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## Authors

Cai, Shuhui Jin, Guiyun Tauxe, Lisa <u>et al.</u>

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# Archaeointensity results spanning the past 6 kiloyears **Gassifrom eastern China: Implications for extreme behaviors** of the geomagnetic field

Shuhui Cai<sup>a,b</sup>, Guiyun Jin<sup>c</sup>, Lisa Tauxe<sup>b,1</sup>, Chenglong Deng<sup>a,d</sup>, Huafeng Qin<sup>d</sup>, Yongxin Pan<sup>d,e</sup>, and Rixiang Zhu<sup>4</sup>

<sup>a</sup>State Key Laboratory of Lithospheric Evolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029, China; <sup>b</sup>Scripps Institution of Oceanography, University of California, San Diego, La Jolla, CA 92093-0220; School of History and Culture, Shandong University, Jinan 250100, China; <sup>d</sup>University of Chinese Academy of Sciences, Beijing 100049, China; and <sup>e</sup>Key Laboratory of the Earth and Planetary Physics, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029, China

Contributed by Lisa Tauxe, November 16, 2016 (sent for review July 20, 2016; reviewed by Lennart Vincent de Groot, and Monika Korte)

Variations of the Earth's geomagnetic field during the Holocene are important for understanding centennial to millennial-scale processes of the Earth's deep interior and have enormous potential implications for chronological correlations (e.g., comparisons between different sedimentary recording sequences, archaeomagnetic dating). Here, we present 21 robust archaeointensity data points from eastern China spanning the past ~6 kyr. These results add significantly to the published data both regionally and <sup>22</sup> q:12, 13 globally. Taking together, we establish an archaeointensity reference curve for Eastern Asia, which can be used for archaeomagnetic dating in this region. Virtual axial dipole moments (VADMs) 25 **Q:14** of the data range from a Holocene-wide low of ~27 to "spike" values of ~166 ZAm<sup>2</sup> (Z: 10<sup>21</sup>). The results, in conjunction with 26 Q:15 our recently published data, confirm the existence of a decrease in paleointensity (DIP) in China around ~2200 BCE. These low intensities are the lowest ever found for the Holocene and have not been reported outside of China. We also report a spike intensity of 165.8  $\pm$  6.0 ZAm<sup>2</sup> at ~1300 BCE ( $\pm$ 300 y), which is either a prelude to or the same event (within age uncertainties) as spikes first reported in the Levant.

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rchaeomagnetism is an effective way to understand the Availation of the geomagnetic field over periods of centuries to millennia during the Holocene. Large fluctuations of the geomagnetic field over the past few thousands of years have been reported, for example, as archaeomagnetic jerks in Europe (1, 2)and eastern Asia (3, 4), as spikes in the Levant (5–7), Turkey (8), and North America (9), as large decreases in paleointensity (DIPs) [DIPs of Kent and Schneider (10)] at ~3000 BCE and  $\sim$ 2200 BCE as well as a possible local high around 1300 BCE in China both reported by Cai et al. (11, 12). Apparent progress has been made on understanding variations of the geomagnetic field during the Holocene in the past few years (13). However, detailed pictures of the global field remain indistinct. Therefore, large numbers of globally distributed data of the highest quality are necessary to further characterize the features of the geomagnetic field. However, the existing paleointensity data from eastern Asia, especially those considered to be "high quality" (passing strict criteria), are sparse. In this study, we carried out detailed rock magnetic and paleointensity studies on samples collected from eastern China spanning the last 6 kyr; these will supplement the published dataset of this area significantly and provide further context for the elusive features of the geomagnetic field mentioned above.

#### **Materials and Methods**

The artifacts studied in this paper come from four locations in eastern China, including Shandong, Liaoning, Zhejiang, and Hebei provinces (Fig. 1A). We investigated various materials varying from pottery and porcelain sherds to bricks (Fig. 1 B-E) collected from living sites and kilns, whose ages span the past ~6 kyr. The detailed sampling background and the list of sample

information are in Supporting Information, Archaeomagnetic Background Q:20 and Sampling, and Table S1. Rock magnetic experiments, including hysteresis loops, isothermal remanent magnetization (IRM) acquisition curves, firstorder reversal curves (FORCs), variation of magnetization versus temperature (M-T), and scanning electron microscope (SEM) images as well as elemental spectrum analysis, were conducted on representative samples. The "IZZI" paleointensity protocol (14), as well as partial thermal remanent magnetization (pTRM) checks (15), total TRM anisotropy correction (16), and cooling rate correction (17), was adopted in this study. The detailed experimental procedures can be found in Supporting Information, Experimental Procedures.

