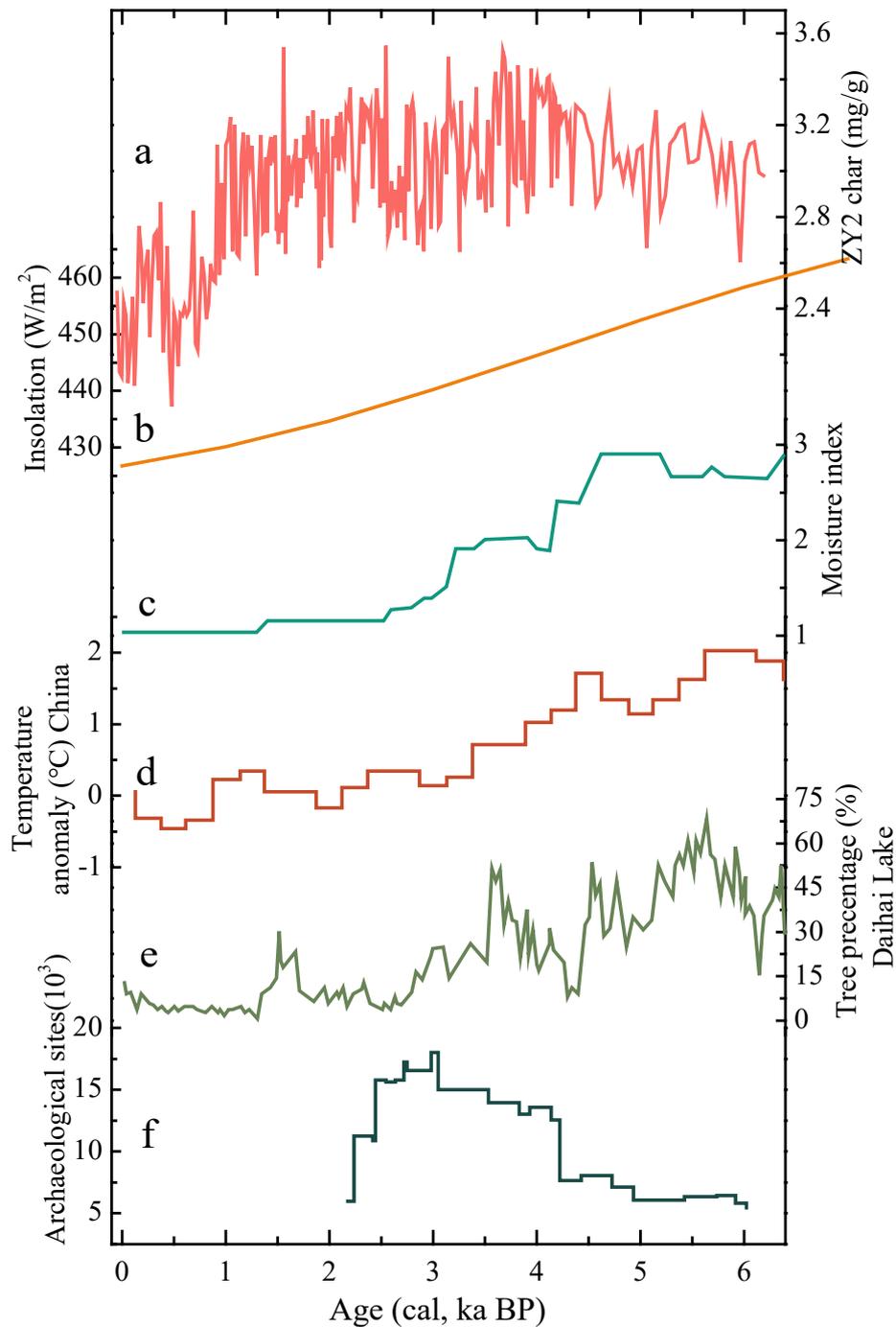
234         The record of fire based on char in core ZY2 also showed a series of centennial  
235 to multi-centennial fluctuations (Fig. 2d). The North Atlantic cold events (5.5, 4.2, 2.8,  
236 1.4, and 0.4 ka BP) during the mid-late-Holocene (54, 55) were large-scale phenomena  
237 rather than local ones (56). Most of them could be attributed to weakening solar  
238 activity, which in turn affected the ocean and atmospheric circulation on earth, causing  
239 abrupt climate change (57, 58). The period of abrupt change corresponds to the  
240 variation in the Holocene Asian Summer Monsoon (36), which could have affected the  
241 fire activity in northern China. The detrend char records showed that the 5.5 and 4.2 ka  
242 BP cold events coincided with episodes of high fire activity; while the cold events of

