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Compositional analysis below the production region level: A case study of porcelain production at Dehua, Fujian, China

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ABSTRACT

Compositional analysis of Chinese porcelain often uses the production region as an analytical unit, whereas the possible compositional similarities and differences between different production loci within the region have rarely been considered. This research assesses the worth of conducting chemical composition analysis at the micro level and evaluates the effectiveness of combining laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) with portable X-ray fluorescence (pXRF) in characterizing compositional variation below the level of the production region. It focuses on porcelain production of the Song and Yuan dynasties (960–1368 CE) at Dehua, Fujian, China, where dozens of large-scale kilns produced enormous amounts of porcelains for export. A total of 19 kiln sites from five villages at Dehua were analyzed in this research. Results from both LA-ICP-MS and pXRF show that there are two distinct production groups at Dehua—Gaide and Longxun-Sanban. Geological differences and different ceramic-making traditions might both contribute to the distinct separation of the two compositional groups. Results from social network analysis further suggest that there are strong inter-kiln compositional similarities within the same production subregion, but kiln sites in the same village are more closely connected than those outside the village. These results demonstrate that the change of analytical scale in compositional analysis can provide more nuanced insights into the organization of production and the patterns of interaction between different production loci within the broader production region. In addition, this research shows that pXRF, though not as accurate as LA-ICP-MS, is capable of finding compositional patterning within a small region of ceramic production.

1. Introduction

Compositional analysis of material objects has long been used by archaeologists to find groups in the data (e.g., Baxter, 2001; Bishop et al., 1988; Bishop and Neff, 1989; Bishop et al., 1982; Harbottle, 1982; Neff et al., 1988). The identified compositional groups are often subsequently interpreted as evidence for different production or social units (Luke et al., 2006; Reents-Budet et al., 2000; Teoh et al., 2014). However, there is some disagreement about the level of production units that compositional groups can represent. Some scholars urged caution in assigning compositional groups to individual production units. Through extensive ethnoarchaeological studies in Mexico, Peru, and Guatemala, Arnold and his colleagues argued that compositional analysis is unlikely to differentiate between individual workshops below the level of the production community, even if these workshops used different clay sources that are less than 3–4 km apart (Arnold, 2000; Arnold et al., 1991; 1999; 2000). Rather than identifying individual production units, composition analysis might best reveal "community resource areas" or "community signature units," which refer to production communities that use raw materials within a limited range of probably no more than

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3–4 km (Arnold, 2000). Costin (2000:387) also supported the use of more neutral terms, like "resource-sharing" or "resource use" groups, to label compositional groups. On the other hand, some researchers are more sanguine about identifying workshops, individual artisans, or even single production events using compositional data. Stark et al. (2000) found that pottery made at two pottery-making villages that are only 2 km apart have distinct differences in paste composition. Adan-Bayewitz et al. (2009) were able to use chemical composition analysis to distinguish between ceramics of two close production locations (about 200 m apart) in Roman Galilee. Freestone et al. (2009a; 2009b; 2010) illustrated that individual batches of glass and ceramics can be recognized through compositional analysis and advocated the use of "the batch" as an analytical unit in attribution studies. Martinón-Torres et al. (2014) applied the concept of "the batch" to the study of the arrowheads from the Terracotta Army of China and discovered that different bundles of arrowheads represent discrete metal batches. Working on the Muisca votive goldwork of Colombia, Martinón-Torres et al. (2015) successfully recognized individual artisans through compositional analysis and concluded that compositional data could be used to identify individual artisans and single production events beneath the broader category of "provenance regions" or "technological traditions/styles."

Identifying individual production units or events is by no means easy because it not only requires the use of high-precision analytical techniques but also depends on the natural variability of raw materials as well as the social factors of raw material selection and procurement. The difficulties can also be compounded by "cumulative blurring" effects that increase sample variability (Blackman et al., 1993). However, the challenges of differentiating between workshops or individual artisans do not signify that compositional analysis should stop at the level of "community resource areas" or broader "provenance regions." On the contrary, compositional analysis at the micro level should be conducted whenever possible, particularly when dealing with objects from production loci. The discrimination of individual production units, if successful, would undoubtedly provide fine-scale reference groups for provenance or attribution studies and would allow "higher-resolution inferences about the organization of production that take us beyond the superficial ascription of products to generic production regions" (Martinón-Torres et al., 2014:549). Even if individual production units below the level of resource communities or provenance regions cannot be clearly differentiated based on chemical composition, compositional analysis of material objects from different loci of production would still provide a fine-grained perspective on production organization and the structure of interaction between different production units. Patterns in chemical signatures reflect the recipes and raw materials used during the production process, which are often part of the production secrets and reflect producers' cultural choices and practices (Neupert, 2000; Sillar and Tite, 2000; Stark et al., 2000; Underhill, 2003). Frequent interaction between producers would promote knowledge and technology transmission (Coto-Sarmiento et al., 2018; Roux, 2015) and lead to greater compositional similarities between different production loci. Ethnoarchaeological studies also showed that potters within a community usually select and use raw materials differently than other communities do (Arnold, 1980; 2000; Arnold et al., 1991). Compositional analysis of material objects at the micro level can thus provide a bottom-up approach to examine the structure of interaction between different production units and delineate the production community.

