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ORIGINAL PAPER

Provision of iron objects in the southern borderlands of the Han Empire: a metallurgical study of iron objects from Han tombs in Guangzhou

Wengcheong Lam[1] · Qianglu Zhang[2] · Jianli Chen[3] · Sum Yi Wu[4]

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Abstract

It is widely acknowledged that the iron industry developed during the Han period led to the widespread distribution of iron implements, but how such iron implements were supplied to the peripheries, especially the southern frontiers within the Lingnan region where evidence of local manufacturing has not been widely found, remains unclear. This paper presents the results of analyses of iron objects from Han tombs in Guangzhou, which was a major center in Lingnan, as a means of shedding light on the iron supply system in the region. The metallurgical and SEM-EDS analyses identified cast iron, fined iron, solid-state decarburization of cast iron, and bloomery iron within the tested assemblage. Since evidence for local cast iron manufacturing has not yet been identified in Lingnan, the discovery of iron or steel objects made by the cast iron process suggested that a supply and transportation system for final products might have developed linking Guangzhou, and perhaps other centers in Lingnan as well, to iron production centers located outside the region. Meanwhile, the comparison of slag inclusions (SIs) in bloomery iron products from Guangzhou and smelting slag samples from the Guiping-Pingnan area of Guangxi, which were dated between the Han and Southern dynasties period, did not strongly support a link between the two areas. More studies are needed to further test this potential link. Nevertheless, through collective consideration of the available evidence, we would argue that the supply of iron daily items did rely on external sources, which suggests that a relatively well-developed transportation network might have existed between Lingnan and other parts of the Han Empire.

Keywords Han Empire · Lingnan region · Guangzhou · Cast iron industry · Bloomery iron · Distribution · Burial goods

Introduction

The iron industry played a critical role in the state financial system of the Han Empire (206 BCE-220 CE). By 117 BCE, the Han state implemented the iron monopoly to take over the mining, manufacturing, and selling of iron implements throughout its territory as a way to increase its income (Wagner 2008, 192–210, 246). In ancient China, iron objects in the Central Plains region were made of cast iron (i.e., using blast furnace to smelt the ore and then cast objects that usually have about 4% of carbon, see Wagner 1993, 336). The technology appeared as early as the seventh century BCE following the initial development of bloomery iron smelting (i.e., smelting iron ores in the solid state) during the eighth century BCE, and was then widely adopted by territorial states during the second half of the first millennium BCE (Lam 2014; Lam and Chen 2017). Since blast furnaces usually required substantial amounts of labor and fuel (Wagner 2008, 144-146), the development of cast iron tradition may have been the key factor behind the Han state's decision to monopolize cast iron manufacturing by setting up large-scale ironworks in iron-rich regions (Lam et al. 2018). In addition to the appearance of

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✉ Wengcheong Lam
wlam@cuhk.edu.hk

[1] Department of Anthropology/Department of History, CUHK, Shatin, Hong Kong

[2] Guangzhou Municipal Institute of Archaeology, Guangzhou, China

[3] School of Archaeology and Museology, Peking University, Beijing, China

[4] Department of Anthropology, CUHK, Shatin, Hong Kong

large-scale iron production centers, the development of the iron industry during the Han period was characterized by the increasing distribution of iron implements alongside the expansion of state. In southwest (Chen et al. 2008a; Li 2011; Li et al. 2018, 2019), southeast (Chen et al. 2008b), and northeast China (Chen 2014), the metallurgical analyses of iron implements have suggested that iron or steel implements made by the cast iron process were widely distributed. Having said that, the iron industry in the southern peripheral region of Lingnan (i.e., present-day Guangdong, Guangxi region, and northern Vietnam) has as yet been overlooked in the literature (Huang et al. 2016b being the only exception).

After 112 BCE, the Han Empire conquered the Nanyue kingdom and imposed the commandery-county system upon the entire Lingnan region. Supported by the state, the development of trading ports such as Hepu in Guangxi and Panyu in Guangdong (present-day Guangzhou city) fully transformed Lingnan from a peripheral frontier region to an important player in the state economic system focused on the procurement of exotic goods (Xiong 2014; Xiong 2015). The popularization of intensive farming in the region also served as the foundation for economic development and, eventually, fueled the demand for various iron implements used in agricultural production and daily activities (Bai 2005; Liu 2017). There is, however, no evidence to date for the manufacturing of cast iron within the entire Lingnan region. But, were some iron implements locally made, and to what extent were iron implements imported into Lingnan from production centers outside of the region? Both questions remain unresolved, yet their answers are essential for addressing the development of transportation and the economy in Lingnan during the Han period.

In order to extrapolate the questions mentioned above, it is necessary to investigate iron technology in the region via metallurgical analysis. Even though the traditional technique in Central Plains China of using cast iron could be efficient, the production of cast iron requires considerable investment in facilities (e.g., furnaces, air supply system, and fining system), abundant workforce as well as substantial ores and fuel to generate the high temperature and reducing environment to melt iron and produce free-flowing slag (Wagner 1993, 48). Also, the large-scale consumption of raw materials and considerable amounts of products made by one cast needs a developed transportation network and copious market in order to make the manufacturing economical. In contrast, the requirement for all those aspects noted above in the case of bloomery iron manufacturing is far less. As evidenced by the recent discovery of bloomery iron in Daye of Hubei during the Qing period (1644-1912 CE) (Larreina et al. 2018; also see relevant discussion in Wagner 1993, 263-264), bloomery furnace produces small amounts of iron with a relatively low carbon content, which requires less heat and fuel. Bloomery iron production might be a more suitable “technological choice” in areas where production was constrained by the limitation of workforce, fuel, or under-developed communication routes. Guangzhou, or even the entire Lingnan, was constrained by the three factors discussed above. Together with the discovery of blommery iron smelting sites in the Guiping-Pingnan area in Guangxi that were recently reported (Huang and Li 2011, 2012a, b; Huang 2013; Huang et al. 2014), the identification of bloomery iron products in the region could be conceived as a potential signal of local iron manufacturing. The metallurgical study of iron artifacts and iron technology from Lingnan could therefore provide key information towards addressing, at least partially, the questions regarding local manufacturing and interregional transportation. It may also contribute to our understanding of the iron supply system in the region as well as the economic connection between the Han Empire and its peripheries.

This article presents the results of analyses of iron objects from Guangzhou, which was one of the main centers in Han period Lingnan. Even though more analyses of samples from other regional centers (e.g., Hepu) are needed in the future, the results presented in this paper show that the majority of iron tools and weapons in the Guangzhou assemblage were made of cast iron or solid-state decarburization of cast iron. Since no evidence for local cast iron manufacturing has hitherto been found, this pattern suggests that the residents of Guangzhou were well-connected to the provisioning system outside of Lingnan. Meanwhile, the discovery of bloomery iron in the assemblage suggests the possibility of local manufacturing. However, it remains unclear whether the bloomery iron was manufactured by the local smelting system in Lingnan based on the comparison between the slag from the smelting locales and slag inclusions in iron objects. Overall, the dominance of implements made of cast iron or a related steel-making technique evidenced by the study of iron from Guangzhou should be attributed to the existence of a developed transportation network between Lingnan and other parts of the empire, which may also have been a key underlying mechanism significantly contributing to the economic development of the Han Empire’s southern peripheries.

Background of Guangzhou and the development of iron industry in the South

This article focuses on the manufacturing techniques of iron objects discovered in the Guangzhou area through metallurgical analysis. In order to facilitate the explanation of analytical results, this section will briefly introduce the geographical and historical background of the research area as well as the development of the iron industry in the Han period.

Geographical background of Guangzhou and the Han Empire's control of Lingnan

Within the Lingnan region, Guangzhou, known as Panyu in the Han period, was a major center characterized by the high intensity of cemeteries surrounding the central walled town (Fig. 1). Geographically, the Lingnan region is circumscribed by the Nanling mountains that separate this region from the Yangtze river valley, with just a few mountain passes controlling the major North-South traffic routes (Liu 2014). In order to facilitate transportation to Lingnan, Qin Shihuang ordered the construction of the Lingqu canal in Guilin to connect the Pearl river to the Xiang river system in 217 BCE. The construction of pathways through the Nanling Mountains continued during the Han period, which further stimulated and facilitated overland transportation and communication between Lingnan and other parts of the Han Empire. Nonetheless, the limitation in transportation options still highly constrained communication in and out of Lingnan and, according to textual sources, this was not significantly improved until the Tang period (618–907 CE) (ibid.).

Before the arrival of the Qin army, the Lingnan region was home to Baiyue ethnic groups (or Hundred Yue) who might have had connections with other contemporary Yue ethnic groups in the southeast coast region (Brindley 2015, 28-35). In 214 BCE, the Qin state conquered the region and divided it into three commanderies: Nanhai, Guilin, and Xiang. After the collapse of the Qin state, Zhao Tao, a general originally from the Hedong commandery, took over and established the Nanyue kingdom. When the Han state implemented a policy forbidding the sale of iron goods to the Nanyue kingdom (*Shiji* 113, 2969 1997), the latter launched a campaign against the Changsha kingdom in revenge, which perhaps suggests that the supply of iron to the Nanyue kingdom might have been very restricted. According to the unearthed inventory record of iron implements found from the tomb of a high official of the Nanyue kingdom at Luobowan, Guigang (Guangxi 1988), a number of iron implements were imported to Lingnan from a center named Dongyang, probably in present-day Anhui province. During the Nanyue kingdom period, local iron manufacturing appeared to be generally less developed in comparison with other regions, particularly in the Central Plains, even though both Guangdong and Guangxi provinces were relatively rich in iron ores (Fig. 2) (Guangdong 1994; Guangxi 1992)

The Lingnan region was finally integrated within the Han imperial administration system after Emperor Wu's conquest in 112 BCE. Even though Lingnan was still constrained by its geography and undeveloped communication system, the migrations of exiled officials (*Hanshu* 12, 357 1997) and merchants might have introduced materials and technology into the former backwater, such as the use of oxen as draft animals and iron agricultural tools. Models from an Eastern Han tomb in Foshan, Guangdong, clearly demonstrated the existence of rice paddy fields and probably iron plow-shares in agricultural production during the Eastern Han period (Xu 1981). All the above factors combined together to accelerate the process of large-scale migrations from the North, which eventually led to rapid demographic growth in the Lingnan region (Wang 2014) and huge demand of iron implements in order to support the new economic developments. Thus, the investigation of the iron supply system would shed insight into factors underlying the economic development in the region during the Han period.

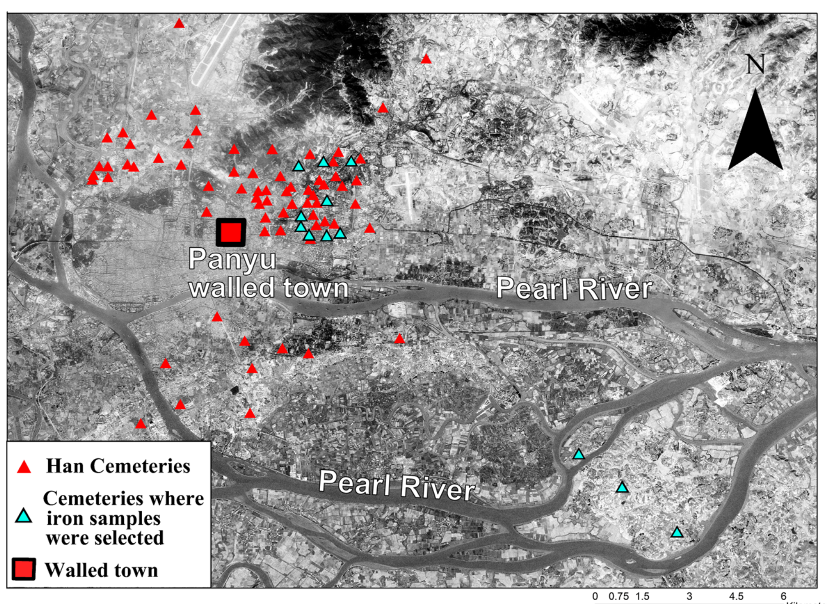

Fig. 1 Distribution of Han cemeteries surrounding Panyu and locations of cemeteries from which iron objects were selected for metallurgical analysis. (Redrawn from Guangzhou and Guangzhou 1981, 3, Fig. 1; background map: 1969 April Corona image)