243 2.8 and 1.4 ka BP and the Little Ice Age (~0.4 ka BP) were periods of low fire activity  
244 (Fig. 2d). This means that the threshold effect (as discussed above) of climate change on  
245 fire activity could also partly explain the centennial fluctuations in fire in northern  
246 China, suggesting that the cold events before ~4 ka BP promoted fire activity, while  
247 subsequent cold events decreased the fire activity.

248         The paleoclimatic and archaeological evidence suggests that the abrupt droughts  
249 and cold climate resulted in warfare, population migration, and dynasty change (59).  
250 This is especially true for northern China, where society was more sensitive to severe  
251 climatic conditions due to the relatively high latitudes (60). There were three large  
252 population migrations from northern to southern China during 1.9–1.8 ka BP (late Han  
253 Dynasty), 1.6–1.4 ka BP (Wei-Jin Southern & Northern Dynasties), and 1.1–0.9 ka BP  
254 (Tang-Song Dynasty) (61). The reduction in the population of northern China generally  
255 reduced the intensity of human activity, which decreased fire activities (20). The  
256 migrations in 1.9–1.8 and 1.6–1.4 ka BP coincided with low fire activity (Fig. 2d);  
257 while the migration in 1.1–0.9 ka BP coincided with high fire activity (Fig. 2d). The  
258 Tang-Song period occurred during the Medieval Warm Period, which likely  
259 overwhelmed the population reduction in northern China and dominated fire occurrence  
260 in this period.

261         Different spatial scales have different fire regimes. The climate–vegetation  
262 relationship can generally drive both global and sub-continental fire regimes, while

263 human-induced fires are prominent mainly on a local scale (62-64). Small-scale human  
264 activity can affect natural fire regimes, even in pyrogeographic settings in which climate  
265 exerts strong, top-down controls on fuel (65). Fortunately, muddy shelf areas may  
266 contain an integrated burning imprint of the mainland, which avoids the influence of  
267 small-scale human activities on local fires. The discussion above means that variations  
268 of char are dominated by climate change, both on the millennium timescale and the  
269 centennial timescale. Even if the obvious decrease in fire activity at ~1 ka BP could  
270 attribute to the establishment of the semi-pastoral, semi-agricultural economy and the  
271 fire management in northern China, it was ultimately associated with the cold-dry  
272 climate. Therefore, char can barely retrieve traces left in the environment during the  
273 civilization evolution in China.



274 **Fig. 3.** Comparison of the millennial-timescale variation in BC records from ZY2 with  
 275 other records since 6.2 ka BP. (a) Fire activities recorded by char from core ZY2 in the  
 276 SYS (this study); (b) extraterrestrial insolation of mid-month insolation 65N for July  
 277 (33); (c) moisture index-based pollen in northern China (39); (d) temperature anomaly  
 278 in China (38); (e) percentage of tree pollen recorded from the sediment core in Lake  
 279 Daihai (40); (f) change in the total number of archaeological sites in northern China  
 280 from 6 to 2 ka BP (50).

281

## 282 4.2. The soot variation footprint the ancient civilization evolution in China

283 The soot abundance increased slightly during the mid-Holocene (Fig. 2c),  
284 similar to the char variation at that time. The correlation between soot and char at 4~6.2  
285 ka BP was significant (Fig. 4a;  $R^2 = 0.46$ ,  $P < 0.01$ ,  $n = 58$ ; removing two outliers, Fig.  
286 S3). Soot is produced via high-temperature ( $> 600^\circ\text{C}$ ) gas-to-particle conversion under  
287 dry conditions and can reflect high-intensity biomass burning and is generated with  
288 char (14). Therefore, the slight increase in soot during the mid-Holocene could be  
289 predominantly attributed to natural factors. However, the correlation between soot and  
290 char decreased during 3~4 ka BP (Fig. 4b;  $R^2 = 0.33$ ,  $P < 0.01$ ,  $n = 60$ ) and showed no  
291 correlation after ~3 ka BP (Fig. 4f), suggesting the influence of anthropogenic factors  
292 on soot emission. Besides release from high-intensity biomass burning, soot can be  
293 generated from the combustion of fuel (*e.g.*, petroleum, coal, and charcoal) (13, 14).  
294 Although the Hou Hanshu, a famous Chinese historical document about the Han  
295 Dynasty, indicates that China first collected and used petroleum in the Wei-Jin,  
296 Southern & Northern Dynasties (1730–1369 BP), petroleum was not used widely (66).  
297 However, China has long used coal and charcoal as an important source of energy (67).

