# Hanzhong bronzes and the highly radiogenic lead in Shang period China

3 Kunlong Chen<sup>1,2</sup>, Jianjun Mei<sup>1,3</sup>, Thilo Rehren<sup>2,4</sup>, Siran Liu<sup>1</sup>, Wei Yang<sup>5</sup>,

- 4 Marcos Martinón-Torres<sup>6</sup>, Congcang Zhao<sup>7</sup>, Yoshimitsu Hirao<sup>8</sup>, Jianli Chen<sup>9</sup>, Yu Liu<sup>10</sup>
- 5
  6 1. Institute of Historical Metallurgy and Materials, University of Science and Technology
  7 Beijing, Beijing, 100083, China
- 8 2. UCL Institute of Archaeology, London, WC1H 0PY, UK
- 9 3. Needham Research Institute, Cambridge, CB3 9AF, UK
- 4. Science and Technology in Archaeology and Culture Research Center, The Cyprus
   Institute, Nicosia, Cyprus
- 12 5. Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing, 100029, China
- 13 6. Department of Archaeology, University of Cambridge, Cambridge, CB2 3ER, UK
- 14 7. School of Cultural Heritage, Northwest University, Xi'an, 710069, China
- 15 8. Faculty of Humanities, Beppu University, Beppu, 874-8501, Japan
- 16 9. School of Archaeology and Museology, Peking University, Beijing, 100080, China
- 17 10. Institute of Archaeology, Chinese Academy of Social Sciences, Beijing 100710, China

# 18 Abstract

19 For decades, the origin of the bronzes with distinct highly radiogenic lead isotopic 20 ratios in Shang period China has constituted a research puzzle. This paper presents 21 new lead isotope data of bronze objects from Hanzhong, representing one of the key regional bronze cultures during Shang period China. On the basis of a synthetical 22 23 investigation of the typological, chemical and lead isotopic features of Hanzhong 24 bronzes and their relations to other regional bronze cultures, we propose the Qinling area as the potential region of origin for the metals containing highly radiogenic lead 25 used by several contemporaneous but culturally/ politically distinct entities across a 26 27 vast territory. Taking into account both archaeological and geological evidence, this 28 working hypothesis is expected to draw attention not only merely to the geological 29 provenance of metal resources but also to the mechanisms of metal production and

30 circulation as well as broader social-economic dynamics.

# 31 Keywords:

Lead isotopes; provenance; Shang; China; bronze; highly radiogenic lead; regional
specialisation; multiple-direction exchange.

# 34 Highlights:

- New lead isotope results from a key regional bronze culture of Shang period
  China.
- The Qinling region is proposed as a source of the mysterious highly radiogenic
  lead in Shang period bronzes.
- Regional specialisation and a multiple-direction interregional exchange are
  shown in Bronze Age China
- A new understanding of ancient metallurgy in broader social-economic contexts
  is offered.

# 43 **1. Introduction**

44 The production of splendidly cast bronzes, one of the most prominent features

45 characterising China's early civilisations (Chang, 1986: 365-7) reached its zenith

46 during the middle and late Shang period (ca. 1300-1046 BCE). To support the large-

- scale production of ritually important vessels, weapons and other implements, copper,
- tin and lead had to be exploited in ore-rich mountainous regions, remote from the

49 centres of Shang civilisation, and transported to the Central Plain in the lower reaches

50 of the Yellow River (Yue and Liu, 2006), the core and metropolitan region of the

51 Shang polity (Figure 1). The circulation of these key resources and products must

52 have resulted in complex trans-cultural inter-regional interactions, which in turn

53 played a significant role in shaping the developmental trajectory of the early states in

54 Bronze Age China (Liu and Chen, 2012: 369-81).

55 Seeking to identify the geological origins and supply routes of raw metals and the

56 circulation patterns of the finished bronzes, chemical and/or isotopic analyses have

57 therefore been a core topic of archaeometallurgical research in China just as in many

other regions in the world (e.g. Peng, et al., 1999, Jin, 2008, see also Pollard, et al.,

59 2015, Radivojević, et al., 2018 for most recent reviews on methodology). However,

60 the distinctive highly radiogenic lead (HRL) isotopic composition of many Shang

61 period bronzes has puzzled scholars from various disciplinary backgrounds and

become a long-standing issue in Chinese archaeology since it was first identified in

the 1980s. After looking at the data collected by the authors, Ernst Pernicka (2013,

64 personal communication) even joked that archaeological scientists would have given

up the lead isotope method if they had started with the Shang period bronzes.

66 Over the last few years, another round of discussion on this question has attracted

67 interest from a broad academic community (e.g. Sun, et al., 2016, Jin, et al., 2017,

Liu, et al., 2018a, Liu, et al., 2018b). While the mysterious HRL metal and its

69 geological origin still sit in the heart of the debate, we begin to see a promising new

70 tendency paying more attention to general archaeological contexts and questions (Jin,

et al., 2017, Liu, et al., 2018b). In this paper we report new analytical data, briefly
review previous studies, propose a potential geographic region of origin for the HRL
and discuss the implications of this new model, thus moving towards a new

- vunderstanding of bronze metallurgy and its social-economic context in Bronze Age
- 75 China.

# 76 2. The Hanzhong bronzes

The Hanzhong basin, situated in southwestern Shaanxi, is located on the upper 77 reaches of the Han River, surrounded by the Qinling mountain range to the north and 78 the Daba-Micang Mountains to the south. The basin has geographic connections 79 reaching southwest to the Chengdu Plain, north to the Wei River valley and south to 80 the Middle Yangtze region, making it an important centre for communication 81 82 networks between various parts of China (Figure 1). Since the 1950s, dozens of 83 ancient bronze hoards or caches have been found around the confluence of the Han and Xushui Rivers (e.g. Duan, 1963, Tang et al., 1980, Chai et al. 2005), forming the 84 so-called Hanzhong bronze group, also known as the 'Chengyang bronze group', after 85 the two counties of Chenggu and Yangxian in the eastern part of the basin (Zhao, 86 87 1996). The total assemblage of Hanzhong bronzes comprises more than 700 objects from 33 caches at 19 different sites. The bigger caches/hoards such as Longtou, Sucun 88 and Machang (Chen et al., 2016: online supplement 1) often contain stylistically 89 mixed but chronologically similar assemblages of bronze artefacts (Zhao, 2006, Sun, 90 2011). The whole bronze assemblage includes hundreds of weapons and Yang discs, 91 92 dozens of ritual vessels, masks, Zhang sceptres, sickle-shaped objects, and a few tools and ornaments (Figure 2) (Cao, 2006, Zhao, 2006). As a group, these bronzes, often 93 found accidentally by farmers, have been assigned to the "Baoshan Culture", which 94 takes its name from the only properly excavated site in the region, contemporary with 95 96 the Shang culture of the Central Plain (XDWX, 2002: 176-9). Typological studies 97 suggest an approximate date ranging from the Upper Erligang to the late Anyang (Yinxu) periods, spanning from the fourteenth to the eleventh centuries BCE (Cao, 98 99 2006, Zhao 2006), although some pieces in the group may be dated to the Western Zhou period (e.g. Li 2007, von Falkenhausen, 2011). 100