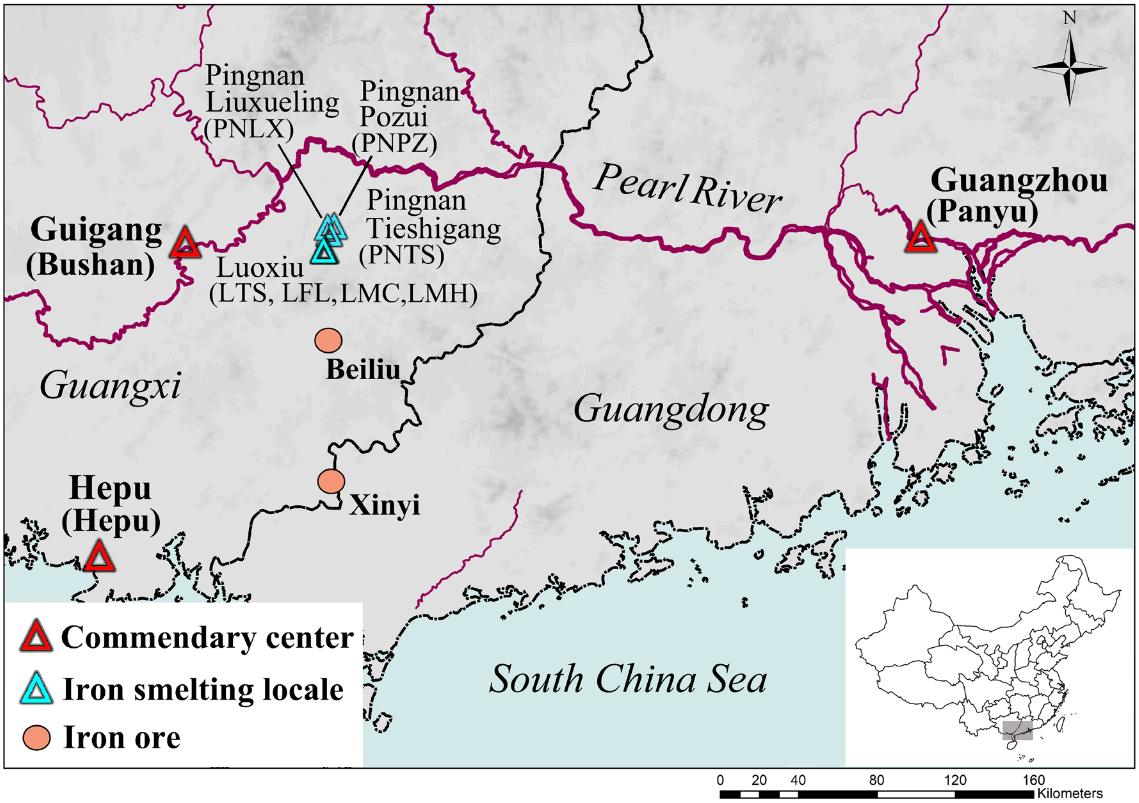

Fig. 2 Location of major centers in Lingnan, bloomery iron smelting locations found in the Guiping-Pingnan area, and iron ores located in the region. Data: bloomery iron smelting sites (Huang and Li 2012a, b; Huang et al. 2014); iron ores: Guangdong 1994, Fig. 2; Guangxi 1992, 146-149. Drawn by author in ArcGIS using a data set from Harvard's China Historical GIS (http://www.fas.harvard.edu/~chgis/data/chgis/downloads/v4/)

Development of iron technology and the iron industry in the South in the Han period

During the Qin-Han period, previous studies have suggested the use of four major approaches or methods for manufacturing wrought iron (iron with carbon content in the range 0.1–0.3%) and steel (iron with carbon content in the range 0.5–1%): solid-state reduction of bloomery iron, solid-state decarburization of cast iron, fined iron, and crucible steel (Chen and Han 2007; Wagner 1993, 288). Bloomery or direct process iron involves the direct reduction of iron from ores using a bloomery furnace. In contrast, the second and third types were made from cast iron. Solid-state decarburization of cast iron involves the annealing of a cast iron object in an oxidizing atmosphere in order to produce wrought iron or steel for smithing (Wagner 1993, 291). Fined iron refers to the process of converting cast iron into wrought iron or medium-carbon steel in a hearth or open fire, which is thus known as the indirect process (see Percy 1864, 579; Wagner 1993, 290-291). With the exception of the last type (crucible steel), which was rarely found and appeared probably after 200 CE (Zhou et al. 2014), the other three types of manufacturing techniques were all identified in the Han period in previous research (e.g., see cases in Chen 2014, 236-307).

Even though the improvement of agricultural technology and population growth in Lingnan are both reflected in archaeological findings, the development of cast iron technology as the basis for the everyday tools and implements is still poorly understood. As mentioned above, one critical factor is that no cast iron production sites have yet been found in main centers in the entire Lingnan region. Although it is generally understood that surface visibility during archaeological surveys in Lingnan is relatively low, cast iron foundries usually generated voluminous material residues such as slag and ceramic molds, and are often encountered in urban centers when archaeological intensive works were conducted (Lam et al. 2018; Shaanxi 2018). During the past decades, archaeological works have intensively investigated areas surrounding Panyu walled-town in present-day Guangzhou (Fig. 1) and identified palaces of the Nanyue kingdom and a ceramic workshop (Guangzhou 2003b), but no iron manufacturing remains were found. Thus, the absence of iron manufacturing remains in archaeological investigations in Guangzhou reinforces the idea that large-scale iron production, especially of cast iron, was unlikely to take place in major centers in Lingnan (Huang 2015).

In the Western Han period, the Han state controlled regional iron manufacturing industries by setting up iron officials across the state (Wagner 2008, 192–217), but according to textual records, there was no iron official setup in Lingnan. One text in *Hou Hanshu* (76, 2462 1965) mentioned that a commandery official, Ren Yan, taught local people in Jiuzhen (present-day northern-central Vietnam) to cast iron agricultural implements during the early Eastern Han period (the first century). This is also reflected in excavated texts from *Shuifudi* (Hulsewe 1985, 27, A8) which indicated that the local government was responsible for providing agricultural implements. Whether Ren Yan's efforts resulted in the development of a large-scale iron industry remains unclear. But if there was an inadequate level of iron manufacturing in the region, did the Han state manage to supply to its southern peripheries with agricultural tools via transportation from external sources? Moreover, were other daily-use implements, such as iron knives, also procured via external sources? Since iron was a commodity essential to the state's economy, the nature of production and distribution of iron in the South, the identification of indicators showing the extent to which iron objects were supplied by the production system outside Lingnan, and interregional transportation, are all essential to the study of the economic system of the Han state.

Given the significance of iron implements for the Han state and the huge demand involved, some scholars (e.g., Qu 2009) have started to challenge the mainstream idea in the literature that the local industry in Lingnan did not develop until the Southern dynasties (420–589 CE) (Huang 2015; Xu 1981). More importantly, recent archaeological work has identified clusters of bowl-shaped pits used for iron smelting in the Guiping-Pingnan area of Guangxi, which were collectively dated to the period between the Han and Southern dynasties according to the diagnostic characteristic of sherds and radio carbon-dating results (Huang and Li 2011, 2012a, b; Huang 2013; Huang et al. 2014) (Fig. 2). Metallurgical analyses of slag confirmed that these pit-furnaces were employed for smelting bloomery iron. Similar bloomery iron furnaces were also found in Wuzhou of Guangxi dating to Southern dynasties (Meng and Zou 2017). In addition, one iron object from an Eastern Han tomb in Guiping, the same area where iron smelting sites were found, was identified via metallurgical study as bloomery iron (Huang et al. 2016b). When viewed together, these findings reasonably suggest that there might have been a long tradition of manufacturing bloomery iron starting in the Han dynasties in southern China. The new discoveries also imply that the supply of iron in Lingnan might have relied, to a certain extent, upon the local manufacturing system, as some studies have previously argued (Zheng 1996). In contrast, previous studies of ironworks in the Central Plains have hitherto revealed no unambiguous evidence for the manufacturing of bloomery iron (e.g., Chen et al. 2011; Du et al. 2011, 2012; Li 1995; Shaanxi 2018).

Despite the growing interest in the iron industry in Lingnan during the Han period, the reconstruction of the entire iron manufacturing system there is hindered by several challenges. Previous studies of ratios of non-reduced elements (NRCs), trace elements, and isotopes have demonstrated that these dimensions could be useful for showing the connection between the final bloomery iron objects and relevant slag (or parent ores) (e.g., Blakelock et al. 2009; Charlton et al. 2012), but much gangue in parent ores is already reduced and entered into slag during the smelting process of cast iron, leaving limited direct evidence available for the study of provenance in cast iron. However, the study of iron objects themselves could at the very least provide some indirect evidence to trace their potential provenance based upon the manufacturing techniques and skills evidenced.

Although one should not over emphasize the presence or absence of bloomery smelting as the major difference between the core and peripheries in the Han period, the study of iron making techniques offers the first step towards a better understanding of regional contrasts in the manufacturing and transportation of iron implements in Han China. To be more specific, if cast iron, solid-state decarburization of cast iron, or fined iron are identified in Lingnan, these items are very likely to have been imported from external sources, either through commercial exchange, state-controlled transportation, or importation associated with migrations to the South. In contrast, since bloomery iron manufacturing had been identified in Lingnan, the discovery of bloomery iron suggests that the sample might have been manufactured by a local production center, especially if the ratios of chemical composition ratios in slag inclusions (SIs) in iron objects match those of smelting slag.

In summary, this study attempts to analyze "technological signature" of iron samples in order to understand their potential source, and thereby contribute to ongoing discussion concerning the production and supply of iron in Han dynasty Lingnan. For this purpose, the main aim of this study is to first present a metallurgical analysis of manufacturing techniques represented by the iron objects from Han tombs in Guangzhou, as a way to evaluate the overall characteristic of the iron industry in the region. By analyzing SIs in bloomery iron objects that were identified in the collected samples, we hope to further test the hypothesis if a potential connection existed between the production sites and bloomery iron objects found in Lingnan. The results of these two main questions would then lay a foundation for extrapolating the transportation system within Lingnan as well as the connection between the southern frontiers and other parts of the Han state.

Methodology and sample collection

In order to test the hypotheses set out above, this study collected samples from Han tombs in Guangzhou, which in the Han period was an urban center called Panyu within the

Nanhai commandery. In the Lingnan region as a whole, residential sites dating to the Qin and Han periods have rarely been excavated and reported on. In contrast, a huge number of Han tombs have been excavated in the environs of Panyu city (Fig. 1) (e.g., Guangzhou 1998, 2003a, 2004; Guangzhou and Guangzhou 1981, 2006), yielding a considerable number of iron objects suitable for metallurgical study. However, no systematic scientific studies of such iron objects have yet been carried out and there has been no attempt to synthesize and interpret the results within broader historical contexts. Only a few iron objects from the Nanyue mausoleum have been subjected to metallurgical analysis before (Beijing 1991). Since these materials were dated to the Early Western Han period and only reflect the situation before the conquest of Nanyue by the Han state, this project selected iron objects from Middle/Late Western Han and Eastern Han tombs that were found in residential sites surrounding the ancient city of Panyu. A total of 57 iron objects were selected for metallurgical analyses (for information and identification results, see Supplement A). Even though the means by which such iron objects were procured by the interred individuals must remain a question to be answered through subsequent research, the metallurgical analysis should at least provide information that might allow some of the questions mentioned above to be addressed.

This study selected samples including iron swords, iron knives, and few other types of iron tools (such as axes and chisels) that are usually found in Han tombs in the region (Fig. 3). For objects that were broken, multiple samples from different fragments under the same numbering code were taken for metallurgical analysis before the conservation procedures were conducted. For larger objects such as swords, multiple samples from different parts of the object were also collected. When the types based on

Fig. 3 A selection of iron objects from Han tombs in Guangzhou selected for metallurgical analysis. (1. Axe 23042; 2. Axe 23044; 3. Axe 23048; 4. Caldron stand 23050; 5. Sword 23066; 6. Ring 23064; 7. Long sword 23032; 8. Ring 23034; 9. Sword 23031; 10. Sword 23018; 11. Spear-head 23049; 12. Ring-pommel knife 23038; 13. Unknown iron tool 23029; 14. Knife 23037; 15. Ring-pommel knife 23015; 16. Unknown iron tool 23028; 17. Nail 23027; 18. Ring-pommel knife 23016; 19. Hook 23045; 20. Small knife 23043; 21. Ring-pommel knife 23026; 22. Chisel 23041; 23. Hook 23062; 24. Hook 23019; 25. Ring 23006; 26. Ring 23036; 27. Hook 23033; 28. Banded iron bar 23017; 29. Hook 23022; 30. Unknown iron tool 23012 31. Hook 23039)

Table 1 Types of iron artifacts from Han tombs in Guangzhou selected for metallurgical analysis

Types	Rings	Nails	Knives	Big knives	Swords	Chisels	Axes	Caldrons	Caldron stands	Spear-heads	Others	Sum
Total	14	8	13	1	6	3	3	1	1	1	6	57

morphological attributes in our selected samples from Guangzhou (Table 1) are compared with previously studied assemblages from the political core region of the Guanzhong basin in Shaanxi (Shaanxi 2018), there seems to be no obvious difference between them. Metallurgical analysis is thus essential for further exploring the manufacturing and supply system of iron implements found in the southern peripheries of the Han Empire.