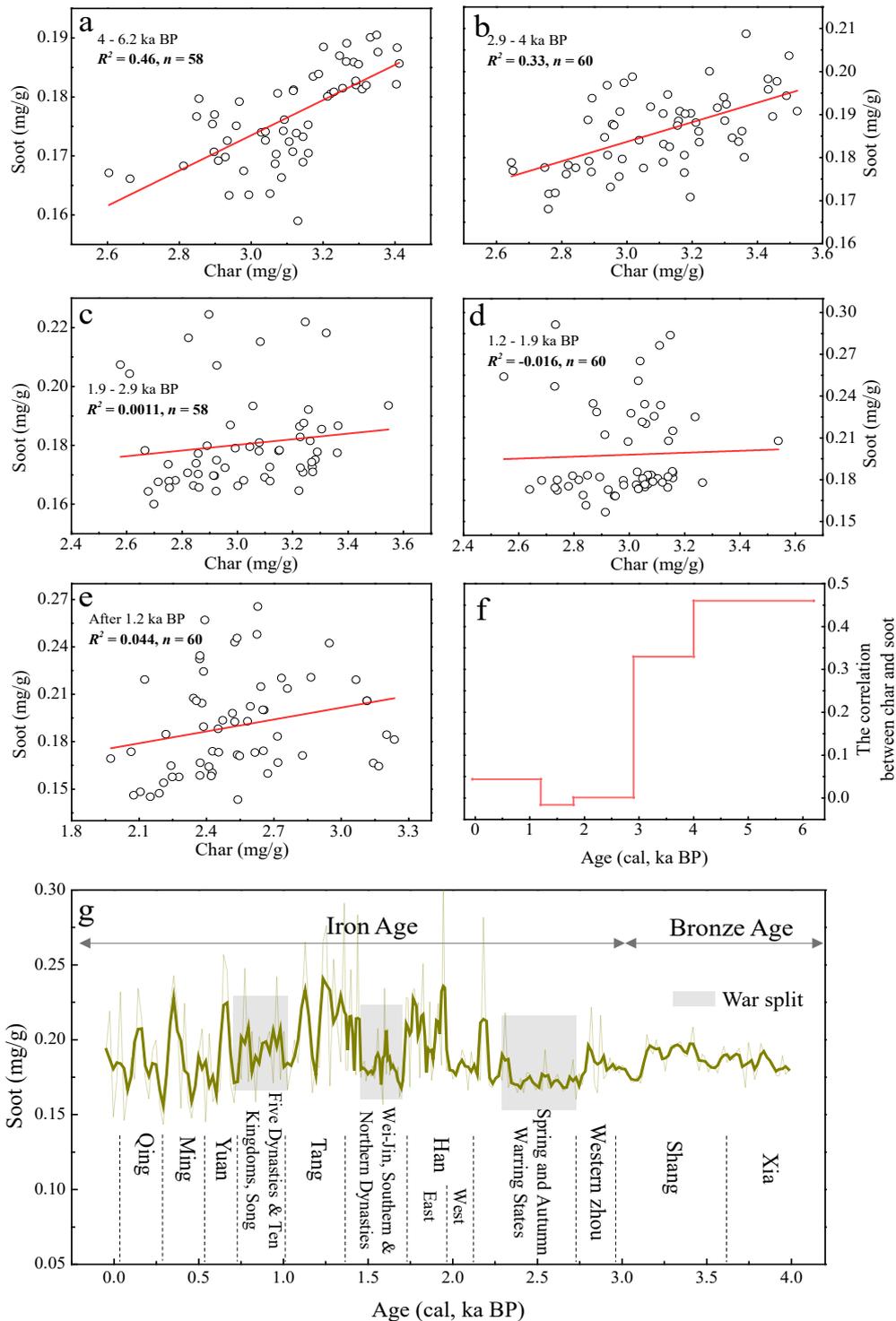
298 The Xia Dynasty, the first Chinese dynasty, was established more than 4000  
299 years ago, when China entered the Bronze Age. Bronze is an alloy of copper and tin or  
300 lead. Its melting point is between 700 and 900°C, which exceeds the production

301 temperature of soot ( $> 600^{\circ}\text{C}$ ). At that time, charcoal and bamboo charcoal were the  
302 major fuels for smelting bronze (68). At smelting temperatures, charcoal fuel promoted  
303 soot production, which caused the increase in soot during 3~4 ka BP (Fig. 4g). The  
304 anthropogenic soot emissions reduced the correlation between soot and char during 3~4  
305 ka BP (Fig. 4f).