#### Results

The rock magnetic results (Figs. S1-S3; Supporting Information, Rock Magnetic Results) indicate thermally stable fine-grained magnetite and titanium (Ti)- and/or aluminum (Al)-substituted magnetite as the dominant magnetic carriers for most samples, which suggest the suitability of these samples for paleointensity experiments. To obtain the most robust paleointensity data, we need to select the results based on a series of criteria [Paterson

#### **Significance**

The geomagnetic field is an intriguing fundamental physical property of the Earth. Its evolution has significant implications for issues such as geodynamics, evolution of the life on the Earth, and archaeomagnetic dating. Here, we present 21 Q:11 archaeointensity data points from China and establish the first archaeointensity reference curve for eastern Asia. Our results record rarely captured extreme behaviors of the geomagnetic field, with an exceptionally low intensity around ~2200 BCE (hitherto the lowest value observed for the Holocene) and a "spike" intensity value dated at  $\sim$ 1300  $\pm$  300 BCE (either a precursor to or the same event as the Levantine spikes). These anomalous features of the geomagnetic field revealed by our data will shed light on understanding geomagnetic field during the Holocene.

Author contributions: S.C. and R.Z. designed research; S.C. and H.Q. performed research; G.J. contributed new reagents/analytic tools; S.C., L.T., and C.D. analyzed data; S.C., L.T., and Y.P. wrote the paper; G.J. provided archaeological samples and age constraints; L.T. the first author is a postdoc in my laboratory; I have participated in all of the analyses conducted there and helped with data analysis and writing of the manuscript; C.D. provided mentorship and support of the first author; H.Q. provided assistance in the Beijing paleomagnetic laboratory for analyses; Y.P. provided mentorship of the first author during the project and helped with manuscript writing; R.Z. provided access to the Beijing laboratory, was primary mentor of the first author during doctoral research, and provided O:16 monetary support for the project. Q:17, 18 118

Reviewers: L.V.d.G., Utrecht University; and M.K., University of Potsdam. The authors declare no conflict of interest.

Data deposition: All data have been deposited in the Magnetics Information Consortium (MagIC) database, https://earthref.org/MagIC. This is the primary database for all paleo- Q:19 magnetic and rock magnetic data.

<sup>1</sup>To whom correspondence should be addressed. Email: ltauxe@ucsd.edu.

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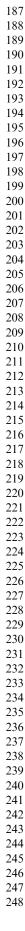
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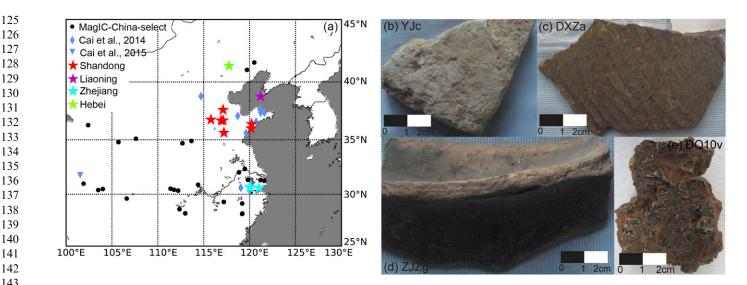


Fig. 1. (A) Site map of this study and the published data from China. Red/magenta/cyan/green stars are the locations of Shandong/Liaoning/Zhejiang/Hebei in this study. Light blue diamonds and downward-pointing triangles represent data locations published by Cai et al. (11, 12). Black solid circles represent locations of the archaeointensity data in China from the MagIC database after data selection. For data selection criteria, please see the text. (B-E) Various archaeomagnetic artifacts analyzed in this study, including brick from Yinjia site, Dezhou, Shandong (B); pottery fragments from Daxinzhuang and Zhaojiazhuang sites in Shandong (C and D); and slag from Laoshushan site, Huzhou, Zhejiang (E).

et al. (18)], for example, those suggested by Shaar and Tauxe (19). The selection statistics used in this study are listed in Table S2. Based on these, 97 specimens from 21 samples out of 407 specimens from 72 samples are considered to yield robust paleointensity estimates. The accepted results at the sample level are listed in Table S3, whereas those at the specimen level are