The iron samples were prepared under standardized preparation. They were mounted in a two-component epoxy resin, ground grinded with SiC paper (from grade 180 to 3000) under water and then polished using diamond suspension (1 and 0.05 μm). Then, the sample was observed under an optical microscope (Leica DLLM) after a 2% natal etching to study the microstructure such as the distribution of carbon content, different kinds of inclusions in the artifact, and evidence of forging or welding. After standardized preparation, iron samples with SIs (slag inclusions) were then subjected to SEM-EDS (scanning electron microscope with energy dispersive spectrometry) analysis employing the equipment (SEM: JSM-7800f, EDS: Oxford X-Max) in the Department of Physics at CUHK. Before the SEM-EDS analyses, the samples were coated with platinum. Measurement was performed with an accelerating voltage of 15 kv, spot size of 8 μm^3, working distance of around 10 mm, and 40-s collection time. Both backscatter and secondary electron modes were employed to help assess the microstructure of SIs. During the testing, the results were collected by the "standardless quantitative method," which relies on the data set and calculation model provided by AZTEC software. The EDS data were collected in weight%, combined with oxygen by stoichiometry and expressed in oxides (i.e., Na_2O, MgO, Al_2O_3, SiO_2, P_2O_5, K_2O, CaO, TiO, MnO, FeO), and normalized to 100%. In addition, only inclusions greater than 10 × 10 μm were analyzed in order to reduce the "localized concentration effects" noted by Dillmann and L'Héritier (2007, 1815). We employed both spot analyses to collect the chemical compositions of particle features or phases, and bulk analyses to collect the average chemical compositions of each selected SI. In order to obtain representative data, we aimed to collect data for at least 20 SIs covering the entire cross-section of polished samples if their condition was good enough (i.e., most of the sample had a solid metallic body). For welded objects, we analyzed at least 10 SIs in each welded layer (zone) with a sufficiently good condition.

As introduced above, three methods for making steel or wrought iron (solid-state decarburization of cast iron, fined iron, and bloomery iron) might have been employed in Lingnan at the same time, and metallurgical analysis was therefore the key approach needed to identify the types of iron or steel used. We previously explained the major differences between these types of iron in one of our earlier studies (Lam et al. 2018). Summarized below are the key criteria needed to facilitate the identification of different types of iron.

The first type, made by solid-state decarburization of cast iron, usually contains very few if any SIs, since much gangue in ores was mostly reduced during the smelting process. For the second type, fined iron, the metallic body usually includes large numbers of SIs comprising materials from the furnace, flue, ashes, and fluxes used in the process. Thus, its microstructure is sometimes similar to that of bloomery iron. However, SIs in fined iron and bloomery iron were introduced by different processes. Non-reduced compounds (NRCs, i.e., elements that are usually not fully reduced in the direct process such as Al, K, and Mg) in SIs in bloomery iron were derived primarily from parent ores due to the relatively low temperature and incomplete reduction process. In contrast, NRCs in SIs in fined iron formed during the second stage of refining, and are derived from the furnaces, fuels, and fluxes used. For this reason, the ratios of NRCs in SIs in these two types of iron would show different patterns. Dillmann and his team (Dillmann and L'Héritier 2007; Disser et al. 2014) analyzed 138 known samples of fined iron and bloomery iron, and employed multivariate analysis (see Eq. (1)), based on the ratios of NRCs including MgO, Al_2O_3, K2O, SiO_2, CaO, and MnO, in order to calculate eight coefficients. By applying these eight parameters to other samples containing SIs but of unknown type, this statistical method was able to compute a logit(p) for SIs and corresponding probabilities for samples that had resulted from an indirect or a direct process. Our study of iron from a Han ironworks at Taicheng, Shaanxi, confirmed the initial applicability of this multivariate approach in differentiating fined iron from bloomery iron (Lam et al. 2018), alongside other indicators such as highly deformed SIs and the presence of "sub-double phase inclusions" (i.e., SIs include fayalite, iron oxide, and glassy matrix but do not display eutectoid phase separations as in double phase inclusions).

Even though the multivariate method has proved to be successful in addressing the type of iron used in architectures in medieval France (Disser et al. 2014), more empirical studies are still needed in order to fully evaluate the use of this method for the discrimination of indirect and direct process products in ancient China, in particular given the differences of technological context and types of objects involved between these cases. As demonstrated in previous studies (Han 1987), scrap iron would often be reused, and high-quality objects (e.g., swords) usually reveal hundreds of layers produced by refining and forging; both processes could generate blurring effects caused by new SIs being introduced. We therefore suggest combining the multivariate statistical method of average NRCs with the identification of high Ca-P phases in SIs. Previous literature suggests that high Ca-P phases are often identified in SIs of fined iron objects and are probably an important indicator of the indirect process (Chen and Zhang 2016; Han and Chen 2013; Han and Ke 2007:614; Huang et al. 2016a; Liu et al. 2019; Yang et al. 2014). Due to thermodynamic reasons, P in cast iron during the fining process will be firstly reduced and then appear in the α-Fe phase of iron as a P eutectic (Chen and Zhang 2016). In order to prevent the formation of iron phosphide in the reducing environment, Ca-rich fluxes (e.g., limestone) would be added to facilitate the formation of $3CaO.P_2O_5$, which would then be trapped in SIs. Therefore, the combination of multivariate analyses that were based on bulk chemical compositions together with the spot-scanning results indicative of high Ca-P phases in SIs could provide a more reliable approach for identifying fined iron and bloomery iron products (Lam et al. 2018; for the specific criteria, see Table 2). In this paper, we define high Ca-P phases as those containing more than 15 wt% CaO and P_2O_5 respectively, since a previous experimental study has shown that bloomery iron slag can contain up to 10 wt% P_2O_5 if ores contain relatively high P (Crew 2000; Török and Thiele 2013). In addition, in the table of statistical results, we will list various scales of Ca and P wt% (e.g., both ≥ 5 wt%, ≥ 10 wt%, ≥ 15 wt%, and ≥ 20 wt%) to facilitate the explanation of spot analyses.

Equation (1) (Disser et al. 2014, 326, Eq. (4); parameters of the logistic regression estimated were based upon Disser et al. 2014, 328, Table 5)

$$\begin{aligned}\text{Logit}(p) = \beta^0 &+ \beta^{Mg}\left[\%MgO^{**}\right] + \beta^{Al}\left[\%Al_2O_3{}^{**}\right] + \beta^{Si}\left[\%SiO_2{}^{**}\right] \\ &+ \beta^{P}\left[\%P_2O_5{}^{**}\right] + \beta^{K}\left[\%K_2O^{**}\right] + \beta^{Ca}\left[\%CaO^{**}\right] \\ &+ \beta^{Mn}\left[\%MnO^{**}\right]\end{aligned}$$

Logit(p): the result of the multivariate statistical study; Intercept β^0: 5.22; β^{Mg}: 0.13; β^{Al}: − 0.95; β^{Si}: 0.007; β^{P}: 0.16; β^{K}: -0.84; β^{Ca}:0.088; β^{Mn}: 0.018

For bloomery iron, although various factors including ores, types of furnaces, fuel ash, fluxes, and various furnace operating parameters might have contributed to slag formation, previous studies (e.g., Blakelock et al. 2009 and studies discussed within) have shown that, to a certain extent, the chemical compositions of slag inclusions in bloomery iron are closely related to the smelting slag produced by the same smelting system. Thus, the comparison of NRC ratios in smelting slag and SIs trapped in iron objects is confirmed to be a "simple and useful strategy for the potential tracing of the provenance of iron objects" (ibid., 1756). Blakelock's experimental study based on the SIs in objects and the relevant slag produced during the experimental smelting and smithing suggested that the ratios of MgO/K_2O and SiO_2/MnO were consistent between smelting slag and SIs in given objects manufactured by the same system. Meanwhile, the Al_2O_3/SiO_2, Al_2O_3/MgO, Al_2O_3/K_2O, and Al_2O_3/CaO ratios were shown to be higher in the smelting slag than in the SIs of objects manufactured by the same system, even though these ratios could be more prone to variable fuel and ash contributions. Based upon Blakelock's approach, Charlton et al. (2012) employed a multivariate model to transform the ratios of compounds in smelting slag and SIs in order to calculate the NRC compositional patterns, which more accurately identifies SI types and reveals the compounds in SIs generated from the same ironmaking system.

According to the mechanism explained above, this study compares the SIs in bloomery iron objects found in Guangzhou with smelting slags from the Guiping-Pingnan area of Guangxi (Huang and Li 2011, 2012a, b; Huang

Table 2 Identification standards of direct and indirect process employed in the article.

Case	Logit(p) shows a high probability of indirect process	Logit(p) shows a high probability of direct process	Logit(p) shows an undetermined stage	Identification of high Ca-P compounds	Result
1	Y	–	–	Y	ID
2	–	Y	–	N	D
3	Y	–	–	N	ID
4	–	Y	–	Y	Undetermined/ possibly ID
5			Y	Y	Possibly ID

ID indirect process, *D* direct process

2013; Huang et al. 2014), which is the suspected source of some Guangzhou iron objects. Iron working sites in the Guiping-Pingnan area extends over a wide hilly area measuring 100 km^2 along the upper Pearl river valley, and comprises a number of previously identified bloomery smelting locales: namely, Luoxiu with four loci (coded LTS, LFL, LMH, and LMC), Pingnan Liuxueling (PNLX), Pingnan Pozui (PNPZ), and Pingnan Tieshigang (PNTS) (see Fig. 2). Even though the multivariate model (Charlton et al. 2012) can generate a faster result than using the bivariate oxide comparisons, there is a greater need for bulk compositional analyses of SIs and smelting slag in order to conduct the analysis, and such data were not available in previously published studies. Therefore, we compared NRC ratios using bivariate graphs as a basic means of data exploration. Through looking at the NRC ratios (i.e., Al_2O_3/ SiO_2, Al_2O_3/MgO, Al_2O_3/K2O, Al_2O_3/CaO, MgO/K_2O and SiO_2/MnO) in SIs and smelting slag, this study attempts to test the hypothetical links between the bloomery iron objects from Guangzhou and smelting slag from Guangxi.

Identification results

Metallographic analysis

Given their relatively poor level of preservation, the majority of iron samples were unidentifiable to the type of iron or steel due to heavy corrosion (Supplement A). In this study, only ten objects had a sufficiently sound metallic body to allow detailed metallographic examination (Table 3). Even though it is extremely difficult to identify corroded objects, any traces of grain structure, if still recognizable, are useful for understanding the original manufacturing technique involved. According to the traces of its grain structure, one object (23019) was probably made of cast iron (Fig. 4a). Meanwhile, a total of 13 samples were identified as either low/medium carbon steel or wrought iron (Table 4). Among these samples, traces of SIs were barely identified (e.g., 23032 Fig. 4b). But given the poor condition of the metallic body, one cannot entirely rule out the possibility that SIs were originally present. In other words, these samples might have been solid-state decarburization of cast iron, fined iron, or even bloomery iron.

The ten objects with preserved metal core can be divided into two groups. The first group, containing three samples (23016, 23041, 23042), includes objects made of medium or low carbon steel (i.e., primarily ferrite with pearlite) (Fig. 4c, d, and e). Since very few micro-scale SIs were sparsely identified within the body of metal, these samples should be identified as steel decarburized from solid-state cast iron. In addition, these three objects comprise an axe, a chisel, and a ring-pommel knife; the latter being the only sample of this kind with preserved metallic body in this study. It is noteworthy that one of these objects, 23016, shows an uneven distribution of carbon content within the metallic body, with pearlite + ferrite in the center and mostly ferrite to either side (Fig. 4c), thus indicating that the object had been decarburized unevenly. Previous metallurgical studies of iron in Han China suggest that steel decarburized from solid-state cast iron was commonly used to make ring-pommeled knives (e.g., Lam et al. 2018; Liu et al. 2014). The example (23016) from Guangzhou appears to have been made using a technical tradition similar to that in the Central Plains.

The second group of artifacts (seven objects) differs in that they all include varying frequencies of elongated, non-metallic SIs within the metal (Figs. 5, 6, and 7). Also, these samples reveal traces of various manufacturing processes. Sample 23045 (chisel) has evidence that the object was formed by forging at least one piece of low-carbon steel (Fig. 5b) into the socket part of the chisel for hafting (Fig. 5a). Sample 23018 was clearly made by welding at least two pieces of iron/steel together; one side (the upper part) is mild-carbon steel with a layered structure, while the other side (the lower part) is wrought iron (Fig. 6a). The microstructure of 23043 shows a layered structure of steel with various carbon contents, suggesting that the raw material was folded multiple times (Fig. 6b). In addition, for samples 23018 (Fig. 6a), 23043 (Fig. 6b), 23048 (Fig. 7a), and 23067 (Fig. 6c), some SIs within the body of the metal were characterized by a sub-double phase structure with iron oxide and a glassy matrix (Figs. 5c and 6d). Meanwhile, a three-phased structure consisting dendritic wüstite with fayalite and glassy matrix (Fig. 7c, d) was observed in SIs in one sample (23050). As indirect and direct processes might present different types of SIs, these objects had to be subjected to further statistical analysis for the purpose of identification.