306 China entered the Iron Age during the Western Zhou Dynasty (~3 ka BP). The  
307 smelting temperature of iron is about  $1500^{\circ}\text{C}$ , meaning that more fuel is used for  
308 smelting (68). During the Iron Age, soot and char were not correlated, suggesting that  
309 anthropogenic factors dominated soot emission. Soot abundance increased at ~2 ka BP  
310 (Fig. 4g), corresponding to the Han Dynasty (2156–1730 BP), when coal and charcoal  
311 fuel began to be used for large-scale metal smelting and agricultural tool production (67,  
312 68). The Song Dynasty (990–671 BP) started a national coal monopoly, and coal  
313 became an important resource related to the national economy and people's livelihoods  
314 during the Ming and Qing Dynasties (582–39 BP) (67, 69). However, soot abundance  
315 decreased beginning in ~1.3 ka BP (Fig. 4g), when indigenous coking technology was  
316 developed (70). In the Tang Dynasty (1332–1043 BP), coke was in its embryonic stage.  
317 By the Song Dynasty (990–671 BP), coking technology had matured and coke ovens  
318 were studied in archeology (70). In indigenous coking technology, a kiln with an  
319 ignition hole in the side wall is used to ignite the coking coal stacked in the kiln. This

320 suggests that the application of indigenous coking technology reduced the emission of  
321 soot compared with the direct combustion of raw coal.

322 China underwent several dynasty changes and united and split after the Xia  
323 Dynasty (4096~3625 BP). The Spring and Autumn Warring States (2721~2171 BP),  
324 Wei-Jin Southern & Northern Dynasties (1730~1369 BP), and Five Dynasties & Ten  
325 Kingdoms (1043~971 BP) periods were the three main splits in Chinese history,  
326 corresponding to the main three valleys in soot abundance at ~2.5, ~1.5, and ~0.8 ka BP  
327 (Fig. 4g). Periods with low soot abundance generally coincided with dynasty changes  
328 (Fig. 4g), when there were more wars. During periods of social stability, coal and  
329 charcoal were the main energy source for agricultural tool and handicraft production,  
330 metal smelting, and heating (68). Wars led to social unrest and the displacement of  
331 people, reducing soot emissions from coal and charcoal use in people's daily lives.



332 **Fig. 4.** (a) to (e) show the correlations between char and soot at 0.05–1.2, 1.2–1.88,  
 333 1.88–2.9, 2.9–4, and, 4–6.2 ka BP, respectively. (f) Change in the correlation between  
 334 char and soot since ~6.2 ka BP; (g) changes in soot from 0 to 4 ka BP and the  
 335 corresponding Chinese dynasties; signal smoothing was performed by averaging three  
 336 adjacent points.

## 337 **5. Conclusions and Implications**

338           The char-BC record revealed that the colder, drier climate before ~4 ka BP  
339 enhanced the fire activity slightly; this was followed by an obvious reduction in fire  
340 activity. The decoupling between climate change and fire occurrence suggested that the  
341 climate-induced precipitation and vegetation cover had a threshold effect on the fire  
342 regime. The synergistic effect of human activity and climate change resulted in a  
343 significant decline in fire ~1ka BP. Ultimately, the climate dominated the variation in  
344 fire activity in northern China both on the millennium timescale and the centennial  
345 timescale, therefore the char can barely retrieve traces persevered in the marine  
346 environment during the civilization evolution in China. However, the variation in soot  
347 clearly marked the large-scale use of coal and charcoal since ~2 ka BP (Han Dynasty)  
348 and adoption of indigenous coking technology after ~1.3 ka BP (Tang Dynasty), which  
349 is consistent with historical documents and archaeological discoveries. Periods of social  
350 unrest in the cold weapon era coincided with the low soot abundance, suggesting that  
351 wars reduced in soot emission from coal and charcoal use in people's daily lives. The  
352 soot signal demonstrated that anthropogenic forces overwhelmed natural factors as the  
353 cause of soot emissions after ~4 ka BP (Bronze Age) in northern China. The time node  
354 represents a key piece of information to establish the natural background of soot-BC in  
355 the marine sediments and to specify the concept of the Anthropocene.