Previous research has revealed two notable features of the Hanzhong bronzes. On the
one hand, the whole assemblage shows remarkable stylistical/typological diversity,
comprising various groups of products that are typologically attributable to distinct
regions, including the Shang metropolitan areas in the Central Plain (Figure 2: Subgroup A), the Wei River valley in the north of the Qinling Mountains (Figure 2: Subgroup B), and the middle-lower Yangtze River Valley (Figure 2: Sub-

- 107 Cao, 2006, Chen, 2010a: 117-23, von Falkenhausen, 2011, Chen, et al., 2016). On the
- other hand, the close correlation among the indigenous archaeological culture, 108
- stylistically local items (sickle-shaped objects, Zhang sceptres and socketed axes, 109
- Figure 2: Sub-group D) and their characteristic metal compositions (mainly unalloyed 110
- copper and the "natural alloys" containing arsenic, antimony and nickel), suggests the 111
- 112 existence of indigenous metalwork in the Hanzhong region during the Shang period
- (Chen, et al., 2009, Chen, 2010a: 104-9, Chen, et al., 2016). 113
- 114 Jin, et al. (2006) reported lead isotopic results of 31 Hanzhong bronze objects and
- suggested the region as a foothold in the "northern route" through which the Central 115
- Plain polities contacted those in the Sichuan Basin, which is relevant to the 116
- 117 exploitation of metallic resources in the regions further southwest, the bordering area
- 118 of Yunnan, Guizhou and Sichuan provinces.

#### 119 3. Materials, methods and analytical results

- This paper presents new lead isotope data of 84 samples from 72 objects among the 120
- 121 Hanzhong bronzes. The objects sampled cover a good range of various typologies,
- chronology and aforementioned sub-groups, and therefore are thought to be 122
- representative of the entire group (Figure 2). The lead isotope composition of our 123
- 124 samples was analysed utilizing a Thermo Electron Corporation MAT262 Surface
- Ionization Mass Spectrometer (TIMS) at Beppu University in Japan. 125
- The results are presented in Table S1 in the Supplementary materials as three isotopic 126 ratios with <sup>204</sup>Pb as the denominator, together with the methods of sample preparation
- 127
- and measurement. The table also shows lead isotope data published by Jin, et al. 128
- 129 (2006), lead concentrations, and the alloy types following the conventional threshold
- 130 of 2wt% for alloys classification. Detailed results and description of chemical
- compositional analysis can be found in Chen, et al. (2009), Chen (2010a) and Chen, et 131
- al. (2016: supplementary material S2). In total, lead isotopic data of 115 samples of 132
- 104 bronze objects from Hanzhong are now available for discussion, with 87 of them 133
- 134 being complemented with their elemental composition.
- The lead isotope compositions of the Hanzhong bronzes span a broad range, from 135
- 17.531 to 23.853 for <sup>206</sup>Pb/<sup>204</sup>Pb, 15.452 to 16.507 for <sup>207</sup>Pb/<sup>204</sup>Pb and 38.104 to 136
- 44.681 for <sup>208</sup>Pb/<sup>204</sup>Pb. Among them, more than three quarters of the samples (88 of 137
- 115) are highly radiogenic, defined here as  ${}^{206}Pb/{}^{204}Pb > 20$  and  ${}^{208}Pb/{}^{204}Pb > 40$  (Jin, 138
- 2008: 292-302, Liu, et al., 2018b). Though there is concern that possible 139
- 140 contamination from the burial environment may alter the lead isotope composition of
- the patina from base metal with low lead content (e.g. Snoek, et al., 1999, Gale and 141

Stos-Gale, 2000), the non-clustered distribution of the lead isotope ratios and the good
agreement with published data suggest lead contamination from the burial
environment was negligible. Therefore, the results are considered robust and taken to
represent the base metal of the objects.

Lead concentration is a major concern when applying lead isotope analysis for the 146 147 provenance of copper-based metals in antiquity (Gale and Stos-Gale, 1982). While the 148 contribution of lead from tin and its influence on the lead isotope composition are generally negligible (Gale and Stos-Gale, 2000, Molofsky, 2009), it is crucial to 149 150 ascertain whether the measured isotopes from one specific object are derived from traces of lead within the copper, or from the lead added during the alloying process 151 152 (Gale and Stos-Gale, 1982, Pollard and Bray, 2015). The issue appears even more 153 significant in Bronze Age China, given the prevalence of leaded tin bronze (taken here as  $Pb \ge 2 \text{ wt\%}$ ) among hundreds of chemically analysed objects. Although it is 154 complicated to decide how much lead in copper-based alloys can signify deliberate 155 addition of lead, several scholars have tentatively suggested that a concentration of 1 156 157 wt% Pb is sufficient to indicate addition of lead, and for the isotope signature being dominated by the lead source (Zhu and Chang, 2002, Jin, 2008: 41, Liu, et al., 158 159 2018b).