Chemical compositions of samples with SIs and manufacturing techniques

As introduced above, in order to study the manufacturing techniques of samples with SIs, the bulk compositional analyses of SIs (Supplement B) were subjected to multivariate analyses. The calculation of the logit(p) value of SIs in 23050 will be used below to illustrate the data treatment process. Since the forging processes also contributed to the formation of SIs in the metal, we first conducted WARD and PCA analyses on the SIs dataset (Fig. 8) after converting the five oxides (MgO, Al_2O_3, K_2O, SiO_2, and CaO) into log-ratio data (XiNRC) following the treatment procedures suggested by Disser et al. (2014, 324, Eq. (2)). The grouped results were then displayed as mass% values on bivariate plots (Fig. 9) to determine which group of SIs was more relevant to the refining/reduction stage of the manufacturing process. In this

Table 3 Description of metallurgical results of selected iron objects

CUHKLab no.	Date	Original record no.	Types of objects	Numbers of samples taken from the same artifact	Materials and techniques	Metallographic and slag inclusions structure
23016	Western Han	2003DZM73:6	Ring-pommel knife	1	Steel made by solid-state decarburization of cast iron	The distribution of carbon content is not even. The central part (lower part in the metallography) is pearlite + ferrite (hypoeutectoid steel). The edge (upper part in the metallography) consists of ferrite with small amount of pearlite, indicating the carbon content is relatively lower (Fig. 4c)
23018	Western Han	2005GBXM19:7	Large iron sword	4	23018(4) was the only sample with sound metallic body. Other samples from the same artifact were completely corroded. This sample was welded by at least one piece of wrought iron and one piece of medium carbon steel, but whether they were made of fined iron or bloomery iron is undetermined	Zone 1 (upper part) is pearlite + ferrite containing relatively dense and elongated SIs. The microstructure shows at least two different processing directions. Zone 2 (lower part) shows ferrite + few pearlite but only includes relatively few SIs (Fig. 6a)
23041	Eastern Han	2000YHGT4(13):001	Chisel	1	Steel made by solid-state decarburization of cast iron	Pearlite + few ferrite with very few SIs (Fig. 4d)
23042	Eastern Han	2000YHGT3J13东隔梁:001	Axe	3	All samples are steel made by solid-state decarburization of cast iron	Pearlite + ferrite (hypoeutectoid steel). Carbon content is about 0.3–0.4%. The microstructure is rather homogeneous with extremely few and small SIs (Fig. 4e).
23043	Eastern Han	2000GYST1⑦:052	Small knife	1	Possibly fined iron	From bottom to the top includes at least 8 layers of steel with slightly varied carbon content and were segregated by high P band. SIs were highly deformed and unevenly distributed in some layers. Some SIs are with relatively high Mn. Also, the microstructure of SIs shows sub-double phase structures (Fig. 6b, e)
23044	Western Han	2006GDJM045:003	Axe	2	One sample 23044(1) preserved sound metallic body. It is low-carbon steel, but whether it was made of fined iron or bloomery iron is undetermined	Ferrite + few pearlite. Grains are equalized and about 5–6 grade. SIs were slightly deformed aligning with the processing direction. The microstructure of SIs includes iron-oxide with fayalite (Fig. 6d).
23045	Western Han	2006GDJM045:004	Chisel	2	The two samples are probably fined iron	The microstructure belongs to hyproeutectic steel, including mostly ferrite and pearlite. The carbon content is uneven and increases from the lower part (mostly ferrite) to the upper part (ferrite + pearlite) of the photomicrograph. Widmanstätten structure is identified in some location. Carbon content is not evenly distributed. SIs were deformed

Table 3 (continued)

CUHKLab no.	Date	Original record no.	Types of objects	Numbers of samples taken from the same artifact	Materials and techniques	Metallographic and slag inclusions structure
						and aligned with processing direction (Fig. 5)
23048	Western Han	2006GDJM045:007	Axe	1	Bloomery iron	Ferrite with small amount of pearlite. Carbon content is higher in the upper part than the lower part in the metallography. SIs were highly elongated and deformed, including fayalite and iron oxide. The distribution of SIs is relatively uneven (Fig. 7a).
23050	Western Han	2005BHM46:9	Caldron stand	1	Bloomery iron	Pearlite + ferrite with widmanstätten structure. The carbon content is highly heterogeneous. Large SIs are irregular with slight degree of deformation. The microstructure of most SIs includes wüsite + fayalite two-phase structure (Fig. 7b, c, and d).
23067	Eastern Han	2010GXZM1:40	Long sword	2	One sample 23067(2) preserved sound metallic body, and is identified as fined iron	Pearlite + ferrite with elongated SIs. The orientation of SIs in the middle (vertical) is different from that on both sides (horizontal), suggesting the samples were welded by multiple pieces (Fig. 6c)

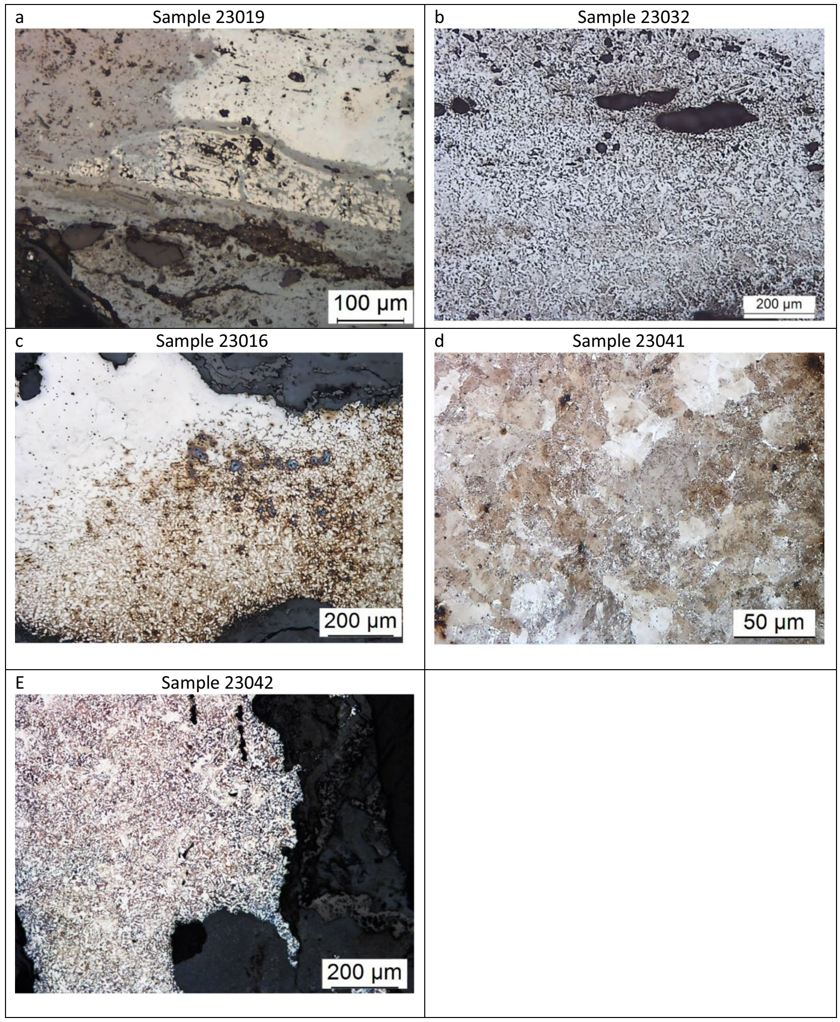

Fig. 4 Optical photomicrographs of selected iron samples. **a** Corroded but shows the microstructure of cast iron (ledeburite + pearlite + cemetite); **b** Corroded but shows a trace of ferrite and pearlite grains; **c** Ferrite + pearlite, and the carbon content gradually increases from the left side (ferrite) to the right side (ferrite + pearlite). The uneven distribution of carbon content indicates that the object had been decarburized unevenly; **d** Pearlite + ferrite and **e** is characterized by ferrite + pearlite. All optical photomicrographs were taken under plane polarized light, unless otherwise stated

case, we selected cluster 2 for the multivariate analysis, since the values of this cluster were relatively concentrated and better aligned towards the zero point. Also, the other two clusters include SIs with very high Si content (> 80 wt%), which might have been impurities (e.g., sand) that entered into the metallic body during the forging process.

After identifying the cluster of SIs that was more likely to be related to the refining/reduction process, the average logit(p) value of selected SIs was then calculated according to the multivariate framework (Eq. (1)). In order to avoid the introduction of errors due to the overestimation of the Fe content, we followed Disser et al.'s methodology (2014, 325) to calculate a new compositional ratio (%Oxide**) for each oxide except iron before applying the logistical regression model. For sample 23050, the logit(p) value is − 4.22, and this sample is likely to have been made by the direct process (Table 5). For samples with relatively small sample-size of SIs (e.g., less than 15 SIs analyzed by bulk analyses) due to

Table 4 Results of metallurgical identification of iron samples from Guangzhou

Types	Cast iron	Steel decarburized from solid-state of cast iron	Fined iron	Possibly fined iron	Fined iron or bloomery iron (un-determined)	Bloomery iron	Medium/low carbon steel or wrought iron (heavily corroded object)
Lab no.	23019	23016, 23041, 23042	23067	23045, 23043	23018, 23044	23048, 23050	23012, 23026, 23028, 23032, 23034, 23037, 23038, 23061, 23062, 23063, 23064, 23065, 23066
Sum	1	3	1	2	2	2	13

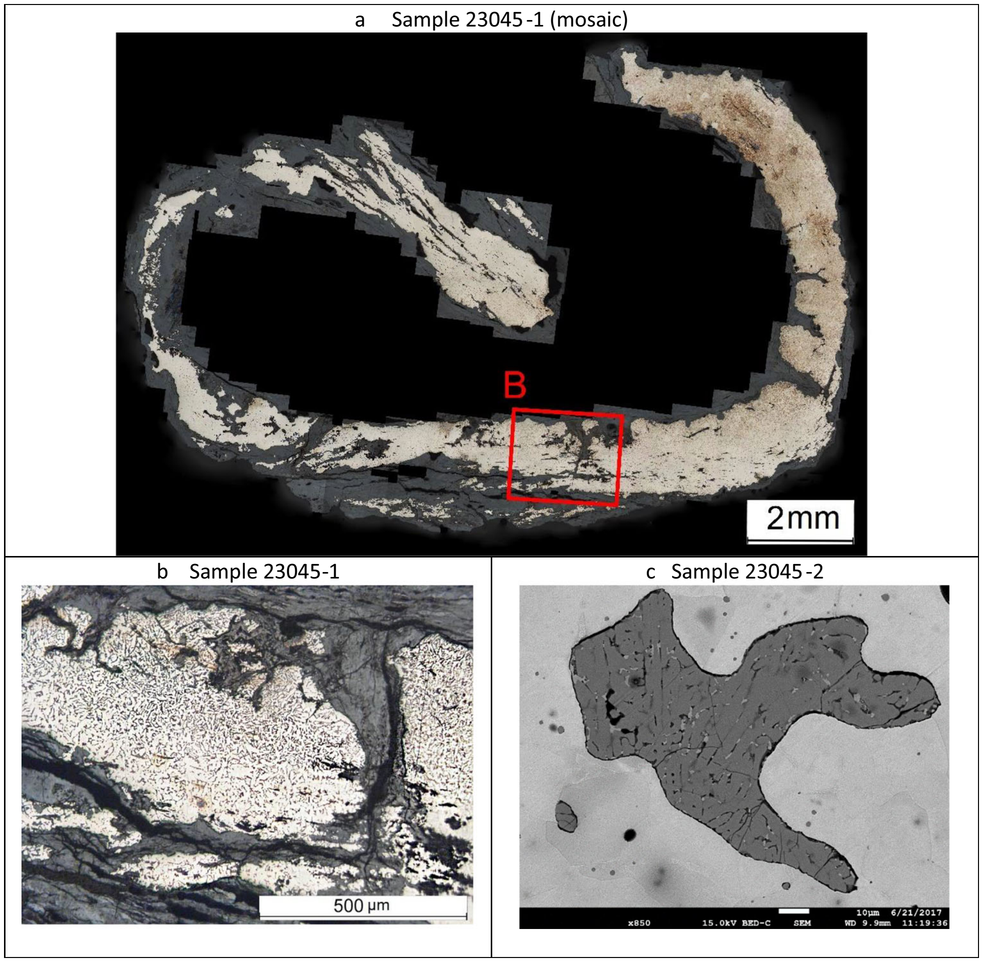

Fig. 5 Photomicrographs and SEM images of 23045-1 and 23045-2. **a** A C-shaped cross-section, indicating the object was forged into shape. The metallic body is characterized by ferrite + pearlite with elongated SIs; **b** The photomicrographs of the red square in (**a**) which is characterized by ferrite + pearlite. The carbon content is uneven and increases from the lower part (mostly ferrite) to the upper part (ferrite + pearlite) of the photomicrograph; **c** A SEM (backscattered) image of 23045-2 showing fayalite (grey phase) and iron oxide (light grey phase) between the fayalite

the relatively small area of sound metal preserved, the PCA and WARD analyses might generate large error variance. In that case, we used all SIs in an iron object collected to calculate its logit(p). In addition, since sample 23018 includes at least two layers (zone1 and zone 2), all the SIs in each zone will be subjected to individual classification analysis, and the logit(p) of each of these zones was therefore calculated and presented separately in Table 5. According to the logit(p) calculation, only the value of 23067 (cluster 3) indicated that the sample was probably made of fined iron (indirect process); all other samples with a metallic body fell within the direct process or undetermined range of logit(p) values.