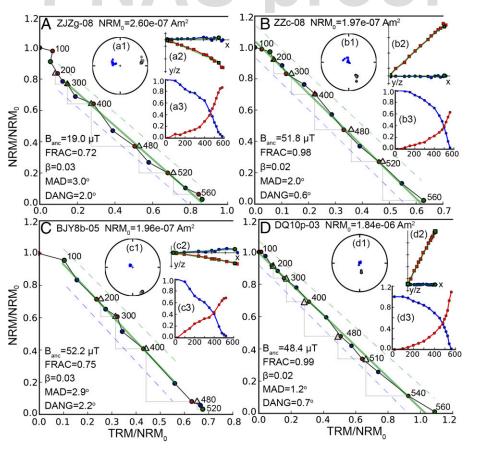
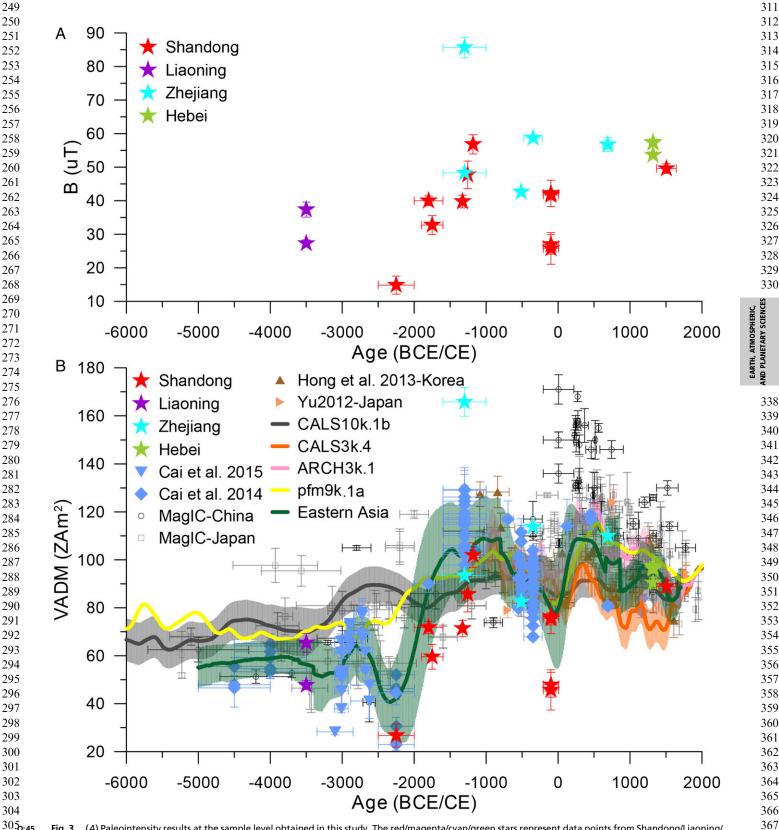


Fig. 2. Arai plots and the associated equal area projections (A1-D1), orthogonal projections (A2-D2), and natural remanent magnetization (NRM) lost-TRM Q:44 gained curves (A3-D3) of representative accepted specimens. Numbers on the Arai plot and orthogonal projections are temperature steps in centigrade (degrees Celsius). The plots are made with the software of Thellier\_GUI (19). For detailed description of these plots, please see the reference.



305**0:45** Fig. 3. (A) Paleointensity results at the sample level obtained in this study. The red/magenta/cyan/green stars represent data points from Shandong/Liaoning/ Zhejiang/Hebei. (B) Comparison of the VADMs in this study with the published data in eastern Asia and predictions from the global models. Light blue diamonds and downward-pointing triangles are published data in Cai et al. (11, 12). Black circles/squares are the selected published data in China/Japan from the MagIC database. Peach rightward-pointing triangles and brown triangles are recently published data from Japan/Korea (4, 20). The gray/orange/pink/ yellow lines are the predictions from global models of CALS10k.1b (25)/CALS3k.4 (24)/ARCH3k.1 (22)/pfm9k.1a (23), respectively, evaluated at the center of 309<sub>Q:46</sub> China (35°N, 105°E). The green line is the running average curve of Eastern Asia (calculated with our data and recently published data (4, 11, 12, 20)], whereas the shading represents 1 SD in the bootstrapped results.

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373 listed in Table S4. Fig. 2 A-D shows representative plots of ac-374 cepted specimens; these generally show good linearity on the Arai plots and have a single directional component going straight 375 to the origin except for a limited secondary component removed 376 377 by 100–150 °C in the orthogonal projection plots (Fig. 2A2–D2). The anisotropy and cooling rate corrections are shown in Fig. S4. 378 Alterations of the samples during the anisotropy correction ex-379 periments are all less than 8% except one over 10% (Fig. S4A), 380 whereas those during cooling rate correction are all limited, with 381 the percentage values of less than 3% (Fig. S4C). The ratios of 382 maximum and minimum eigenvalues of the ATRM tensors ( $\tau_1/\tau_3$ ) 383 vary between 1.02 and  $\sim 2.25$  ( $\sim 82\%$  of them are less than 1.5). The 384 extent of anisotropy correction is generally between 0.9 and 1.1 (Fig. S4B) with some exceptions of  $\sim 0.65$  and 1.3, whereas the cooling 385 rate correction is generally less than 8% (Fig. S4D). 386

The paleointensity values determined for four locations range from 14.8 to 85.7  $\mu$ T (Fig. 3*A*). The data transformed to virtual axial dipole moments (VADMs) (Fig. 3*B*) range from ~27 to ~166 ZAm<sup>2</sup>.