Figure 3:a plots a lead isotope ratio against inverse lead chemical concentration as 160 recently proposed by Pollard and Bray (2015). Theoretically, the mixing lines of two 161 components would become linear in the chart and can be used to illustrate the 162 163 controlling component (copper or lead in this case) of the isotope data. We also plot the data with two isotope ratios grouped by different lead contents for comparison 164 (Figure 3:b). There is no clear linear correlation between 1/Pb (1000 ppm<sup>-1</sup>) and the 165 lead isotope ratio (<sup>206</sup>Pb/<sup>204</sup>Pb) (Figure 3:a), as many of the plots are horizontally 166 squeezed in a very narrow area around  $1/Pb \approx 0$  due to their relatively high lead 167 168 contents but vertically scattered over a broad range of lead isotope values. Figure 3:b is also characterised by the substantial overlap of the isotope ratios despite their 169 various lead concentrations. These patterns, revealed by both isotopic and elemental 170 compositions, suggest that the copper (indicated by the low lead samples) and lead (as 171 shown by the high lead ones) used to cast most of the Hanzhong bronzes have similar 172 HRL isotope signature, although it is interesting to see that a considerable number of 173 lead-rich samples (Pb  $\geq$  2%) seem to be more radiogenic than the rest (<sup>206</sup>Pb/<sup>204</sup>Pb > 174 23). 175

As mentioned before, typological diagnosis and chemical analyses have differentiatedfour sub-groups among the assemblage of Hanzhong bronzes, implying their diverse

178 origins from distinct regional bronze cultures (e.g. von Falkenhausen, 2011, Chen, et al., 2016). When the data are classified by these sub-groups and plotted in Figure 4, it 179 is surprising to see that lead isotopic composition of samples from different sub-180 groups again substantially overlap with each other and are hardly distinguishable. 181 That is to say, even though objects assigned to distinct sub-groups were most likely 182 183 fabricated in various regions and cultural/political contexts, the raw metals used 184 (copper and/or lead), especially the ones that have HRL isotope composition, seem to have originated from a common source. This observation is very significant for our 185 understanding of metal material sources and their circulation networks. 186

#### 187 4. Discussion

## 188 4.1 The highly radiogenic lead metal in Shang period bronzes

Since the pioneering work of Jin Zhengyao in the early 1980s (Jin, 2004), for decades 189 the provenance of the metals for the HRL Shang bronzes has puzzled researchers from 190 various disciplines. More than 60% of the analysed Shang period bronzes (n>800) 191 192 were found to have distinctive highly radiogenic lead isotopic compositions,  $^{206}$ Pb/ $^{204}$ Pb > 20 and  $^{208}$ Pb/ $^{204}$ Pb > 40, which distinguish them from most known lead 193 deposits and bronze artefacts worldwide (Zhu and Chang, 2002, Jin, 2008, Sun, et al., 194 195 2016, Liu, et al., 2018b). Their occurrence is geographically widespread in a vast area of several million km<sup>2</sup> in China and involves most of the major Shang period regional 196 cultures (Figure 1). The HRL bronzes identified in different regions are hardly 197 differentiated from each other by their lead isotope ratios (Figure 5), despite their 198

remarkable typological/cultural varieties (e.g. Bagley, 1999).

200 Another interesting point is that the chemical compositions of HRL bronzes cover

various copper alloys with distinct lead concentrations. On the one hand, as most of

the HRL bronzes contain a significant amount of lead (> 2 wt%) that dominates lead

isotope ratios of the measured objects, the source should be plumbiferous. On the

other hand, some artefacts with low lead contents (< 1 wt%) and malachite samples

from various sites also show similar lead isotope ratios (Jin, 2008: 39-43), suggesting

that the HRL in the alloy could have derived from copper ore as well. The same

207 uncorrelated patterns between lead concentration and isotope ratios have been

208 observed from our results of Hanzhong bronzes as indicated in Figure 3.

209 It is also important to note that the wide appearance of HRL bronzes is

chronologically limited to approximately 300 years between the Upper Erligang

211 (early-middle Shang period) and the Yinxu Phase III (ca. 1450-1150 BCE) (Figure 6,

Figure 7). Despite forming the majority of Shang period objects, HRL bronzes are

hardly found among bronzes from the pre-Shang Erlitou and the later Zhou periods 213 (Jin, 2008). HRL bronze first appears in early Shang cities at Zhengzhou and Yanshi 214 in Henan (Peng, et al., 1999, Jin, 2008, Tian, 2013), Yuanqu in Shanxi (Cui, et al., 215 2012) and Panlongcheng in the Middle Yangtze River (Peng, et al., 1999), and is 216 subsequently identified in almost every major bronze group dating to the middle-late 217 218 Shang period, such as Anyang (Yinxu) in Henan (Jin, 2008), northern Shanxi and Shaanxi (Cao, 2014, Liu, 2015), Sanxingdui in Sichuan (Jin, et al., 1995) and Xin'gan 219 Dayangzhou in Jiangxi (Jin, et al., 1994). A significant proportion (~60%) of the 220 221 collections of Shang bronzes in the Arthur M. Sackler Museum in Washington D.C. 222 and the Sen-oku Hakuko Kan in Kyoto also have this distinctive isotopic signature (Barnes, et al., 1987, Hirao, et al., 1998). Towards the end of the Shang period, HRL 223 224 bronze rather quickly disappeared except for the continued presence at the site of

Jinsha in Sichuan for around another one (?) hundred years (Jin, et al., 2004).

226 In our view, two important observations can be derived from previous research.

227 Firstly, the fact that the HRL bronzes are relatively tightly circumscribed

chronologically but widespread geographically indicates that it is most likely that a

single source region had provided the HRL metal for many distinct regional bronze

cultures in China during the Shang Period (e.g. Jin, et al., 2017, Liu, et al., 2018b).

231 Secondly, the various lead concentrations of HRL bronzes, from lead-free unalloyed

copper to alloys containing dozens percent of lead, suggest the source would have

supplied both copper and lead during its exploitation (e.g. Jin, 2008: 33-47, Tian,
2013, Liu, et al., 2018b).

Theoretically, it is possible that more than one source of HRL was exploited during 235 the Shang period China, as recently proposed by Liu, et al. (2018a). However, 236 according to the lead isotope data for lead and copper ores from China (Jin, et al., 237 238 Figure 3), radiogenic lead is rather rare and only several mineral deposits including 239 the ones in north-east Yunnan, Qinling and Zhongtiao Mountains are reported to yield metalliferous ores with lead isotopic values as high as Shang bronzes. Considering the 240 rather 'sharp' chronological beginning and end of use of HRL in such a vast 241 geographically and culturally diverse territory, current evidence tends to be more 242 consistent with the asumption of a single source or source region. With more sources, 243 244 each potentially with their own geological, political and economic constraints, we might expect more variability in terms of the chronology. Liu, et al. (2018a) note that 245 HRL signatures are found in some pigments and glass after the Shang period (but so 246 far not in metal objects) and use this as key evidence to argue for multiple sources of 247 HRL for the Shang bronzes. However, there are obvious risks when assuming that the 248 249 parameters affecting the supply of non-metals in the post-Shang period can be so

easily applied to Shang-period metals. Thus, on balance, we favour the hypothesis ofa single source.