Even though the logit(p) value of most iron objects with SIs show that, with high probability, they were made by the direct process, the results of spot analyses of different phases in SIs must be taken into consideration. In Table 6, we list the frequencies of points with various concentrations of wt% CaO and P_2O_5 that were recorded. In sample 23067, the object identified as fined iron (indirect process), there are four points with more than 15wt% of CaO and P_2O_5. Given the probability suggested by the logistical regression model and the identification of $3CaO.P_2O_5$, this sample matches the criteria of fined iron. Meanwhile, among the other six samples containing SIs that are probably related to direct process manufacture based on their logit(p) values, only two samples (23050 and 23048) had no phases or features with wt% of CaO and P_2O_5 both above 5% (Table 6). In other words, these two objects did not have high Ca-P phases in their SIs, indicating a high probability that they were made by the direct process.

However, it is important to note that among the samples identified as "direct process" or undetermined based upon logit(p) values, there are two objects (23043 and 23045) in which a few phases with more than 20 wt% of CaO and P_2O_5 respectively were identified. According to the identification criteria discussed above (Table 2), sample 23043 should be identified as possible fined iron. Sample 23045 also contains SIs with relatively high Ca and P content but has logit(p) values suggesting a high probability of direct process manufacture. Since high Ca-P phases in SIs were a typical by-product of the indirect process, and sample 23045 has phases containing more than 20wt% of both CaO and P_2O_5, this

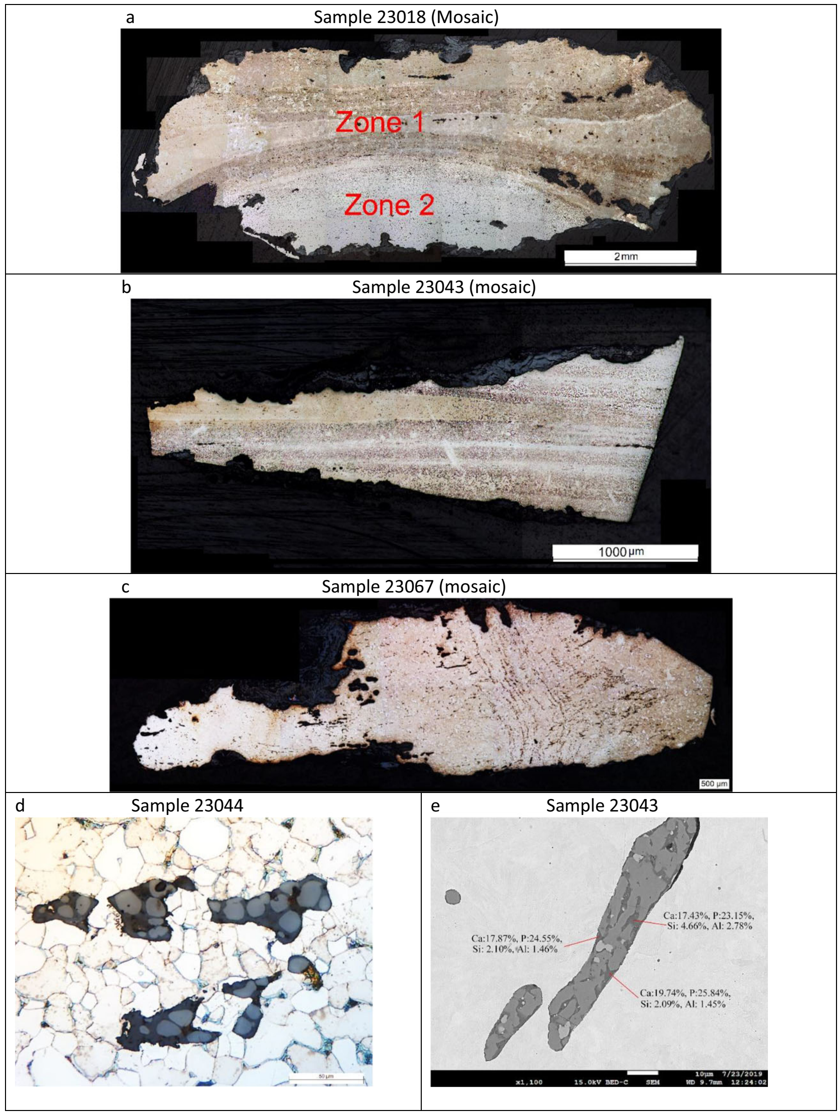

Fig. 6 Photomicrographs and SEM images of selected samples. **a** A welding structure. Zone 1 is pearlite + ferrite containing relatively dense SIs. Zone 2 shows ferrite but only includes relatively few SIs; **b** Multiple layers of mild-carbon steel with small, elongated SIs. The layer structure in the microstructure indicates that the sample was folded multiple times during the manufacturing process; **c** Pearlite + ferrite with elongated SIs. The orientation of SIs in the middle (vertical) is different from that on both sides (horizontal). The samples might have been welded by multiple pieces, but no clear welding line is present; **d** Ferrite with SIs characterized by sub-double phase inclusions; **e** A SEM (backscattered) image showing an elongated SI in 23067. The phases pointed by the red lines contain relatively high Ca and P content, as showed by analytical results

sample is more likely to be associated with the indirect process, hence possible fined iron. For samples 23018 and 23044, even though certain phases or features contain relatively high wt% of CaO and P_2O_5 (both ≥ 10 wt% but below 15 wt%), we would suggest identifying these two samples as "undetermined." As we explained above, since furnace ceramics, fluxes, and fuel ash could all contribute to the formation of SIs in an object, more empirical studies are needed to investigate whether the multivariate framework of NRC ratios in SIs could be employed in the study of iron technology in ancient China as successfully as it was in the study of the ironmaking in medieval French (Disser et al. 2014). In this case study, the two samples (23043 and 23045) that yielded "contradictive" results between the multi-variate approach and the identification of high Ca-P phases undergone forging to a considerable extent during the smithing process. The impurities entered into the metallic body or fuel ash would have generated blurring effects on the chemical compositions of SIs collected by bulk analyses. If the identification of high Ca-P phases is a more important indicator of the indirect process, especially in the types of iron objects that were heavily forged or made by welding, then, our identification results would further suggest that the indirect process was the dominant technique used in the manufacture of iron implements in the Guangzhou assemblage.

Comparison of SIs in suspected direct-process objects from Guangzhou and smelting slag from Guangxi

We suggested above that there is a high probability that two samples (23048 and 23050) were made by the direct process. This section will compare our samples' NRC ratios in SIs with those of smelting slag from Guangxi published in the literature (Huang and Li 2011, 2012a, b; Huang 2013; Huang et al. 2014), in order to test whether the bloomery iron from

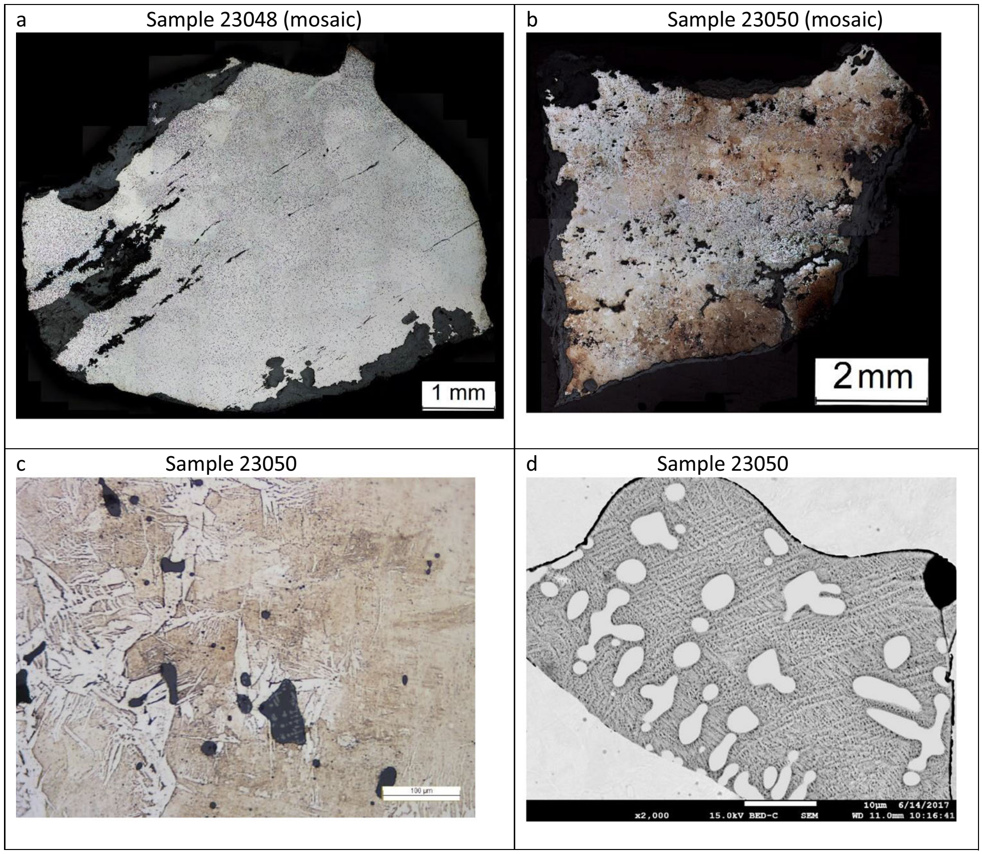

Fig. 7 Optical photomicrographs of iron objects relating to direct process manufacturing. **a** Characterized by ferrite + pearlite with elongated SIs. Carbon content is higher in the upper part than the lower part in the image; **b** Pearlite + ferrite but the carbon content is highly heterogeneous in the cross-section. Also, SIs are irregular in shape and not deformed; **c** Pearlite with ferrite, and widmanstätten structures were embedded within the grain structure; **d** A SEM (backscattered) image of a SI in 23050. The light phase is dendritic wüstite within the fayalitic matrix (dark grey phase)

Guangzhou might have originated in the smelting systems found in the same broad geographic region.

For the purpose of provenance analysis, we first compared the ratios of Al_2O_3/SiO_2, Al_2O_3/MgO, Al_2O_3/K_2O, and Al_2O_3/CaO between smelting slag and the groups of SIs in the two objects that we identified as more relevant to smelting and employed in the multi-variate analysis (Fig. 10a, b). Since the NRC ratios of some slag are much larger than other slag samples, which makes the ratio differences compressed, all ratios in the bivariate plots were plotted on a logarithmic scale. It is noteworthy that samples from most locales, such as PNPZ and PNTS, varied to a certain extent, which might be attributable to the chronological differences between these smelting sites, source variations, or even the different smelting systems employed. Also, in the bivariate plots of Al_2O_3, SiO_2, CaO, and MgO, some samples from LTS, LMH, PNPZ, PNTS, and PNLX show ratios that were higher than the SIs of the iron objects (i.e., plotted in top right quarter defined by the selected groups of SIs of the two objects) (Fig. 10a, b). In other words, the SIs in the two bloomery iron from Guangzhou are possibly derived from the smelting system represented by slag from these locales.