#### Discussion and Conclusions

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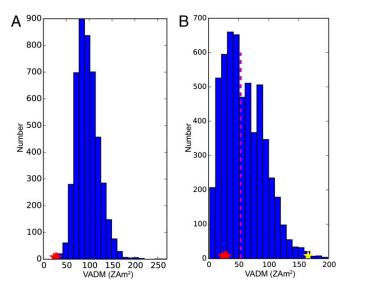
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392 Compilation of the Regional Model of Eastern Asia. Here, we present <sup>393</sup> q:21 21 archaeointensity data points from eastern China spanning the 394 q:22 past ~6 kyr. We compare our results with those published from 395 eastern Asia that met minimal acceptance criteria (only those obtained through a double-heating protocol, based on averages 396 of at least three specimens and with an SD of mean intensity less 397 than 10% or 5 µT), allowing us to detect the regional variation of 398 the geomagnetic field (Fig. 3B). Our data are generally in good 399 400 **Q:23** agreement with published data from eastern Asia at similar time periods, especially with those data published recently (4, 11, 12, 401 20). However, we document larger field variations including 402 extremely low ( $\sim 27 \text{ ZAm}^2$ ) and high ( $\sim 166 \text{ ZAm}^2$ ) values. 403 q:24 Combining our data with those recently published (4, 11, 12, 20), 404 we calculated the paleointensity variation curves (green line in 405 Fig. 3B) using a parametric bootstrap technique similar to that 406 used by Gallet et al. (21). We resampled 1,000 times at each data point considering uncertainties of both age and VADM, and 407 then applied a running average with a time window of 200 y 408 409 **Q:25** shifted by 10 y on the dataset (only time intervals including more than three data points were calculated). The established curve is 410 411 **Q:26** a composite archaeointensity reference curve for eastern Asia, which has applications for archaeomagnetic dating in this area. 412 The data for this curve can be found in Table S5. 413

Our revised eastern Asian curve agrees well with the ARCH3k.1 414 (22) and pfm9k.1a (23) models over the past 3 kyr, but deviates 415 from the older CALS3k.4 (24) and CALS10k.1b (25) models at 416 certain time periods, perhaps because of the effect of including 417 sedimentary data in the CALS type models, which are not absolute and are difficult to calibrate (26) and may be overly 418 smoothed. At ages older than ~3 ka, both CALS10k.1b and 419 pfm9k.1a models are in poor agreement with our data, especially 420 when the field reaches the minima at ~3000 BCE and ~2200 421 BCE and the local maxima around 1300 BCE (Fig. 3B). Our data 422 Q:27 therefore have the potential for greatly improving future global 423 field models. 424

425 Extreme Behaviors of the Geomagnetic Field. Cai et al. (11) reported two extreme low intensities with VADMs equal to or less than 426 30 ZAm<sup>2</sup> at  $\sim$ 2200 BCE from Liangchengzhen (LCZ) and 427 Zhaojiangzhuang (ZJZ) site in Shandong. Our results record 428 429 **Q:28** another low-intensity value (~26.7 ZAm<sup>2</sup>) from the ZJZ site. Including the data from Sichuan reported by Cai et al. (12), we now 430 have recorded four low-intensity values from three different 431 sampling sites in total. Taken together, these data strongly sup-432 port the existence of periods of very low paleointensity or "DIPs" 433 in China at ~3000 BCE and ~2200 BCE, especially at the latter 434 period. These low intensities are the lowest yet determined from



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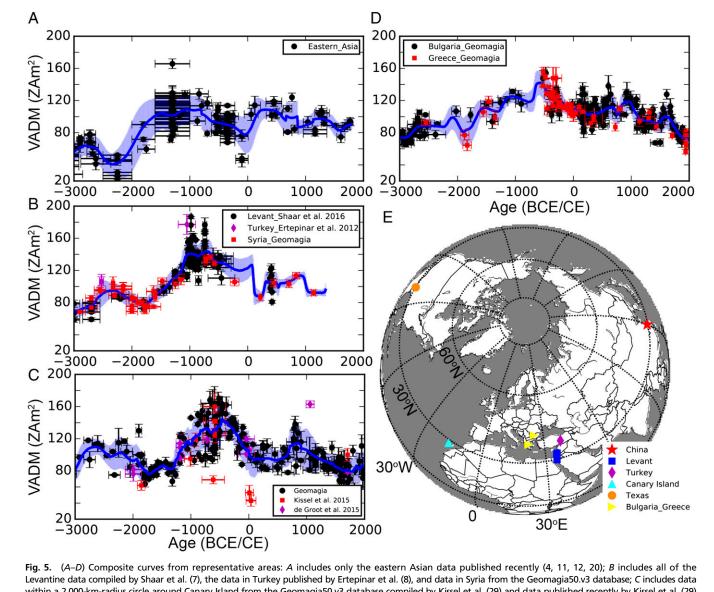
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**Fig. 4.** Histograms of published paleointensity data (blue bars) during the past 10 kyr from GEOMAGIA50.v3 database (39) (A) and during the past 200 Ma (not including data from the recent 10 kyr) from the MagIC database (*B*). Red and yellow stars are the extremely low and high values reported from China. The dashed magenta line in *B* represents median value of the data during the past 200 Ma.

any study anywhere for the Holocene (Fig. 4*A*), which can be either a local geomagnetic anomaly or not captured in other areas. This calls for additional widely distributed data at similar time periods to further characterize their global features and geodynamic mechanisms.