Talking about the geographic location of this source, however, becomes difficult and 252 controversial. Jin Zhengyao first proposed that the raw materials for casting HRL 253 bronzes in Yinxu came from Yunnan and developed the hypothesis of the "southwest 254 255 origin" of HRL metal in the following series of papers on the materials from 256 Zhenghzou (Erligang), Sanxingdui and Xin'gan Dayangzhou (e.g. Jin, 2008, Tian, 2013, Jin, et al., 2017). Although the suggestion has been supported by geologists 257 258 (Zhu and Chang, 2002), the lack of archaeological evidence for contacts between Yunnan and central China during the Shang period has raised serious questions from 259 archaeologists. Considering the geographic intermediate position of the Qinling 260 261 Mountains, Saito, et al. (2002) suggested the region as another possible source of HRL but provided no isotopic and archaeological evidence. A number of other 262 regions including Jiangxi, Hunan (Peng, et al., 1999, Zhu and Chang, 2002), the 263 minor Qinling area in Henan, Qingchenzi in Liaoning (Zhu and Chang, 2002) and 264 265 even Africa (Sun, et al., 2016) have been proposed by other scholars, but none of them is so far conclusive and sometimes conflicts with the archaeological evidence 266 and existing analytical results (see also Chen, 2010b, Jin, et al., 2017, Liu, et al., 267 2018b for detailed review). The geological source of the HRL Shang bronzes has 268 269 therefore remained unknown.

### 270 4.2 The implications of the new data from Hanzhong

The lead isotope analysis, together with the systematic typological and technological 271 research presented in this paper throws new light on the discussion of HRL bronzes of 272 Shang period China. As shown in Figure 5, the results of our analysis are consistent 273 with previous studies and the lead isotope ratios of Hanzhong bronzes are hardly 274 differentiable from other major Shang period bronze groups. More importantly, a 275 substantial part of the typologically and chemically distinctive objects such as sickle-276 shaped objects and Zhang sceptres (Sub-group D in Figure 2) are found to be made of 277 HRL metal as well (mostly unalloyed copper, Figure 3, Figure 4). Since it is widely 278 accepted that these objects are characteristic products of the Hanzhong community 279 (e.g. Cao, 2006, Zhao, 2006, Chen, et al., 2009, von Falkenhausen, 2011, Chen, et al., 280 281 2016), a tight interrelationship between Hanzhong's local metallurgical industry and the HRL source is therefore upheld by both archaeological, technological and isotopic 282 evidence. It is interesting to see that all of the four samples from the repairing patches 283 of different vessels (No. HZ029, 102, 105, 116) also have HRL composition, even 284 though one sample from the main (repaired) body (HZ104) has common lead (see 285

Supplementary Table S1). These repair patches are most likely to have been added to
the vessels when they were used in Hanzhong (Chen, 2010a: 97-99) and would further
testify the HRL signature of local metalwork.

The Qinling area, where the Hanzhong Basin is located, is a well-known metallogenic 289 region of multimetallic ore deposit clusters (e.g. Qi and Hou, 2005, Ren, et al., 2007). 290 291 The Mujiazhuang copper deposit in the Zhashan area of the south Qinling Orogenic 292 Belt, about 200 km to the northeast of Hanzhong, had been identified to have highly radiogenic lead isotopic compositions (see Supplementary Table S2 for detailed 293 294 analytical results) (Zhu, et al., 2006). To the south of the basin, the Pb-Zn deposit in Mayuan has been classified as Mississippi Valley Type (MVT), which tends to have 295 296 more radiogenic and varied lead isotope signatures (Liu, et al., 2015), even though the 297 published data for this deposit were not highly radiogenic.

298 The rich mineral resources in the region have long been noticed and used by the ancient communities there. Several ancient mining pits for turquoise and hundreds of 299 hammer stones have recently been discovered in the area centred at the conjunction of 300 the Xiyu and Luo Rivers in Luonan County (Li, et al., 2016). Pottery sherds found in 301 302 the mining pits were assigned to archaeological cultures ranging from the Neolithic to the Late Bronze Age, consistent with the absolute date of 2030-500 BCE obtained 303 from eight radiocarbon dates (samples of bone and charcoal, four of each) (Xian, 304 2016: 76-80). It is suggested that the Luonan region is a potential source of the 305 turquoise industry at Erlitou, the key site of the Early Bronze Age in the Central Plain 306 307 (Xian, et al., 2018). This research provides crucial evidence for the early exploration and exploitation of mineral resources in the Qinling region, even though ancient 308 copper/lead mining and smelting sites in this area are yet to be identified. 309

310 It is also important to point out that the date of Hanzhong bronzes, and the Baoshan

archaeological culture to which they have been assigned, span from the Upper

Erligang to the Yinxu Phase III (Cao, 2006, Zhao, 2006), which is virtually

synchronous with the period when HRL bronzes were widespread (see Figure 7). The

Jinsha site, which has yielded the only later HRL bronze group dated to early Western

Zhou (Jin, et al., 2004), is located in the Sichuan Plain, hence not very far from

Hanzhang and showing a close relationship with the Baoshan culture (XDWX, 2002).