356

357

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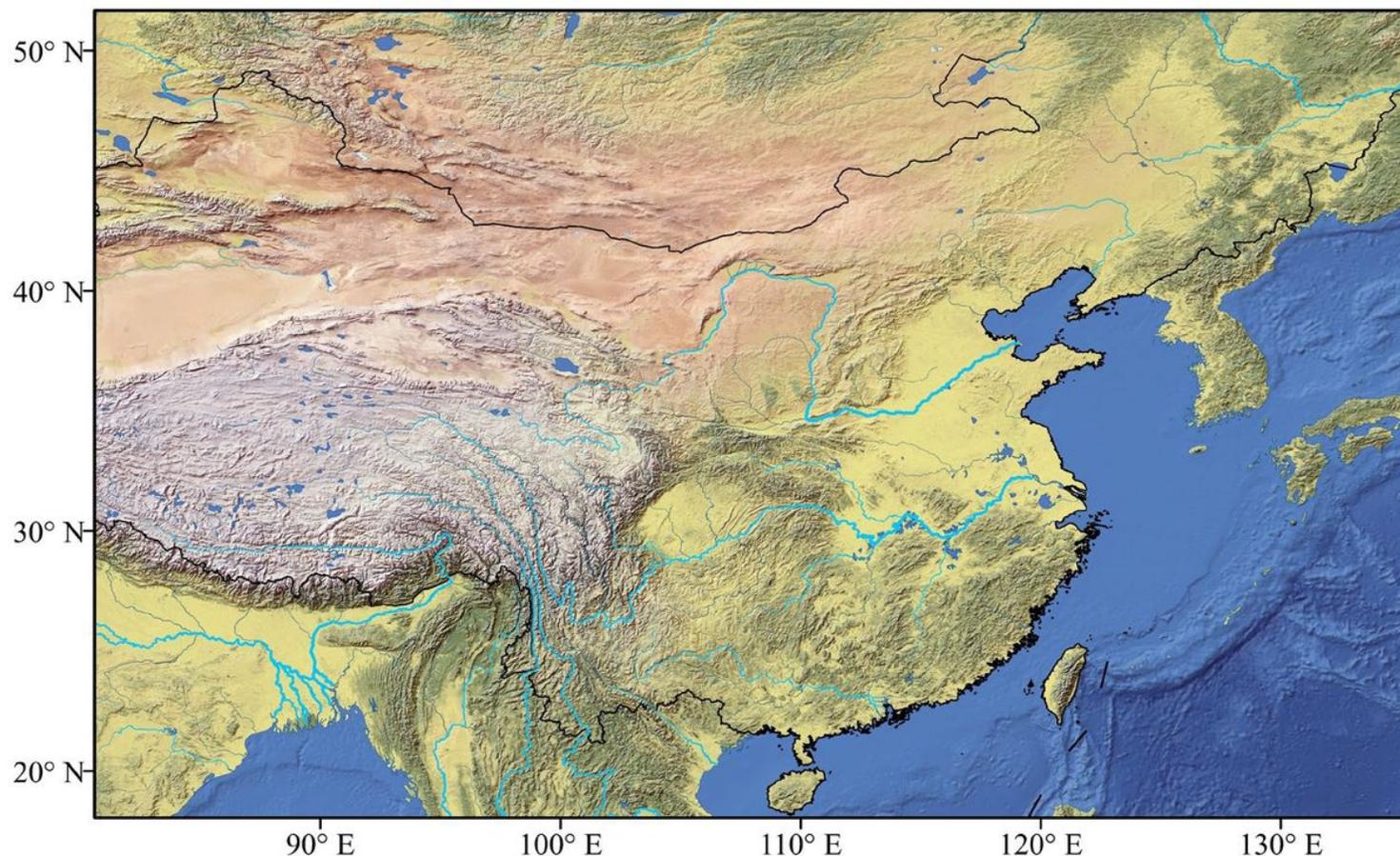
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