465 In addition to periods of extremely low field intensities, Cai 466 et al. (11) reported a period of possible high field intensity dated 467 around 1300 BCE (±300 y). Here, we obtained additional samples 468 from the same sites and find an even higher value of  $\sim 165.8$  ZAm<sup>2</sup> (Figs. 3B and 5A), which meets the definition of a "spike" sug-q:29 469 gested by Cai et al. (11) of fields in excess of 160 ZAm<sup>2</sup> and is 470 nearly as high as those reported in the Levant around 980 BCE 471 (5–7) and in Turkey at ~1050 BCE (8) (Fig. 5B), but the median 472 age is some 300 y earlier (although there is a large uncertainty in 473 the age of  $\sim 300$  y). The high value recorded by our data could 474 represent a spike around 1300 BCE in China, which might be a Q:30 475 precursor to those recorded in the Levant and Turkey. A high 476 intensity of  $\sim 160 \text{ ZAm}^2$  was reported by de Groot et al. (27) in 477 Canary Islands, whose age is 1058 CE based on radiocar-478 bon dating (28) and could alternatively be ~400-300 BCE con-479 strained by the variation curve of the geomagnetic direction. 480 Under the latter scenario, they suggested a westward motion of 481 the Levantine spikes. In addition, Kissel et al. (29) reported high 482 intensities with VADMs over 160 ZAm<sup>2</sup> in Gran Canaria and locations nearby (e.g., Portugal, Spain, and the Azores) between 483 670 BCE and 400 BCE (Fig. 5C). It seems our data support this Q:31 484 speculation that the spike first appeared in China at ~1300 BCE 485 and then migrated westward to the Levant at ~1000 BCE and 486 finally to Europe at ~670–400 BCE (Fig. 5E). However, the age 487 errors of the Chinese spike overlap with the Levantine spike and 488 could instead represent the same event. Under this interpreta-489 tion, the spike intensity recorded by our data extends the spatial Q:32 490 distribution of the spike reported in Levantine area and Turkey 491 to Eastern Asia. It is interesting to note that Bourne et al. (9) 492 reported a possible spike in sediments at ~3 ka in Texas, im-493 plying that the spike could be a global feature or that there are at 494 least two large flux lobes simultaneously (Fig. 5E). We note, 495 however, that the spike at ~1000 BCE has so far not been not 496 seen in European [e.g., Bulgaria and Greece (Fig. 5D)], or even



within a 2,000-km-radius circle around Canary Island from the Geomagia50.v3 database compiled by Kissel et al. (29) and data published recently by Kissel et al. (29) and de Groot et al. (27); D includes data in Bulgaria and Greece from the Geomagia50.v3 database. The selection criteria of including at least three specimens and with a SD of mean intensity less than 10% or 5 µT were applied on these data. (E) Projections of the locations related to geomagnetic spikes.

Syrian data (Fig. 5B) at similar period. These imply that the spike is likely not global but probably large scale, which fulfills one of the conditions for generating such a spike in numerical modeling (30). However, the composite curves in Fig. 5 show that the field intensity is overall high around 1000-500 BCE at different areas, indicating a dipolar behavior of the field. Taken together, the different records of the spike suggest that it is probably a nondipolar event superimposed on an already strong dipolar field. This speculation is similar to the deduction by de Groot et al. (31) that strong, short-term intensity perturbations are superimposed on a global trend of dipolar decay over the past 1 kyr. This indicates that the variation of the pattern of the geomagnetic field, at least during the past 3 kyr, is possibly driven by the dipole component on which nondipole components superpose occasionally (13). In any case, further data with large spatial distributions and precise age constraints are necessary for achieving an improved understanding of this extreme behavior of the geomagnetic field. 557<sub>Q:33</sub>

The DIP and spike confirmed by our results suggest a large [eightfold increase if calculating with the low intensity of ~20  $ZAm^2$  reported by Cai et al. (11) and our high value of ~166 ZAm<sup>2</sup>] and rapid (within ~1,000 v) fluctuations of the geomagnetic field during the Holocene. Our extreme intensity values are still striking, even when placed in a context of the past q:34 200 Ma. The low values of our data (red stars in Fig. 4B) fall into low end of intensities in the published data and are lower than the median intensity (~54 ZAm<sup>2</sup>) of the past 200 Ma (not including data from the recent 10 kyr). Our low intensities are comparable to the strength of the field during some particular geomagnetic intervals, for example, the Laschamp geomagnetic excursion (32) and the Miocene dipole low (33, 34). Our spike (yellow star in Fig. 4B) is among the highest paleointensities Q:35 recorded over the past 200 Ma. In summary, the extreme behaviors recorded by our data are extraordinary, especially when Q:36 considering the short span of the record, and thus bring challenges to geodynamical modeling (30, 35–38).