Based on the aforementioned observations, here we propose the Qinling area as a

potential target area to locate the geological and geographical origin of the Shang

319 period HRL bronzes. According to this proposal, polymetallic resources (copper and

lead) exploited by the local communities in Hanzhong would have supplied the raw

321 metals for many bronze industries in different regions during the Shang period in

322 China, as indicated by their shared and highly characteristic lead isotope signature and

the archaeologically evidenced trans-cultural correlations (e.g. von Falkenhausen, 323

- 2011, Chen, et al., 2016). The implications of this new hypothesis are briefly outlined 324 below.
- 325

#### 4.3 Social-economic landscape in Shang period China: a metallurgical perspective 326

The early Shang period, when the HRL bronzes first came into use, witnessed the 327 widespread adoption of bronze metallurgy in China, especially south-eastward 328 (Bagley, 1999, Wang, 2014). The increasing expansion of production scale in the 329 Shang metropolitan areas would have undoubtedly increased demand for raw 330 331 materials such as metals, which subsequently facilitated the interregional connections and established routes of exchange between the ore-rich mountainous areas and the 332 333 regional centres where the raw metals were accumulated and worked into artefacts. It 334 has been pronounced repeatedly by specialists in Bronze Age China archaeology that acquisition of metals for the elites would has been an essential motive for the well-335 336 formulated "Erligang Expansion" during the early Shang period (e.g. Bagley, 1999, Wang, 2014, Liu and Chen, 2003: 131-45). 337

Demonstrated as a crucial nodal point of the exchange network since the Erligang 338 339 period, the local communities of Hanzhong would have been stimulated by the external influence and reacted to it by taking advantage of the natural landscapes and 340 resources (Chen, et al., 2016). As von Falkenhausen (2011: 435) has insightfully 341 pointed out, "whatever bronze manufacturing went on in the upper Han River Basin 342 should be viewed in conjunction with the exploitation of the copper-ore resources of 343 the Qinling Mountains". Considering the ore-rich geology and ample fuel supplies 344 from the mountainous area, and the relatively underdeveloped state of agriculture as 345 evidenced by the finds from the Baoshan site (XDWX, 2002: 180), it would have 346 been a rational choice for this region to specialise in the primary production of metals 347 (mining and smelting) and to exchange their metals with cultures in other regions 348 which excelled at other productive activities. This kind of dynamics of interaction has 349 been considered to have occurred regularly in Bronze Age China, as illustrated by the 350 production and exchange of salt (von Falkenhausen, 2006, Liu and Chen, 2012: 273-351 90). The intermediate geographic position of the Qingling Mountains and navigable 352 353 water routes along the Han River would have been crucial in promoting the proposed exchange. 354

During the Yinxu period, the Hanzhong region continuously participated in a trans-355 356 cultural interaction network connecting many regions. While the control of the Shang state seemed to retreat to Anyang and surrounding areas (Li, 1990), reginal bronze 357

358 industries across the vast territory in south China prospered, as exemplified by the spectacular and distinctive artefacts from Jiangxi, Sichuan and Hunan (Kane, 1974, 359 Bagley, 1999, Xu, 2008, Steinke, 2014). With the engagement of these newly 360 established regional centres, the simplified "core-periphery" tributary model (Liu and 361 Chen, 2003: 133-40), even if it was the case during the early Shang period, must have 362 363 given way to a more complex multiple-direction exchange network. Importing copper and lead from specialised mining/smelting communities in Hanzhong through the 364 existing connection routes would have been more cost-effective than producing them 365 locally, even for some regions where ores were available. The increasing interactions 366 with other regional bronze cultures in turn explain why bronzes of various styles and 367 manufacture origins (secondary production) were gathered in Hanzhong (Li, 2007, 368 Chen, 2010a, von Falkenhausen, 2011, Chen, et al., 2016). This pattern of economic 369 specialisation (i.e. production for exchange and import across cultural and geographic 370 boundaries) is predicted by Ricardo's Law of Comparative Advantage, and has been 371 372 demonstrated previously for copper production and exchange in Bronze Age Alps (Shennan, 1999). 373

It is also worth noting that the decrease of HRL bronzes in the Central Plain coincided 374 with the rise of the Zhou in the Wei River valley towards the late Yinxu period. The 375 dramatic change of the political landscape would have undoubtedly affected and 376 377 potentially limited the multiple-direction exchange network engaged with many regional powers of various political standings. Several scholars have already 378 correlated the sudden disappearance of bronzes in Hanzhong with the conquest of 379 380 Shang, although whether the local communities were allies of the Zhou is still controversial (e.g. Li, 1997, von Falkenhausen, 2011, Sun, 2011). 381

# 382 **5. Concluding remarks**

Until direct evidence of metallurgical production, such as mining pits, furnaces, slag 383 and other technological remains dated to the Shang period has been identified in the 384 region, the proposed Qinling area as an origin of the Shang period HRL bronzes 385 should remain as a working hypothesis. However, the significance of this paper is not 386 only merely in demarcating the potential geological ore source but, more importantly, 387 in aligning the archaeological and scientific data into a coherent historical narrative. 388 Even though we are unable to confirm the exact geographical/geological origin of the 389 metal with the evidence available, the proposed model of a single specialised source 390 supplying a number of culturally distinctive regional bronze industries itself is 391 sufficiently intriguing and meaningful for furthering our understanding of Shang 392 period China. Instead of being just an unsolved mystery pertaining to the exact 393

geographical/geological origin of HRL, the subject is shown here to be an excellent
case in point for the discussion of regional relationships and the wider socialeconomic landscape in Shang period China.

While the provenance of ancient artefacts and the location of primary production remains will undoubtedly continue to be one of the primary goals of our research, cautious research has to continue to explore the archaeological/historical dynamics outlined by the existing evidence. Solid cooperative relationships among researchers from diverse backgrounds are undoubtedly crucial for further work on bronze metallurgy in Shang China, given the interdisciplinary nature of archaeological research.

# 404 Acknowledgements

405 The authors are grateful to Fulin Chai, Quntao Zhai, Kyouhei Nishida, Shoji

406 Yamaguchi and Takuma Ishida for their support in the sampling and lead isotope

407 analyses. We also thank Professors Zhengyao Jin, Jessica Rawson, Mark Pollard,

408 Ernst Pernicka, David Killick and other colleagues for sharing their knowledge. This

research received support from the National Natural Science Foundation of China

410 (51304020), Administration of Cultural Heritage of China (2014220, 20080210) and

the Newton International Fellowship awarded by the British Academy (NIF160456).

412 Comments from two anonymous reviewers and the associated editor have helped

413 strengthen our arguments and are gratefully acknowledged.