To further test the hypothetical link, the ratios of MgO/K_2O and SiO_2/MnO in the two datasets were compared (Fig. 10c). As with Fig. 10a and b, the NRC ratios of smelting slag were very diverse and scattered (Fig. 10c). Also, the ratios of MgO, K_2O, SiO_2, and MnO in samples 23048 and 23050 were generally different from those of the majority of smelting slag tested in the Guiping-Pingnan area of Guangxi. But, as the ratios of MgO/ K_2O and SiO_2/MnO in SIs in the two bloomery iron objects are relatively matched, these two objects might have come from a similar smelting system. In terms of the ratios of MgO/K_2O and SiO_2/MnO, it is noteworthy that only one sample from the LTS locale has a result relatively consistent with one sample (23048) from Guangzhou. In contrast, all samples from the LMH, PNPZ, PNTS, and PNLX locales had ratios relatively separate from those of the two iron objects. Therefore, the SIs in the iron objects were unlikely to be derived from the smelting system used at the four locales mentioned above. Also, even though one out of the four slag samples from LTS is consistent with the MgO/K2O and SiO2/MnO ratios of 23048, the ratios of Al_2O_3, SiO_2, CaO, and MgO of this sample in the bivariate plots (Fig. 10a, b) are not higher than the SIs in the iron objects. In supplement C, we present the comparison between all SIs in the two objects and smelting slag in the same three bivariate plots. The results are basically the same, i.e., the SIs in the two objects did not fully match the criteria to indicate that they might have come from the smelting systems represented by any slag from the Guiping-Pingnan area.

As we note before, furnace ceramics, fuel ash, fluxes, and smithing process would all contribute to the variabilities in the

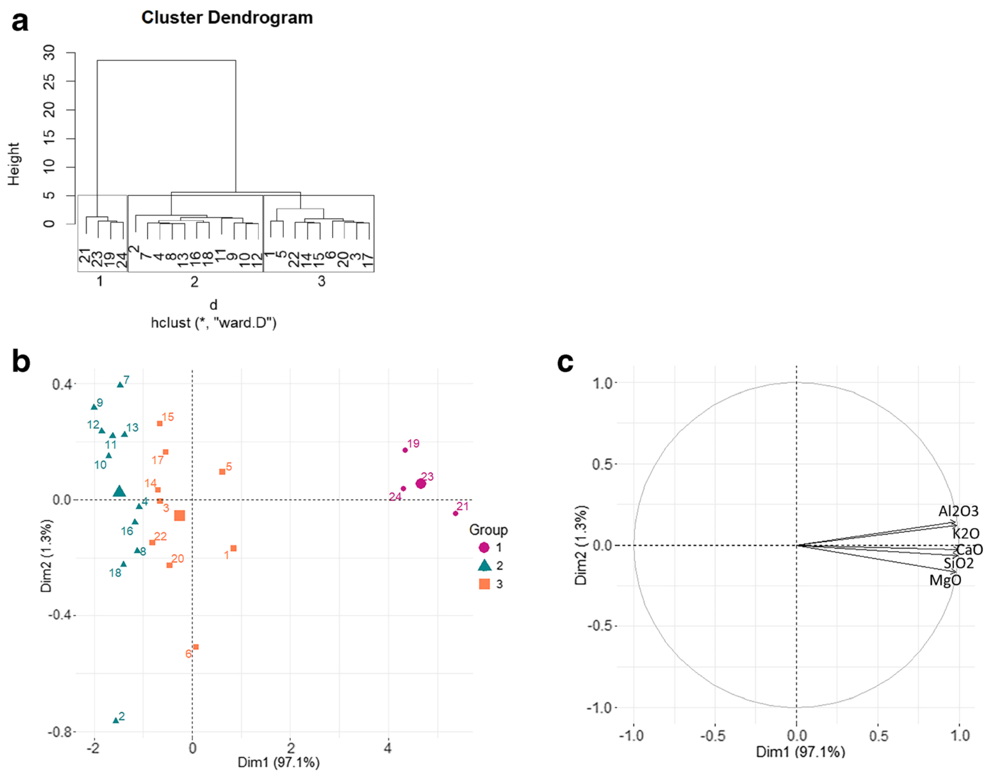

Fig. 8 Pre-treatment of chemical compositions of SIs in 23050 employing statistic methods (WARD and PCA). **a** Dendrogram of hierarchical clustering of XiNRC in 23050 following the WARD method and instructions as presented in Disser et al. 2014; **b** PCA of SIs based on the groupings by WARD method; **c** Variable correction plot that shows the relationship of all variables in the PCA analysis

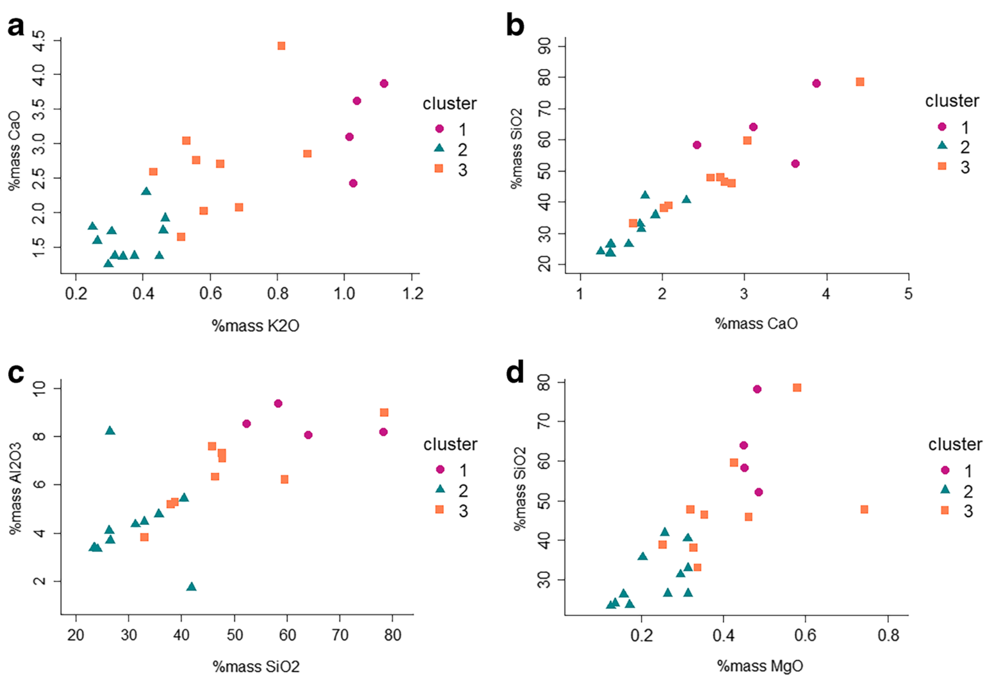

Fig. 9 Bivariate plots of various NRC couples in SIs of sample 23050 according to Ward grouping result in Fig. 8

Table 5 Average chemical compositions (wt%) of the cluster of SIs in samples with dense SIs for multivariate statistical analysis and prediction of iron-making process for samples based on logit(p).

	MgO*	Al_2O_3*	SiO_2*	P_2O_5*	K_2O*	CaO*	MnO*	FeO*	Logit(p) LOG ODDS	Probability of indirect process	Probability of direct process	Results
23018 (zone1, cluster 1)	1.64	8.17	60.52	0.94	3.51	6.04	0.83	17.43	− 5.68		> 0.99	Direct process
23018 (zone2)	0.65	3.03	26.43	4.23	1.21	2.43	1.27	59.73	− 1.59	0.17	0.83	Direct process
23043 (cluster 3)	0.64	5.07	25.34	4.85	1.14	2.03	13.75	46.11	− 6.20		> 0.99	Direct process
23044 (cluster 3)	0.37	2.74	12.27	0.52	0.20	0.65	0.16	82.49	− 9.43		> 0.99	Direct process
23045(1) (cluster 1)	2.89	8.59	33.02	1.63	5.51	7.20	0.49	39.46	− 13.41		> 0.99	Direct process
23045(2)	2.47	5.79	33.99	1.51	3.96	5.18	0.36	44.87	− 8.35		> 0.99	Direct process
23048 (cluster 2)	0.44	5.01	14.12	0.97	0.58	2.29	0.12	75.83	− 14.32		> 0.99	Direct process
23050 (cluster 2)	0.23	4.26	30.26	1.60	0.34	1.62	0.33	60.09	− 4.22	0.01	0.99	Direct process
23067 (cluster 3)	0.55	1.87	9.55	12.89	0.58	1.97	0.60	71.83	2.58	0.93	0.07	Indirect process

Detection limit was empirically determined to be 0.1 wt%. "*" after the average chemical composition indicates that it is below the detection threshold. For original data, see Supplement B

chemical compositions of iron slag. Blakelock et al.'s study (2009, 1741) pointed out that even the two blooms from the same smelting system but worked by two smiths employing different fuel would potentially result in different SI signatures. In addition, the heterogeneity of chemical compositions might have existed in different area of the same piece of slag that was not fully molten (Humphris et al. 2009, 368). Given the wide range of compositional variations evidenced in the case of bloomery slag from the Guiping-Pingnan area, any strong conclusion has to be drawn based on a careful design of sample collection and considerable number of samples analyzed. By considering the analytical uncertainties and the small amount of slag samples from the research area available at this stage, more samples, including various types of slag from production sites and blommery iron products from settlements, have to be collected for more in-depth analyses in the future. However, since no significant pattern could be identified at this point, the comparison of NRC ratios did not empirically support the hypothesis that the Guangzhou iron objects could have been manufactured using any of the known and previously tested smelting systems in the Guiping-Pingnan area.

Discussion

The overall manufacturing technology reflected by the Guangzhou iron assemblage

Based on the object types selected for this research and discussed in other published data (e.g., Guangzhou and Guangzhou 1981), this typical range of iron artifacts in the regional assemblage usually includes ring-pommeled knives, swords, axes, chisels, vessels (e.g., caldrons), and coffin nails. Beyond the caldron stand that might belong to local tradition (Bai 2005), the types of iron objects show a high degree of similarity with those found in the Central Plains. By just looking at the shape and typology of artifacts, no obvious regional contrasts could be observed between those in Lingnan and the Central Plains. Therefore, metallurgical analysis is essential for examining the manufacturing techniques and potential origins of these iron objects.

Even though the selected objects were heavily corroded, the metallographic study identified that at least three objects were made by solid-state decarburization of cast iron and there

Table 6 Counts of SEM-EDS points in SIs with high wt% P_2O_5 and wt% CaO

Sample#	Total points of spots (in phases or crystals) analyzed by SEM-EDS	wt% P_2O_5 and wt% CaO ≥ 5%	wt% P_2O_5 and wt% CaO ≥ 10%	wt% P_2O_5 and wt% CaO ≥ 15%	wt% P_2O_5 and wt% CaO ≥ 20%	Final identification result
23018	80	8	1	0	0	Undetermined
23043	115	18	9	3	1	Possibly indirect process
23044	168	19	4	0	0	Undetermined
23045	209	6	1	1	1	Possibly indirect process
23048	124	0	0	0	0	Direct process
23050	48	0	0	0	0	Direct process
23067	37	11	9	3	0	Indirect process

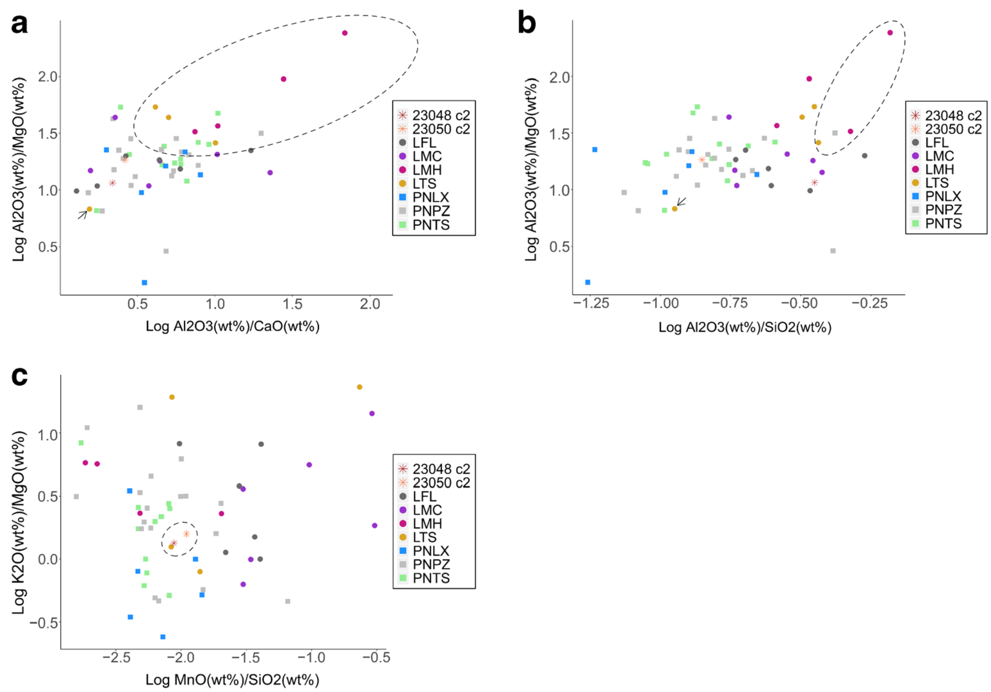

Fig. 10 Bivariate plots of NRC of SIs in bloomery iron (23048 cluster 2 and 23050 cluster 2) from Guangzhou and of smelting slag from the Guiping-Pingnan area. The ratios were plotted on a logarithmic scale, since the NRC ratios of some slag are much larger than other slag samples, making the ratio differences compressed. Plotted ratios of smelting slag within the dotted circles in (**a**) and (**b**) were higher than those in the two iron objects from Guangzhou. The sample from LTS location pointed by an arrowhead in (**a**) and (**b**) is the data-point consistent to the ratio of SIs in 23048 in (**c**). Data of smelting sites are derived from Huang and Li 2011, 2012a, b; Huang et al. 2014

was one potential cast iron object among samples with no SIs. While more experimental studies are needed to further confirm the effectiveness of the multi-variate approach and make it fully applicable in the case of ironmaking in ancient China, as we noted before, the using of this approach together with the spot analyses of high Ca-P phases would be conceived as a rather reliable methodology to differentiate direct and indirect processes. Among the seven samples with SIs, the multi-variate approach and spot analyses show that one (23067) was identified as fined iron, and two (23048, 23050) were made of bloomery iron. In addition, among the four other examples with SIs, we would suggest that two of them (23043, 23045) are possibly fined iron objects, due to the relatively high Ca-P content in the SIs.