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# Supporting Information

#### Cai et al. 10.1073/pnas.1616976114

#### Archaeomagnetic Background and Sampling 0:1

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The samples from Shandong province were collected from 10 different sites covering the prehistoric cultures of early Dawenkou (4300-3500 BCE), Longshan (2500-2000 BCE), and Yueshi (2000-1600 BCE), as well as the historical dynasties of middlelate Shang (1300–1000 BCE), Western Han (206 BCE-8 CE), and Ming (1368–1644 CE). To confirm the unusually low paleointensities discovered by Cai et al. (11), we collected pottery shards from the Liangchengzhen (LCZ) and Zhaojiazhuang (ZJZ) sites. The remaining artifacts from Shandong province were collected from eight sites. The samples from Liaoning province are all from Wenjiacun (WJC) site located in the Lyshun area, Dalian City, which is dated to the middle Dawenkou (~3500 BCE). All of the samples from Shandong and Liaoning were provided by the archaeologists from the School of History and Culture, Shandong University, Jinan, China. The ages of our sites from prehistoric periods and the DXZ site (middle-late Shang) are based on radiocarbon dating (40, 41) as well as archaeological contexts. The ages of other sites were determined mainly based on archaeological background.

The samples from Zhejiang province were collected from part of the published sites in Cai et al. (11), which cover the dynasties of early Tang (618-764 CE), Zhanguo (475-221 BCE), Late Chunqiu (550-476 BCE), and Shang (1600-1000 BCE). Ages of samples from this site are based on typology of the potteries during archaeological survey.

One site from Hebei province is included, which is the Baojiaying (BJY) site located at Longhua county, Chengde City. This site used to be a big kiln factory started during the Liao-Jin dynasty (907-1125 CE) and terminated in Yuan dynasty (1271-1368 CE). A total of 10 cultural layers were excavated on this site. The materials we collected were from the fourth-fifth laver, where some ancient coins from the Yuan dynasty were unearthed. Combing with the typology of the potteries, our samples collected from this site are dated to the Yuan dynasty.

Besides radiocarbon dating (40, 41) and archaeological dating based on archaeological context (e.g., the workmanship and decoration of the potteries, important information preserved on the artifacts such as the ancient Chinese characters, representative artifacts such as ancient corns correspond to special dynasties), we attempted to use the optically stimulated luminescence (OSL) dating method on nine pottery samples (three from Shandong and six from Zhejiang), but none of them gave acceptable results unfortunately. The detailed sampling information is listed in Table S1.

#### Experimental Procedures

Rock Magnetism. To determine the magnetic mineralogy of the studied samples, we carried out detailed rock magnetic experiments including hysteresis loops, isothermal remanent magnetization (IRM) acquisition, first-order reversal curves (FORCs), and variation of magnetization versus temperature (M-T). Powder samples (0.1–0.2 g) were prepared and fixed in nonmagnetic capsules for the hysteresis loops, IRM acquisition, and FORCs experiment, which were measured with the MicroMag 3900 VSM in the Paleomagnetism and Geochronology Laboratory (PGL) at Institute of Geology and Geophysics, Chinese 58 **Q:4** Academy of Sciences (IGGCAS). M-T curves were measured with the magnetic-measurements variable-field translation balance fixed with an oven in PGL for the purpose of determining Curie temperatures ( $T_c$  values) and detecting possible alteration during heating. Microprobe slices (thickness of 30-50 µm) of selected samples were prepared and observed under scanning electron microscope (SEM). The slices were coated with a carbon layer to prevent surface charging of the sample during SEM operation. The SEM experiments were performed with FEI Nova nano450 at 15-kV acceleration voltages in IGGCAS. The imaging and energy dispersive X-ray spectroscopy (EDS) analyses were conducted under the backscattered electron (BSE) mode.

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Paleointensity. A total of 407 specimens from 72 samples (55, 6, 6, and 5 from Shandong, Liaoning, Zhejing, and Hebei, respectively) were processed for paleointensity experiments with a minimum of five specimens per sample. Among all of the specimens, 376 were processed in the PGL, whereas the other 31 were measured in the paleomagnetism laboratory of the Scripps Institution of Oceanography (SIO), University of California, San Diego. For experimental procedure in the PGL, samples were first broken into irregular chips. Then fresh specimens were selected and fixed in cubic ceramic boxes ( $1.2 \text{ cm} \times 1.2 \text{ cm} \times 1.2 \text{ cm}$ ), which have comparable magnetic moments to the background of the magnetometer used for the measurements, with fire-resistant cotton matting. The specimens were heated in a French paleointensity furnace in an argon atmosphere and cooled naturally after heating (~12 h). Heating steps were carried out from 100 to 580 °C with temperature intervals varying from 20 to 100 °C. The residual field of the oven is less than 10 nT for the "zero-field" cooling steps and a laboratory field of 30  $\mu$ T was applied along -z of the specimens with a precision of 0.1  $\mu T$  for the "in-field" cooling steps. The remanence was measured with the 2G 760 SQUID magnetometer after each step. The whole procedure of the experiment was conducted in a shielded room with residual field less than 300 nT.