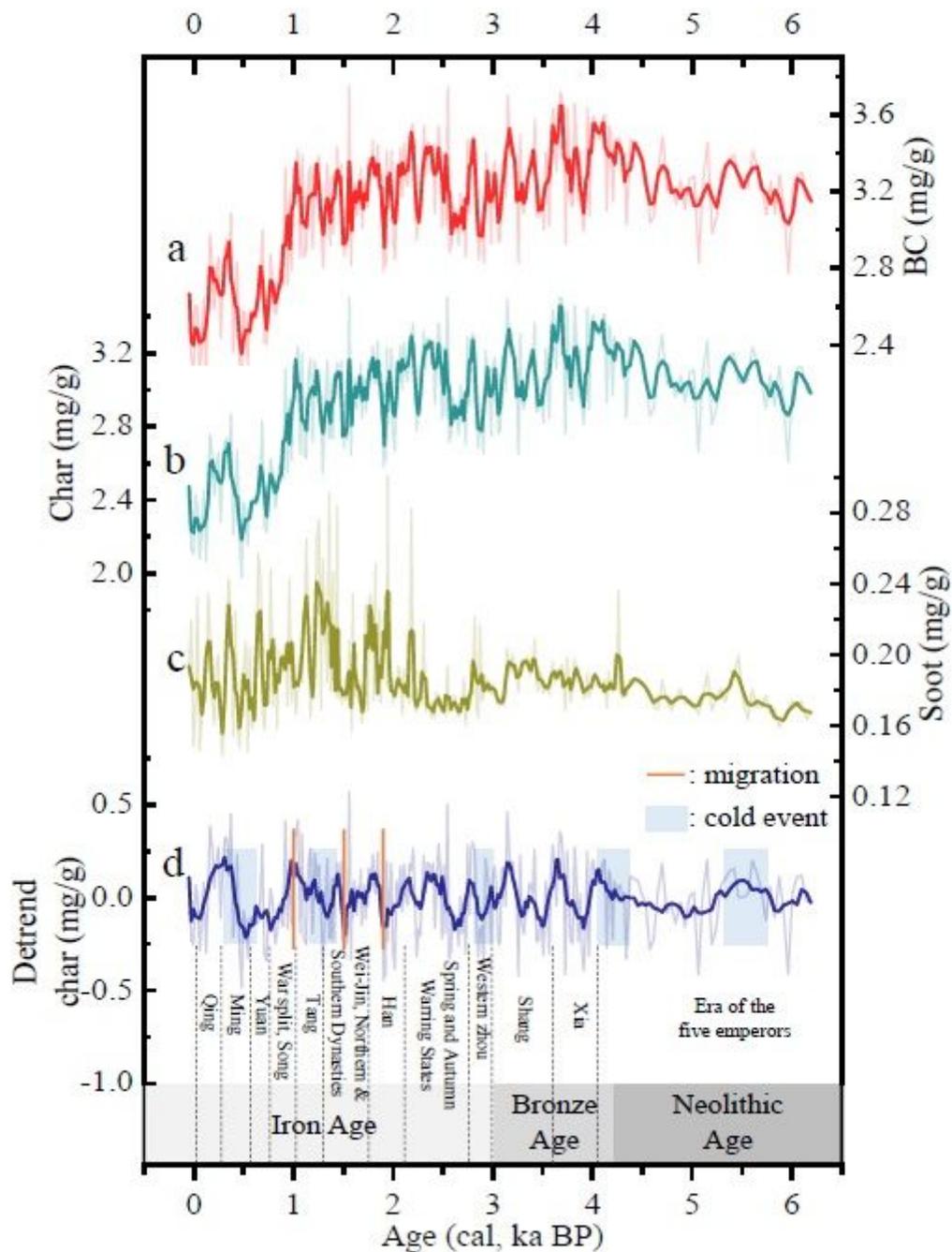
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## Figures