Our study of the iron technology used to produce the objects found in Guangzhou indicates that it was dominated by cast iron and related steel-making technology, which is a pattern similar to that of previous studies of iron technology in the Central Plains during the Han period (e.g., Lam et al. 2018). This pattern of cast iron and fined iron being found together with bloomery iron also matches a previous preliminary study of three iron objects from Han tombs in the Guiping region of Guangxi (Huang et al. 2016b), which identified a cast iron caldron, a fined iron sword, and a second sword made of bloomery iron. It is also noteworthy that the techniques used to manufacture typical Han-style iron objects found in Guangzhou were similar to those used to make objects of the same type found before. For instance, sample 23016 (ring-pommel knife) belongs to the most commonly occurring category of iron objects found in Han tombs and was made by solid-state decarburization of cast iron, which reflects the analytical results of ring-pommel knives found elsewhere (Lam et al. 2018; Liu et al. 2014). However, as noted above, no cast iron foundries were discovered during previous archaeological works near main urban centers in the Lingnan region. Metallurgical studies, therefore, suggest that Lingnan relied heavily on the external supply of objects that were made of cast iron technology, thus indicating intensive connections between Lingnan and other parts of the Han Empire.

Bloomery iron objects and their potential provenance

In this study, two samples with SIs were identified as being probably made of bloomery iron. The two bloomery iron objects were an axe (23048) and a tripod stand (23050) for a bronze caldron. The bloomery iron axe looks similar to another in the Guangzhou assemblage of objects that was made from the solid-state decarburization of cast iron. Since these two axes also look relatively similar to other Han period examples, it is difficult to determine whether they were locally made products. In contrast, in a previous study of Han period ironware, it was noted that iron tripod stands are an indigenous style of objects which were commonly found in the southern and southwestern peripheries of the Han Empire (i.e., Lingnan and Yungui plateau) (Li and Zhou 2014). More sampling is needed to confirm if the use of bloomery iron was related to object types that used on the peripheries of the Han Empire, but the discovery of these two iron objects at least shows that some iron objects might come from ironworks different from those that manufactured and supplied cast iron objects to Guangzhou.

As bloomery iron was identified, this immediately raised the question of whether it had been produced in the Lingnan-based blommeries of the Guiping-Pingnan area. Based on the comparison of NRCs ratios between smelting slag sample and SIs in bloomery iron objects from Guangzhou, we cannot confirm that these products were from the smelting system in the Guiping-Pingnan area of Guangxi; no slag sample from the whole slag collection from seven locales could match the criteria set out above. However, it is noteworthy that, besides the small sample-size of slag published in the literature, our knowledge about the ore sources in the Guiping-Pingnan area is also limited, yet Dillmann and his team's work already showed the importance of identifying the compositions of ores in the provenance study of iron (Desaulty et al. 2009). Also, we have to acknowledge that the chronology resolution of these locales in the area is wide, and these locales collectively cover a period of more than seven centuries (Huang and Li 2011). Due to the lack of precise chronological study of each of these locales, it is impossible to firmly date the iron production to the Han period. Without more supporting evidence, even a match between NRC ratios of SIs in iron objects from Guangzhou and the slag samples from Guangxi smelting site clusters would be just a coincidence, instead of a clear provenance link between these two areas.

Even though bloomery iron could have been locally manufactured in Lingnan during the Eastern Han period, one cannot rule out the possibility that the bloomery iron objects identified in Guangzhou were imports. In previous studies, although cast iron was often the dominant technique evidenced in Central Plains' iron assemblages, bloomery iron was also occasionally discovered in cemeteries there, as well as in mausoleums in various other places (e.g., see such finds published in Beijing and Xuzhou 1997). As we explained before, bloomery iron could have been a "technological choice" in places where skilled labor resources, ores, or fuel were scare (Larreina et al. 2018). Ironworks outside of Lingnan, especially those to the South of the Yangtze river valley, might also have turned to bloomery iron production as a way of overcoming certain technical limitations in the region. On the other hand, even if the bloomery iron objects were locally manufactured in Lingnan, the fact that these two objects represented a small proportion of the assemblage selected in this study suggests that the provision of daily ironware in Guangzhou must have relied more upon external sources.

The transportation of iron implements and Han's control in the Lingnan region

Through metallurgical analysis, this study has demonstrated that the majority of iron objects from Guangzhou were made of cast iron or related steel-making techniques, and that the supply of iron objects relied heavily on external sources. It thus seems reasonable to suggest that the objects identified or the semi-raw materials for making these objects were probably procured via exchange or migrations from sources outside Lingnan. Although this study focuses only on iron objects from Guangzhou, the manufacturing techniques identified through this study indicate that the importation of iron implements from workshops outside Lingnan was the key way through which this important center met its population's demand for iron weapons, agricultural tools, and daily utensils.

The transportation of iron implements to Lingnan might have occurred in various ways. In the Han period, the development of trading ports undoubtedly attracted the migration of merchants from other regions. In addition, Hepu, which was a major port in Lingnan, was often mentioned as a place to which political criminals were exiled. Some personal artifacts, such as ring-pommel knives and even swords, might have been brought into Guangzhou alongside such migrations from the North. While migrations would bring some iron implements into the region, the importance of market exchange or transportation should not be underestimated. For instance, the axe (23042) might have been brought into Guangzhou as either a commercial product or state-controlled item, rather than being a personal item, as agricultural or craft tools of this type were widely used and produced on a large-scale. Our recent study of the distribution of metal objects in Lingnan during the Han period indicates that the transportation network through the Pearl river system was relatively well developed (Lam et al. 2019), although it appears to have been less well integrated when compared with its counterpart in the capital region in Shaanxi (Lam 2020). As the expansion of the Han state occurred alongside the development of interregional and intraregional communications, the identification of cast iron and fined iron in Guangzhou supports the idea that a transportation network developed that interconnected the core or inland areas with southern peripheries for the transportation of mundane, high-volume daily items. With more studies of iron objects and manufacturing waste within the region in the future, we may be able to further clarify the connections between local iron production sites and ironworks outside the region, thus allowing us to attempt a reconstruction of the mechanism underpinning the widespread distribution of Han manufactured goods.

Conclusion

The production of ironware, comprising mainly agricultural tools and equipment, daily utensils, and weapons, was essential to the Han economic system, and even underpinned the political authority of the Han state. But, the means by which iron implements were manufactured in or transported to the

Lingnan region, and other peripheral regions, has not been fully examined. Both metallographic and chemical compositional studies suggest that steel decarburized from solid-state cast iron, fined iron, cast iron, and bloomery iron were all employed in the manufacturing of iron objects found in Guangzhou. The widespread distribution of objects made using cast iron technology in peripheral centers such as Guangzhou should be attributed not only to the expansion of the Han state and associated migrations, but also to the development of the supply network outside Lingnan which, despite the limitations of contemporary transportation technology, ensured the steady flow of goods into the region.

The discovery of bloomery iron objects in Guangzhou has confirmed previous suggestions that bloomery smelting might have existed in the Han peripheries. Even though the provenance link between the bloomery iron objects found in Guangzhou and smelting sites in the Guiping-Pingnan area of Guangxi still requires further research and substantiation, our findings nevertheless raise some critical questions for those conducting further exploration of the Han iron industry, such as the potential for bloomery iron production in other ironworks dating to the Han period. If more samples from other centers in Lingnan are analyzed in the future, it should assist in further clarifying the techniques employed in the manufacturing of iron implements in the Han peripheries. Moreover, such studies should help illustrate the roles played by exchange, transportation, and potential local production in the supply of one of the most critical materials contributing to the economic development in the Han period.

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References

Bai Y (2005) Xianqin lianghan tieqi de kaoguxue yanjiu [Archaeological study on iron works before 3rd Century A.D. in China]. Kexue chubanshe, Beijing.

Beijing & Xuzhou [Beijing Keji Daxue Yajingshi Yanjiusuo, Xuzhou Hanbingmayong Bowuguan] (1997) Xuzhou Shizishan Chuwangling chutu tieqi de jingxiang shiyan yanjiu [Metallurgical report on the analysis of iron objects from the mausoleum of the Chu state at Shizishan in Xuzhou]. Wenwu 1997(7):146–156

Beijing [Beijing Keji Daxue Yejinshi Yanjiushi] (1991) Xihan Nanyue wangmu chutu tieqi jianding baogao [Identification report of iron objects from the Mausoleum of the Nanyue Kingdom]. In: Guangzhoushi wenwu guanli weiyuanhui (ed) The Mausoleum of the Nanyue Kingdom of the Western Han Period. Wenwu chubanshe, Beijing, pp 389–396

Blakelock E, Martinon-Torres M, Veldhuijzen HA, Tim Y (2009) Slag inclusions in iron objects and the quest for provenance: an experiment and a case study. J Archaeol Sci 36:1745–1757

Brindley EF (2015) Ancient China and the Yue: Perceptions and identities on the southern frontier, c.400 BCE–50 CE. Cambridge University Press, Cambridge

Charlton MF, Blakelock E, Martinón-Torres M, Young T (2012) Investigating the production provenance of iron artifacts with multivariate methods. J Archaeol Sci 39:2280–2293

Chen J (2014) Zhongguo gudai jinshu yezhu wenming xingtian [Exploration of the metal smelting and casting civilization in ancient China]. Kexue chubanshe, Beijing

Chen J, Han R (2007) Hanjin Zhongyuan Ji Beifang Diqu Gangtie Jishu Yanjiu [Iron and Steel technology in Central Plain and Northern Region during the Han and Jin Dynasties]. Beijing Daxue Press, Beijing

Chen J, Zhang Z (2016) Jiyu luzha fengxi de gudai chaogan jishu panduan wenti [Identification of fined iron techniques based upon metallurgical analysis of slag]. Nanfeng Wenwu 2016(1):115–121

Chen J, Huang Q, Li Y, Han R (2008a) Kezhang kele muzang chutu tieqi de jinxiang shiyan yanjiu [Metallurgical study of iron artifacts from burials at Kele, Kezhang]. In: Guizhousheng wenwu kaogu yanjiushou (ed) Kezhang Kele-Site Report of 2000. Wenwu chubanshe, Beijing, pp 195–206

Chen J, Yang C, Zhang H, Lin F (2008b) Fujian Wuyishancun hancheng chutu tieqi de jinxiang shiyan yanjiu [Metallurgical analysis of iron objects from Cuncheng walled-town in Wuyishan, Fujian]. Wenwu 2008(3):88–96

Chen J, Hong Q, Qin Z, Liu H, Han R (2011) Lushan Wanchenggang yetie yezhi de yelian jishu chubu yanjiu [Preliminary study on the smelting and metling techniques based on remains from the Wanchenggang iron production site, Lushan]. Huaxia Kaogu 2011(3):99–108

Crew P (2000) The influence of clay and charcoal ash on bloomery slags. In Tizzoni, C.C., Tizzoni, M. (Eds.). Iron in the Alps: deposits, mines and metallurgy from antiquity to the XVI Century, Bienno (BS), pp. 38-48.