In the SIO laboratory, fresh specimens were fixed in 12-mmdiameter glass tubes with glass microfiber paper and potassiumsilicate glue (KASIL). Specimens were heated in the laboratorybuilt paleointensity oven equipped with a fan for cooling. The cooling times to room temperature are about 30-45 min depending on the peak temperature. The residual field of the ovens is less than 10 nT during zero-field steps. Measurements were made on a 2G cryogenic magnetometer. The same heating steps as those in PGL were followed. A laboratory field is applied along -z axis of the specimens, and field value of 30 or 50 µT was chosen depending on the expected ancient field of samples. All of the experiments were conducted in the paleomagnetic shielded room at SIO.

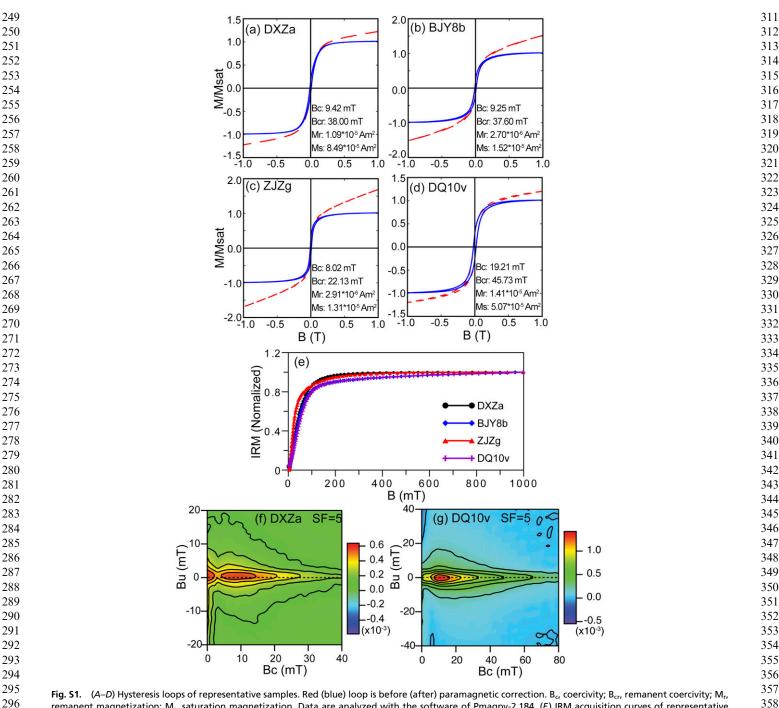
The "IZZI" protocol was followed for all of the specimens during the paleointensity experiment (14). Checks for alteration [partial thermal remanent magnetization (pTRM) checks] were inserted at every other step (15). The bias caused by the anisotropy of TRM (ATRM) in paleointensity for archaeological materials can reach ~10% or more, whereas that caused by cooling rate effect is around 5% with a few exceptions of 15% (11, 12). Therefore, both ATRM and cooling rate effects should be considered when determining the paleointensity values recorded by archaeological artifacts. In this study, the ATRM correction was conducted on each successful specimen in the intensity experiment. An eight-step (baseline, +x, -x, +y, -y, +z, -z, +x) experiment was conducted following the method of Veitch et al. (16), whereby the last step is an alteration check. A total TRM was used for calculation of the anisotropy tensors. Cooling rate corrections were conducted on those specimens heated in the paleointensity ovens in SIO but not on those processed in the French furnace in PGL because the natural cooling system of the latter mimics the original cooling of the pottery when first fired. The cooling rate correction experiment follows the procedure suggested by Genevey and Gallet (17), including three steps: fast cooling, slow cooling, and a second fast cooling as an alteration check. The slow cooling step takes ~12 h in the SIO ovens. Detailed experimental steps can be found in Cai et al. (11). 