#### 414 **References**

- Bagley, R., 1999. Shang archaeology, in: Loewe, M., Shaughnessy, E.L. (Eds.), The Cambridge History
  of ancient China: From the origins of civilization to 221 BC, Cambridge University Press, Cambridge
  pp. 124-231.
- Barnes, I.L., Chase, W., Deal, E., 1987. Appendix 2. Lead Isotope Ratios, Ancient Chinese Bronzes in
  the Arthur M. Sackler Collections. Volume 1. Robert W. Bagley, Shang Ritual Bronzes in the Arthur M.
  Sackler Collections, Harvard University Press, pp. 558-560.
- 421 Cao, D., 2014. The Loess Highland in a Trading Network (1300-1050 BC), Princeton University.
- 422 Cao, W., 2006. Hanzhong chutu Shangdai qingtongqi (Shang Dynasty bronzes unearthed from
  423 Hanzhong), Bashu Press, Chendu. (in Chinese)
- 424 Chai, F., He, T., Gong, C., 2005. Shanxisheng Chengguxian xinchu shangdai tongqi (New discovery of
  425 Shang Dynasty bronzes from Chenggu County, Shaanxi Province). Kaogu yu Wenwu (Archaeology and
  426 Cultural Relics), no.6, 3-4. (in Chinese)
- 427 Chang, K.-c., 1986. The archaeology of ancient China, Yale University Press, New Haven, Conn.
- 428 Chen, K., 2010a. Shaanxi Hanzhong chutu Shangdai tongqi de kexue fenxi yu zhizuo jishu yanjiu
  429 (Scientific Study on the Shang Dynasty Bronzes Unearthed from Hanzhong, Shaanxi Province: Materials
  430 and Manufacturing Techniques), University of Science and Technology Beijing, Beijing. (in Chinese)
- Chen, K., Mei, J., Rehren, Th., Zhao, C., 2016. Indigenous production and interregional exchange: late
  second-millennium BC bronzes from the Hanzhong basin, China, Antiquity 90, 665-678.
- Chen, K.L., Rehren, Th., Mei, J.J., Zhao, C.C., 2009. Special alloys from remote frontiers of the Shang
  Kingdom: scientific study of the Hanzhong bronzes from southwest Shaanxi, China, J Archaeol Sci 36,
  2108-2118.
- Chen, K.-t., 2010b. Qian tongweisu laiyuandi fenxi: Shangdai tongqi zhuzao yuanliao laiyuan yanjiu
  pingyi (Comments on provenance studies of Shang bronze casting raw materials with lead isotope
  analysis) in: Li, Y.-t. (Ed.), Jinian Yinxu fajue bashi zhounian xueshu taolunhui (Conference
  commemorating the 80th anniversary of archaeological excavation in Yinxu), Institute of history and
  philology, Academia Sinica, Taipei, pp. 113-139. (in Chinese)
- Cui, J., Tong, W., Wu, X., 2012. Yuanqu shangcheng chutu bufen tong lianzha ji tongqi de qian tongweisu bizhi fenxi yanjiu (Study on the lead isotope analysis of bronze, slags and wares unearthed from the Yuanqu Shang City), Wenwu (Cultural Relics), no.7, 80-84. (in Chinese)
- 444 Duan, S., 1963. Jieshao Shanxisheng bowuguan de jijian qingtongqi (Introduction of several
  445 bronzeobjects of Shaanxi Provincial Museum). Wenwu (Cultural Relics), no.3, 43–44. (in Chinese)
- Gale, N.H., Stos-Gale, Z., 2000. Lead isotope analyses applied to provenance studies, in: Ciliberto, E.,
  Spoto, G. (Eds.), Modern analytical Methods in Art and Archaeology, Wiley, New York, Chichester, pp.
  503-584.
- Gale, N.H., Stos-Gale, Z.A., 1982. Bronze Age copper sources in the Mediterranean: a new approach,
  Science 216, 11-19.
- Hirao, Y., Suzuki, H., Hayakawa, Y., 1998. Lead isotopic ratios of ancient Chinese bronze in the
  collection of the Sen-oku Hakuko Kan, Journal of Sen-oku Hakuko Kan, 25-46.
- Jin, Z., 2004. Zhongguo xuezhe de shoupian qian tongweisu kaogu yanjiu lunwen (On the first lead
  isotopic paper in archaeology by Chinese scholar), Wenwu Baohu yu Kaogu Kexue (Sciences of
  Conservation and Archaeology) 16, 64-66. (in Chinese)

- Jin, Z., 2008. Zhongguo qian tongweisu kaogu (Lead Isotope Archaeology in China), University of
   Science and Technology China Press Hefei. (in Chinese)
- Jin, Z., Chase, T., Hirao, Y., Peng, S., Mabuchi, H., Miwa, K., Zhan, K., 1994. Jianxi Xin'gan
  Dayangzhou Shangmu qingtongqi de qian tongweisu bizhi yanjiu (The study of lead isotope ratios of
  bronze artefacts from Dayangzhou cemetry at Xin'gan in Jiangxi province), Kaogu (Archaeology), no.8,
  744-747. (in Chinese)
- Jin, Z., Liu, R., Rawson, J., Pollard, A.M., 2017. Revisiting lead isotope data in Shang and Western Zhou
  bronzes, Antiquity 91, 1574-1587.
- Jin, Z., Mabuchi, H., Chase, T., Chen, D., Miwa, K., Hirao, Y., 1995. Guanhan Sanxingdui yiwukeng qingtongqi de qian tongweisu bizhi yanjiu (The study of lead isotope ratios of bronze artefacts from the hoard at the site of Sanxingdui in Guanghan ), Wenwu (Cultural Relics), no.2, 80-85. (in Chinese)

Jin, Z., Zhao, C., Chen, F., Zhu, B., Chang, X., Wang, X., 2006. Baoshan yizhi he Chengyang bufen
tongqi de qian tongweisu zucheng yu xiangguan wenti (Lead isotopes of the bronzes from Baoshan site
and Chengyang bronzes and its related issues), in: Zhao, C. (Ed.), Chengyang Qingtongqi, Cultural Relics
Press, Beijing, pp. 250-259. (in Chinese)