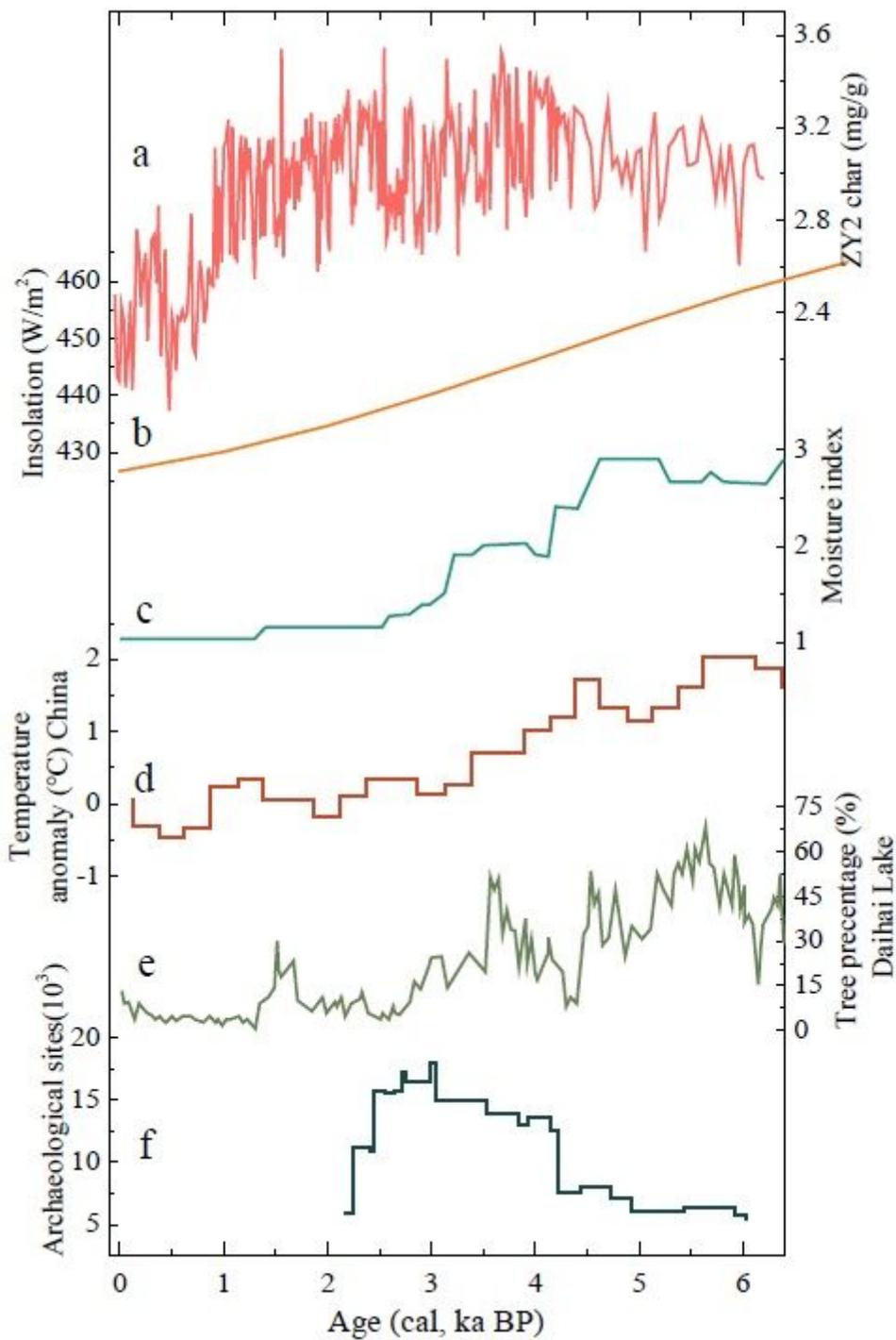
**Figure 1**

A map showing the winter and summer monsoons, Bohai Sea (BS), South Yellow Sea (SYS), Yellow and Yangtze Rivers, Pacific Ocean, Korean (KP) and Shandong (SP) Peninsulas, ZY2 sediment core (red circle), and locations of fire-record sediment core studied previously (yellow circles) (20, 21, 22). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