#### **Rock Magnetic Results**

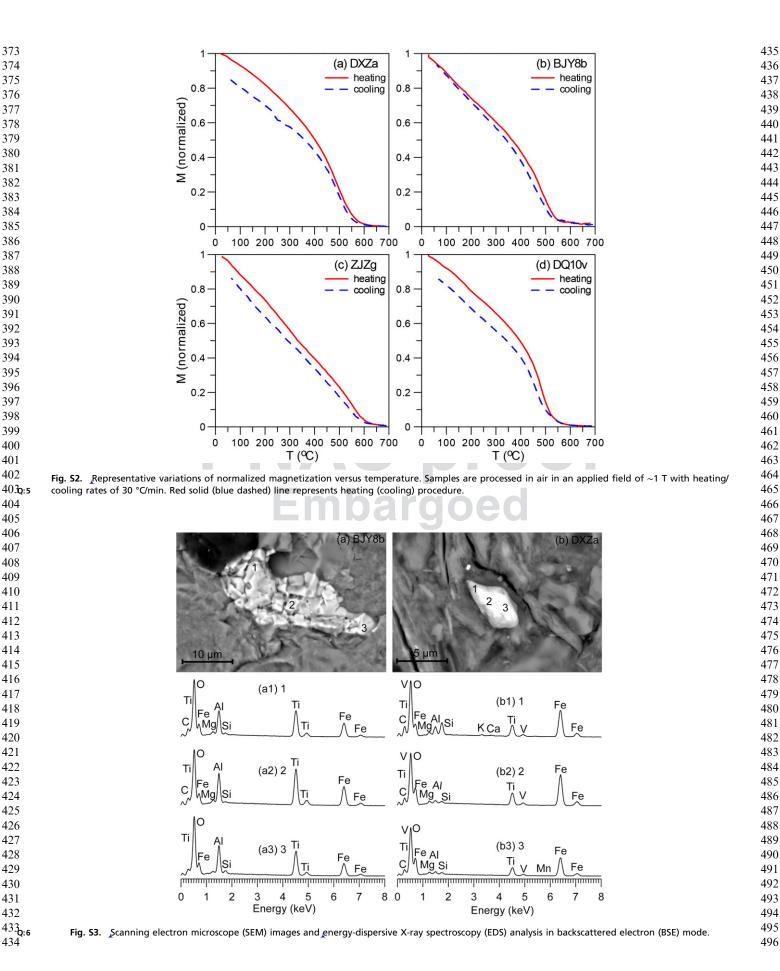
The representative hysteresis loops show slightly "wasp-waisted" shapes (Fig. S1 B and C), suggesting a mixing of either SD and SP grain sizes (42) or varied magnetic minerals (e.g., magnetite and hematite). However, the hysteresis loops of the latter are usually supposed to show "goose-necked" shapes (42, 43), which are absent in our results. This allows us to infer that, even if some of our samples contain hematite, the amount must be limited, which is supported by the following rock magnetic re-sults (Figs. S1E and S2) and NRM demagnetization during paleointensity experiments (Fig. 2). The bulk coercivities (B<sub>c</sub> values) of the specimens are generally low, with a range of  $\sim 10$ to  $\sim 20$  mT, which demonstrates that soft magnetic minerals are the dominant magnetic carriers. The IRM acquisition curves (Fig. S1E) either saturate before 200 mT (DXZa and ZJZg) or increase significantly before 200 mT and keep slightly increasing until 800 mT (BJY8b and DQ10v), which supports the inference of soft magnetic carriers to be dominant. The shapes of FORCs (Fig. S1 F and G) indicate the existence of SD fraction, probably mixed with SP grains (44, 45).

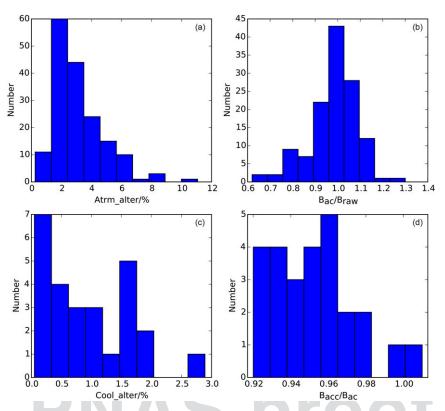
The representative M-T curves show good reversibility (Fig. S2), which indicate that the magnetic carriers are thermally stable. T<sub>c</sub> values calculated by the second derivative method described by Tauxe et al. (43) are ~580 °C (ZJZg) or lower (BJY8b, ~540 °C; DXZa, ~570 °C; DQ10v, ~515 °C). The former demonstrates the existence of magnetite, whereas the latter is usually explained as titanomagnetite in most of the previous studies (11, 12, 46, 47). However, we put forward another possibility for those with T<sub>c</sub> values less than 580 °C in this study. It was demonstrated that not only titanium (Ti) but also aluminum (Al) substitution can reduce the T<sub>c</sub> values of magnetic minerals (48). The SEM images and elemental spectra show the presence of both Ti and Al elements (Fig. S3), indicating both Ti- and Al-substituted magnetic minerals are possible in archaeological materials made of clay minerals. This, combined with the M-T results, suggests that magnetite, Ti-magnetite, and/ or Al-magnetite are the dominant magnetic carriers.

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remanent magnetization; Ms, saturation magnetization. Data are analyzed with the software of Pmagpy-2.184. (E) IRM acquisition curves of representative samples. (F and G) FORC plots analyzed with the software of FORCinel\_1.17 (49).





**Fig. S4.** Histograms of (*A*) alteration percentage during the TRM anisotropy correction experiment, which is the maximum difference (expressed as percentage) of the four pairs (+x/-x, +y/-y, +z/-z, and the two +x steps) of TRM; (*B*) extent of TRM anisotropy correction described by the ratio of intensity value after anisotropy correction (B<sub>ac</sub>) to the raw intensity (B<sub>raw</sub>); (*C*) alteration percentage during cooling rate correction experiment, which is the difference between two fast cooling steps; and (*D*) cooling rate correction factors expressed as the ratio of intensity after both anisotropy and cooling rate correction (B<sub>ac</sub>) to B<sub>ac</sub>.

**Other Supporting Information Files** 

Table S1 (DOCX) Table S2 (DOCX) Table S3 (DOCX) Table S4 (DOCX) Table S5 (PDF)

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