Desaulty AM, Dillmann P, L'Héritiera M, Mariet C, Gratuze B, Joron JL, Fluzin P (2009) Does it come from the Pays de Bray? Examination of an origin hypothesis for the ferrous reinforcements used in French medieval churches using major and trace element analyses. J Archaeol Sci 36:2445–2462

Dillmann P, L'Héritier M (2007) Slag inclusion analyses for studying ferrous alloys employed in French medieval buildings: supply of materials and diffusion of smelting processes. J. Archaeol. Sci. 34: 1810–1823

Disser A, Dillmann P, Bourgain C, L'Héritier M, Vega E, Bauvais S, Leroy M (2014) Iron reinforcements in Beauvais and Metz Cathedrals: from bloomery or finery? The use of logistic regression for differentiating smelting processes. J. Archaeol. Sci. 42:315–333

Du N, Li J, Zhang G, Wang X, Li Y (2011) Shandong Lizi Qiguo gucheng dongbeibu yetie yizhi de diaocha yu yanjiu [Survey and research on iron production sites in the northeastern part of the capital city of the Qi State in Linzi, Shandong]. Jiangxi ligong daxue xuebao 32(6):12–15

Du N, Li Y, Zhang G, Wang X, Li J (2012) Linzi gucheng nanbu liangtie yiwu yanjiu [Study of ancient iron smelting relics in the southern part of the capital city Linzi]. Zhongguo kuangye 21(12):115–120

Guangdong [Guangdongsheng Difangshizhi Bianweihui] (1994) Guangdong shengzhi. dizhi kuangchanzhi [Guangdong chorography: volume of geology and mineral resources]. Guangdong renming chubanshe, Guangzhou

Guangxi [Guangxi Zhuangzu Zizhiqu Bowuguan] (1988) Guangxi guixiang Luobowan Hanmu [The Han Tomb at Luobowan in Guixiang, Guangxi]. Wenwu chubanshe, Beijing

Guangxi [Guangxi Zhuangzu Zizhiqu Difangzhiza Bianweihui] (1992) Guangxi tongzhi. Dizhizhi [Guangxi chorography. Geology volume]. Guangxi renming chubanshe, Nanning.

Guangzhou & Guangzhou [Guangzhoushi Wenwu Guanli Weiyuanhui, Guangzhoushi Bowuguan] (1981) Guangzhou Hanmu [Guangzhou Han Tombs]. Wenwu chubanshe, Beijing

Guangzhou & Guangzhou [Guangzhoushi Wenwu Kaogu Yanjiusuo, Guangzhoushi Panyuqu Wenguanhui Bangongshi] (2006) Panyu Hanmu [Panyu Han Tombs]. Kexue chubanshe, Beijing

Guangzhou [Guangzhoshi Wenwu Kaogu Yanjiusuo] (1998) Guangzhou Shidaigang Xihan tumu fajue jiangbao [Preliminary report on the excavation of Western Han shaft-earth tombs at Shidaigang, Guangzhou]. In Collection of archaeological articles related to Guangzhou. Wenwu chubanshe, Beijing, pp 207–212

Guangzhou [Guangzhoshi Wenwu Kaogu Yanjiusuo] (2003) Guangzhoushi Hengzhigang xihanmu de qingli [Excavation of the Western Han tombs at Hengzhigang in Guangzhou]. Kaogu 2003(5):35–43

Guangzhou [Guangzhoushi Wenwu Kaogu Yanjiusuo] (2003) Guangzhou Haifusi Handai yaochang yizhi de fajue [Excavations on the Han period kiln site at the Haifu temple, Guangzhou]. Kaogu xuebao 2003(3):373–400

Guangzhou [Guangzhoushi Wenwu Kaogu Yanjiusuo] (2004) Guangzhou Huanghuagang Hantang muzang fajue baogao [Excavation of Han-Tang periods tombs at Huanghuagang, Guangzhou]. Kaogu xuebao 2004(4):451–484

Han R (1987) Jilin Yushu Laoheshen Xianbei muzang chutu jinshu wenwu de yanjiu [Reseach on the metal objects from Xianbei tombs at the Laoheshen site in Yushu, Jilin]. In: Jilinsheng wenwu kaogu yanjiushuo (ed) The Laoheshen Site in Yushu. Wenwu chubanshe, Beijing, pp 146–156

Han R, Chen J (2013) Casting Iron in Ancient China. In: Humphris J, Rehren T (eds) The World of iron. Archetype publications, London, pp 168–177

Han R, Ke J (2007) Zhongguo kexue jishushi (kuangyejuan) [China Science and Technology History(volume of mining and smelting)]. Science Press, Beijing

Hanshu [Book of Former Han by Ban Gu] (1997) Annotated by Yan Shi. Zhonghua Shuju, Beijing

Hou Hanshu [Book of Later Han by Song Ye] (1965) Annotated by Li X. Zhonghua Shuju, Beijing

Huang Q (2013) Guangxi Guigang diqu gudai yetie yizhi diaocha ji luzha yanjiu [Surveys of iron smelting sites in the Guigang Region of Guangxi and study of slag remains]. Lijiang chubanshe, Guilin

Huang Z (2015) Nanyueguo kaoguxue yanjiu [Archaeological research of the Nanyue Kingdom]. Zhongguo shehui kexue chubanshe, Beijing

Huang Q, Li Y (2011) Guangxi Pingnan Liuchen pozui yizhi yelian jisu yanjiu [Study on Pozui iron smelting site in Pingnan County of Guangxi]. Youse jinshu. 63(1):140–146

Huang Q, Li Y (2012a) Guangxi Guiping Luoxiu gudai yetie yizhiqu chubu yanjiu [Preliminary studies on ancient iron smelting sites in Guiping city, Guangxi]. Zhongguo kejishi zazhi 33(4):485–494

Huang Q, Li Y (2012b) Guangxi Pingnanxian tieshitang yelian yizhi chubu yanjiu [Preliminary research on smelting sites at Tieshiang in Pingnan county, Guangxi]. Sichuan wenwu 2012(1):92–96

Huang Q, Li Y, Chen J, Gong, H (2014) Guangxi Guigang Liuxueling handai yetie yizhi chubu yanjiu [A preliminary study of the Han dynasty iron-smelting site of Guigang Liuxueling]. Nanfang minzu kaogu (10), Kexue chubanshe, Beijing, pp 231-238.

Huang Q, Li Y, Chen J, Tie F (2016a) Yi luzha fenxi weizhu jieshi gudai liantie jishu de yanjiu yu tansuo [A summary of ancient iron smelting technology based on iron slag analysis]. Zhongguo guojia bowuguan guankan 2016(11):145–153

Huang Q, Li Y, Zhang C, Chen J, Feng G (2016b) Guangxi zhanguo handai muzhan chutu tieqi de kexue yanjiu [The scientific research of the iron unearthed in the tombs of the Warring States period, Han dynasty, Guangxi]. Nanfang Wenwu 2016(1):109–114

Hulsewe AFP (1985) Remnants of Ch'in Law. Brill, Leiden

Humphris J, Martinón-Torres M, Rehren T, Reid A (2009) Variability in single smelting episodes—a pilot study using iron slag from Uganda. J. Archaeol. Sci. 36:359–369

Lam W (2014) Everything old is new again? Rethinking the transition to the cast iron production in the central plains of China. J. Anthropol. Res. 70(4):511–542

Lam W (2020) Integration and the regional market system in the Early Chinese empires: a case study of the distribution of iron and bronze objects in the Wei river valley. Asian Perspectives 59(1):117–158

Lam W, Chen J (2017) Dongzhou shiqi tieqi jisu yu gongye de diyuxing chayi [On the regional variation of iron technology and iron industry during the Eastern Zhou period]. Nanfang wenwu 2017(3):98–106

Lam W, Chen J, Chong J, Lei X, Tam W (2018) An iron production and exchange system at the center of the Western Han Empire: scientific study of iron products and manufacturing remains from the Taicheng site complex. J. Archaeol. Sci. 100:88–101

Lam W, Zhang Q, Chen J (2019) Handai Lingnan tieqi de chuanbo yu yunshu fangshi: yi muzang suizang tieqi de kongjian fenxi wei shijiao [Spread and transportation of iron implements in Lingnan during the Han period: a perspective from the spatial analysis of iron implements contained in tombs]. Nanfang wenwu 2019(3):122–135

Larreina D, Li Y, Liu Y, Martinón-Torres M (2018) Bloomery iron smelting in the Daye County (Hubei): technological traditions in Qing China. Archaeol. Res. in Asia 16:148–165

Li J (1995) Nanyang Haidai yetie [Iron production during the Han period in Nanyang]. Zhongzhou Guji Press, Zhengzhou

Li X (2011) Gu Dianguo Jinshu Jishu Yanjiu [Study of the metallurgical techniques in the Ancient Dian State]. Kexue Chubanshe, Beijing

Li Y, Zhou L (2014) Yungui gaoyuan chutu zhanguo shiqi tieqi yanjiu [A study of ironwares unearthed from the Yunnan-Guizhou Plateau during Qin-Han period]. Jianghan Kaogu 2014(6)

Li Y, Li Y, Yuan H, Juleff G, Zhang M (2018) Early iron objects of southwest China: a case stdy of iron objects excavated from Qiaogoutou cemetery site, Sichuan Province. Archaeol. Anthropol. Sci. 11:1187–1198

Li Y, Ma C, Juleff G, Murakami Y, Zhou Z, Li Y (2019) Microstructural and elemental analyses of slags excavated from Xuxiebian iron smelting site, Sichuan, China. Archaeometry 61(6):1353–1365

Liu X (2014) Lidai wuling jiaotong geju de bianqian [The evolution of Wuling route]. Gouxue xuekan 2014(1):80–90

Liu X (2017) Xianqin lianghan nongye yu xiangcun juluo de kaoguxue yanjiu [Archaeological research on the agriculture and village settlement of the Pre-Qin and Han Periods]. Wenwu chubanshe, Beijing

Liu H, Chen J, Mei J, Jia J, Shi L (2014) A view of iron and steel making technology in the Yan region during the Warring States period and the Han dynasty: scientific study of iron objects excavated from Dongheishan site, Hebei province. China. J. Archaeol. Sci. 47:53–63

Liu Y, Martinón-Torres M, Chen J, Sun W, Chen K (2019) Iron decarburisation techniques in the eastern Guanzhong Plain, China, during Late Warring States period: an investigation based on slag inclusion analyses. Archaeol. and Anthropl. Sci. 11:6537–6549

Meng C, Zou G (2017) Guangxi Wuzhou liuzhao yetie yizhi chutian [Study on iron smelting sites of Six Dynasties in Wuzhou city Guangxi]. Jinsu shijie 192(4):11–17

Percy J (1864) Metallurgy: the Art of extracting metals from their ores, and adapting them to various purposes of manufacture, Volume 2, Iron; steel. John Murray, London

Qu Y (2009) Donghan shiqi tieqi zai Lingnan diqu de tuiguang shiyong jiqi shehui dongying [On the populization of using iron implements in Lingnan during the Eastern Han period and its social motivation]. Guangxi shehui kexue 2009(10):84–87

Shaanxi [Shaanxisheng Kaogu Yanjiuyuan] (2018) Taicheng zhutie: Shaanxi Yangling Handai zhutie yizhi fajue yu yanjiu [Taicheng ironworks: report on the excavation and research of a cast iron foundry of the Han Period in Yangling, Shaanxi]. Shanghai guji chubanxue, Shanghai

Shiji [Records of Grand Historian by Shima Qian] (1997) Annotated by Zhang S, Shima Z, and Pei Y. Zhonghua Shuju, Beijing

Török B, Thiele A (2013) Extracting phosphoric iron under laboratorial conditions smelting bog iron ores. IOP Conf. Ser. Mater. Sci. Eng. 47(012034)

Wagner DB (1993) Iron and steel in Ancient China. Brill, Leiden

Wagner DB (2008) Science and civilization in China: vol.5, Chemistry and Chemical technology, part 11. Ferrous Metallurgy. Cambridge University Press, Cambridge.

Wang Z (2014) Qinhan "wuling" jiaotong yu "nanbian" xingzheng [Traffic communication crossing "Wuling" during the Qin-Han periods and administration in southern broader]. Zhongguoshi yanjiu 2014(3):17–21

Xiong Z (2014) The Hepu Han tombs and the maritime Silk Road of the Han Dynasty. Antiquity 88:1229–1243

Xiong Z (2015) Haidai hepugang kaogu yu haishang shichou zhilu [Archaeological discovery: the Hepu Port on the Maritime Silk Road of the Han Dynasty]. Wenwu chubanshe, Beijing

Xu H (1981) Handai Guangdong nongye shengchan chutian [Preliminary research on agricultural prodcution in Guangdong during the Han period]. Nongye kaogu 1981(4):56–60

Yang J, Li Y, Zhao F, Lou P (2014) Beijing Changping Mapaoquan shangcheng susuo yizhi chutu tieqi de shiyan yanjiu [Experimental studies on the iron objects unearthed from Mapaoquan site in Changping and a discussion of criteria for judging puddling steel]. Zhongguo kejishi zazhi 35(2):177–187

Zheng C (1996) Guanyu Lingnan yetieye qiyuan de ruogan wenti [On several questions related to the origin of the iron industry in Lingnan]. Guangxi minzu yanjiu 1996(3):50–56

Zhou W, Liu S, Liu H, Chen J (2014) Zhongguo chuantong ganguo lianqian zishu zhutian [A preliminary study of traditional Chinese crucible lead smelting technology]. Ziran kexueshi yanjiu 2014(2): 201–215