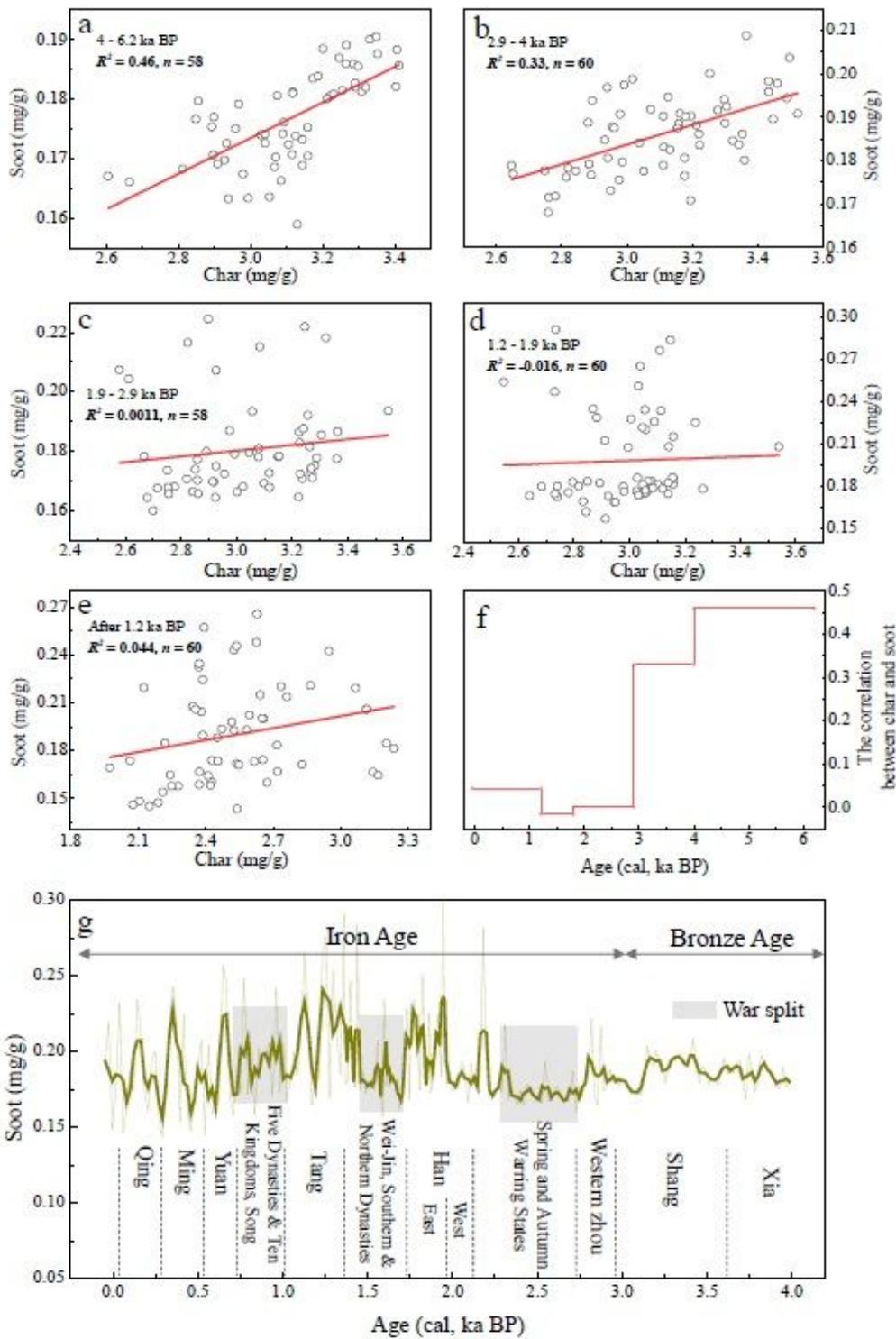
**Figure 2**

Sediment BC data for core ZY2 since ~6.2 ka BP: the variation in (a) BC, (b) char, and (c) soot; (d) the char detrend with 50% loess regression (Fig. S1). Signal smoothing averaging three adjacent points for (a) to (c), and seven points for (d).



**Figure 3**

Comparison of the millennial-timescale variation in BC records from ZY2 with other records since 6.2 ka BP. (a) Fire activities recorded by char from core ZY2 in the SYS (this study); (b) extraterrestrial insolation of mid-month insolation 65N for July (33); (c) moisture index-based pollen in northern China (39); (d) temperature anomaly in China (38); (e) percentage of tree pollen recorded from the sediment core in Lake Daihai (40); (f) change in the total number of archaeological sites in northern China from 6 to 2 ka BP (50).



**Figure 4**

(a) to (e) show the correlations between char and soot at 0.05–1.2, 1.2–1.88, 1.88–2.9, 2.9–4, and, 4–6.2 ka BP, respectively. (f) Change in the correlation between char and soot since ~6.2 ka BP; (g) changes in soot from 0 to 4 ka BP and the corresponding Chinese dynasties; signal smoothing was performed by averaging three adjacent points.

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