- Jin, Z., Zhu, B., Chang, X., Xu, Z., Zhang, Q., Tang, F., 2004. Chengdu Jinsha yizhi tongqi yanjiu
  (Research on the bronzes from the Jinshan site in Chengdu), Wenwu (Cultural Relics), no.7, 76-88. (in
  Chinese)
- 474 Kane, V.C., 1974. The independent bronze industries in the South of China contemporary with the Shang475 and Western Chou Dynasties, Archives of Asian Art 28, 77-107.
- 476 Li, B., 1990. Zhongguo qingtong wenhua de fazhan jieduan yu fenqu xitong (On the development phases
  477 and regional division of bronze cultures of China), Huaxia Kaogu (Huaxia Archaeology), no.2, 82-91.
  478 (in Chinese)
- 479 Li, C., 2007. Qintongqi xuebu ji (Collections of research papers on ancient bronzes), Cultural Relics
  480 Press, Beijing. (in Chinese)
- Li, X., 1997. Lun Yangxian Fanba tong yazhang deng wenti (On the bronze sceptres unearthed from
  Fanba, Yanxian County and related issues), Wenbo (Cultural Relics and Museology), no.2, 13-14, 18.
  (in Chinese)
- Li, Y., Xian, Y., Chen, K., Yang, Q., Shao, A., Zhang, D., 2016. Shaanxi Luonan Hekou lvsongshi kuang
  yizhi diaocha baogao (Investigation report on the ancient turquoise mining site of Hekou in the Luonan
  county, Shaanxi province), Kaogu yu Wenwu (Archaeology and Cultural Relics), no.3, 11-17. (in
  Chinese)
- Liu, J., 2015. Scientific Study on the Shang and Zhou Periods Bronzes unearthed from Northern Shaanxi:
  Cultural connections between loess highland and Anyang in the Late Shang dynasty (Shanbei diqu chutu
  shangzhou shiqi qingtongqi de kexue fenxi yanjiu: jianlun shangdai wanqi jinshan gaoyuan yu anyang
  yinxu de wenhua lianxi), University of Science and Technology Beijing. (in Chinese)
- 492 Liu, L., Chen, X., 2003. State Formation in early China, Duckworth, London.
- Liu, L., Chen, X., 2012. The Archaeology of China: from the late Paleolithic to the Early Bronze Age,Cambridge University Press, New York.
- Liu, R., Rawson, J., Pollard, A.M., 2018a. Beyond ritual bronzes: identifying multiple sources of highly
  radiogenic lead across Chinese history, Sci Rep 8, 11770.
- Liu, S., Chen, K., Rehren, Th., Mei, J., Chen, J., Liu, Y., Killick, D., 2018b. Did China import metals
  from Africa in the Bronze Age?, Archaeometry 60, 105-117.

- 499 Liu, S., Li, R., Chi, G., Zeng, R., Liu, L., Shi, S., 2015. Geochemical Characteristics and Sources of Ore-500 forming Fluids of the Mayuan Pb-Zn Deposit, Nanzheng, Shaanxi, China, Acta Geologica Sinica 501 (English Edition) 89, 783-793.
- 502 Molofsky, L.J., 2009. A novel approach to lead isotope provenance studies of tin and bronze, The Uniersity of Arizona. 503
- 504 Peng, Z., Liu, Y., Liu, S., Hua, J., 1999. Gan-E-Yu diqu Shangdai qingtongqi he bufen tong qian 505 kuangliao laiyuan de chutan (Preliminary study on Shang dynasty bronzes and their Cu-Pb ore sources 506 in Jiangxi, Hubei and Henan provinces ), Ziran kexue shi yanjiu (Studies in the History of Natural 507 Sciences ) 18, 241-249. (in Chinese)
- Pollard, A.M., Bray, P.J., 2015. A new method for combining lead isotope and lead abundance data to 508 509 characterize archaeological copper alloys, Archaeometry 57, 996-1008.
- 510 Pollard, A.M., Bray, P.J., Gosden, C., 2015. Is there something missing in scientific provenance studies 511 of prehistoric artefacts?, Antiquity 88, 625-631.
- 512 Qi, W., Hou, M., 2005. Shaanxi tongkuangchuang leixing ji zhaokuang fangxiang (Copper ore deposit 513 types and prospecting direction in Shaanxi Province), Northwest Erngeology 38, 29-40. (in Chinese)
- 514 Radivojević, M., Roberts, B.W., Pernicka, E., Stos-Gale, Z., Martinón-Torres, M., Rehren, Th., Bray, P., 515 Brandherm, D., Ling, J., Mei, J., Vandkilde, H., Kristiansen, K., Shennan, S.J., Broodbank, C., 2018. 516 The provenance, use, and circulation of metals in the European Bronze Age: The state of debate, Journal 517
- of Archaeological Research. https://doi.org/10.1007/s10814-018-9123-9
- 518 Ren, X., Wang, R., Mao, J., Li, C., Xiang, T., Wang, J., 2007. Mian Lue Ning duo jinshu kuangjiqu quyu diqiu huaxue tezheng yu zhaokuang fangxiang (Regional geochemistry characters and exploration 519 520 direction for Mianxian-Lueyang-Ningqiang multimetal deposit cluster Area, Shaanxi Province, China), 521 Journal of Earth Sciences and Environment 29, 221-226. (in Chinese)
- Saito, T., Han, R., Sun, S., Liu, C., 2002. Preliminary consideration of the source of lead used for bronze 522 523 objects in Chinese Shang dynasty: was it really from the area where Sichuan, Yunan and Guizhou 524 provinces meet? BUMA-V (The Fifth International Conference on the Beginning of the Use of Metal 525 and Alloys), Korea, pp. 21-24.
- 526 Shennan, S., 1999. Cost, benefit and value in the organization of early European copper production, 527 Antiquity 73, 352-363.
- 528 Snoek, W., Plimer, I.R., Reeves, S., 1999. Application of Pb isotope geochemistry to the study of the 529 corrosion products of archaeological artefacts to constrain provenance, Journal of Geochemical 530 Exploration 66, 421-425.
- 531 Steinke, K., 2014. Erligang and the Southern Bronze Industries, in: Steinke, K., D., C. (Eds.), Art and 532 Archaeology of the Erligang Civilization, Princeton University Press, Princeton, pp. 151-172.
- 533 Sun, H., 2011. Shilun Chengyang tongqi cunzai de lishi beijing (On the historical contexts of the 534 occurrence of Chengyang bronzes), Sichuan Wenwu (Cultural Relics of Sichuan), no.3, 33-45. (in 535 Chinese)
- 536 Sun, W.D., Zhang, L.P., Guo, J., Li, C.Y., Jiang, Y.H., Zartman, R.E., Zhang, Z.F., 2016. Origin of the 537 mysterious Yin-Shang bronzes in China indicated by lead isotopes, Sci Rep 6, 23304.
- 538 Tang, J., Wang, S., Guo, C., 1980. Shanxisheng Chengguxian chutu yinshang qingtongqi zhengli jianbao 539 (Brief report of the Shang Dynasty bronzes unearthed from the Chenggu County, Shaanxi Province). 540 Kaogu (Archaeology), no.3, 211–18. (in Chinese)
- 541 Tian, J., 2013. Zhengzhou digu chutu Erligang qi tongqi yaniu (Scientific Study of Erligang Bronze 542 Artefacts unearthed in Zhengzhou area ), University of Science and Technology China. (in Chinese)

- von Falkenhausen, L., 2006. The Salt of Ba: Reflections on the role of the "peripheries" in the production
  systems of Bronze Age China, Arts Asiatiques 61, 12.
- von Falkenhausen, L., 2011. The Bronze Age in the upper Han River basin: some observations, in: Cao,
  W. (Ed.), Hanzhong chutu Shangdai qingtongqi (Shang Dynasty bronzes unearthed from Hanzhong),
  Bashu Press, Chengdu, pp. 378-516.
- 548 Wang, H., 2014. China's First Empire? Interpreting the material record of the Erligang expansion, in:
  549 Steinke, K. (Ed.), Art and Archaeology of the Erligang Civilization, Princeton University Press,
  550 Princeton, pp. 67-98.
- XDWX (Xibei Daue Wenbo Xueyuan), 2002. Chenggu Baoshan: 1998 niandu fajue baogao (Excavation report of the Baoshan site in the Chenggu County in 1998), Cultural Relics Press, Beijing. (in Chinese)
- Xian, Y., 2016. Shaanxi Luonan Laziya caikuang yizhi ji zhoubian lvsongshi chanyuan tezheng yanjiu
  (The Studies on the geographical origin characteristics of turquoise from the laziya mine sites in
  luonan ,shannxi province and the surrounding mine areas), University of Science and Technology Beijing,
  Beijing. (in Chinese)
- Xian, Y., Fan, J., Li, X., Li, Y., Zhou, X., Gao, Z., Wu, M., 2018. Shaanxi Luonan lvsongshi de si
  tongweisu tezheng jiqi chandi yiyi (Research on the source characteristics of Luonan turquoise by using
  strontium isotopic method), Xibei Dizhi (Northwestern Geology) 51, 108-115. (in Chinese)
- 560 Xu, J.J., 2008. The Sanxingdui site: Art and Archaeology, ProQuest, UMI Dissertations Publishing.
- Yue, Z., Liu, Y., 2006. Yinxu Zhutong Yizhi Zongshu (Review of the foundry remains in the Yinxu Site),
  Sandai Kaogu (Archaeology of the Three Dynasties) 2, 358-374.
- Zhao, C., 1996. 1996. Chenggu Yangxian tongqiqun zonghe yanjiu (A comprehensive study on the bronzes unearthed from the Chenggu and Yangxian counties). Wenbo, no.4, 3–26. (in Chinese)
- 565 Zhao, C., 2006. Chengyang qingtongqi (Bronzes unearthed from Chenggu and Yangxian), Cultural
  566 Relics Press, Beijing. (in Chinese)
- 567 Zhu, B., Chang, X., 2002. Ping "Shangdai qingtongqi gao fangshexing chengyin qian" de faxian
  568 (Comments on the "finding of Shang bronze artefacts with highly radiogenic lead isotope"), Gudai
  569 Wenming (Ancient Civilizations) 1, 278-283. (in Chinese)
- 570 Zhu, H., Gao, J., Zhang, D., 2006. Qinling diqu shouci faxian han fangshexing chengyin yichang qian de
  571 tongkuangchuang (Copper deposit with high radioactive lead anomaly discovered first in Qinling area),
  572 Kuangchan yu dizhi (Mineral resources and geology) 20, 461-464. (in Chinese)

## 573 **Figure Captions**

574 *Figure 1.* Map showing the sites/regions yielding HRL bronzes (red dots) and ore deposits mentioned in the text (white square). Pie charts show the proportion of HRL 575 bronzes among the analysed samples from the major Shang sites/areas (1. Jinsha; 2. 576 577 Sanxingdui (n=53); 3. Hanzhong (n=115); 4. Huaizhenfang; 5. Northern Shaanxi and 578 Shanxi (n=195); 6. Lingshi Jingjie; 7. Yuanqu; 8. Yanshi; 9. Zhengzhou (n=37); 10. Anyang (n=67); 11. Runlou (n=20); 12. Taijiasi; 13. Luoshan; 14. Panlongchen 579 (n=37); 15. Dayangzhou (n=19); a. Mayuan; b. Mujiazhuang; c. Luonan) 580 581 *Figure 2. Examples of Hanzhong bronzes (upper) and their alloy types (lower)* 582 583 showing the typological/cultural and material diversity of the assemblage (drawings are reproduced from Cao 2006 and Zhao 2006, chemical data are from Chen 2010) 584 585 Figure 3. (a) A plot of Hanzhong bronzes data, showing 1/Pb against lead isotope 586 ratio <sup>206</sup>Pb/<sup>204</sup>Pb. (b) A <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb diagram for Hanzhong bronzes 587 showing the distribution of lead isotope ratios of artefacts with different lead contents 588 589 590 Figure 4. Plots of lead isotope ratios of Hanzhong bronzes showing the overlap of objects from distinct sub-group, especially at the highly radiogenic region 591  $({}^{206}Pb/{}^{204}Pb \ge 20).$ 592 593 Figure 5. Plots of lead isotope ratios of major Shang period bronze assemblages 594 including Hanzhong. Note the large proportion of HRL values and the substantial 595 overlapping of the artefacts from various sites/regions. (see Supplementary material 596 597 for the sources of data used) 598 Figure 6. Plots of <sup>206</sup>Pb/<sup>204</sup>Pb ratios of objects from different sites/regions showing 599 that the uses of HRL bronzes are almost completely limited to the Shang period. 600 (ELG/PLC=Erligang and Panlongcheng, NS&S= North Shanxi and Shaanxi, TM-601 602 QC= Tianma Qucun). Note that the few objects with HRL signatures from the Yejiashan and Tianma-Qucun sites of the Western Zhou period are typologically 603 dated to the Shang period. (see online Supplementary Material for sources of data 604 605 used) 606 607 Figure 7. Schematic diagram showing the relative chronology of the sites/regions 608 mentioned in the text. Note the date of the whole assemblage of Hanzhong bronzes is in essence synchronous with the wide appearance of HRL objects. 609