A high-resolution compositional study is particularly crucial for examining the large-scale, intense porcelain production in China. Massive kiln complexes (such as Jingdezhen, Longquan, Dehua, and Minqing)—each consisting of dozens or even hundreds of huge kilns—have been widely discovered in China (e.g., Ho, 2001; Li, 2008; Qin et al., 2015; Weng et al., 2017; Yang, 2016; Zeng, 2001; Zhejiang, 2005; Zheng et al., 2018; Fujian Museum, 2020; Jiangxi et al., 2020). Recently, compositional analysis has emerged as an important methodology for archaeologists endeavoring to understand the elemental characterization of porcelains from a single kiln complex (Cui et al., 2012; Duan et al., 2016; Li et al., 2012; Sang et al., 2019; Wu et al., 2020; Xu et al., 2017), to differentiate stylistically similar products from different kiln complexes (Huang et al., 2020; Leung et al., 2000b; Li et al., 2005; Li et al., 2006; Wang et al., 2020; Wang et al., 2018; Zhu et al., 2011; Zhu et al., 2010), and to source Chinese porcelains found overseas (Chen et al., 2016; Cui et al., 2016; Dias et al., 2013; Fischer and Hsieh, 2017; Xu et al., 2019; Zhou et al., 2019; Zhu et al., 2016). However, these studies usually used the production region or kiln complex, which refers to a cluster of kilns located within a specific geographic area (usually the area of a county), as an analytical unit and treated the elemental characterization of the analyzed kiln as being "representative" for the whole region without considering the possible compositional similarities and differences between different kiln sites. By stopping at the level of the broader production region, these compositional studies of Chinese ceramics mainly provided knowledge about "macroprovenience" (Rice, 1981:219), and were thus unable to address the issues of production organization and technological interactions between kilns within the broader kiln complexes or production regions.

This paper evaluates the worth of changing the analytical scale from the production region to the individual kiln in the compositional studies of Chinese porcelain production. It focuses on porcelain production of the Song and Yuan periods (960–1368 CE) at Dehua, a premier production center of export porcelain in southeast China. By analyzing porcelain samples from 19 kiln sites at Dehua using laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) and portable X-ray fluorescence (pXRF), this research seeks to examine the production organization and patterns of interaction between different kiln sites within the same production region.

2. Porcelain production at Dehua, Fujian, China

Dehua, a county located in an inland mountainous region of Quanzhou in southern Fujian (Fig. 1), quickly emerged as one of the premier production centers of export porcelain in southeast China during the Song and Yuan dynasties (960–1368 CE). The prosperity of porcelain production at Dehua even attracted the attention of foreign travelers, such as Marco Polo, who mentioned a city near the port of *Zaitun* (Quanzhou) called *Tin-gui* (possibly Dehua), where fine and cheap porcelain wares were manufactured in large quantities (Polo, 1926). The rise of Dehua as a porcelain production center is attributed to the rich deposits of high-quality porcelain clays (Gao, 1941; Xu, 1985) and, more importantly, to the expansion of maritime trade networks and the development of Quanzhou as a maritime trade center. Quanzhou experienced a remarkable development after the establishment of the Maritime Trade Superintendency *(shibosi)* in 1087 CE, which allowed local and foreign merchants to head directly to Quanzhou for trading, and quickly evolved from a minor regional city to China's largest port city (Clark, 1991; Li, 1986; So, 2000). Dehua, though located in the inland hilly region, has access to the port of Quanzhou via a short route and then the Jinjiang River. According to the *Dehua Xianzhi* [The Gazetteer of Dehua County], the route to Quanzhou port was established in 964 CE.

Porcelain production at Dehua during the Song and Yuan dynasties was enormous at multiple scales. A total of 42 Song-Yuan kiln sites have been discovered through decades of surveys at Dehua (Xu, 1996) (Fig. 1). Archaeological excavations of the two kiln sites—Wanpinglun and Qudougong—in the 1970s revealed the massive size of kilns used at Dehua (Fujian Museum, 1990). For instance, the Qudougong "dragon" kiln, a long, multi-chambered tunnel kiln built upon a hillside, is over 57 m in length. No evidence suggests that these large-scale kilns at Dehua were owned or controlled by the state. Porcelain production at Dehua was oriented towards overseas markets, and numerous discoveries of Dehua wares in ancient shipwrecks and terrestrial sites along ancient maritime routes support this hypothesis (e.g., Cremin, 2007; Goddio, 1997; Mathers and Flecker, 1997; Miksic, 2009, 2017; National

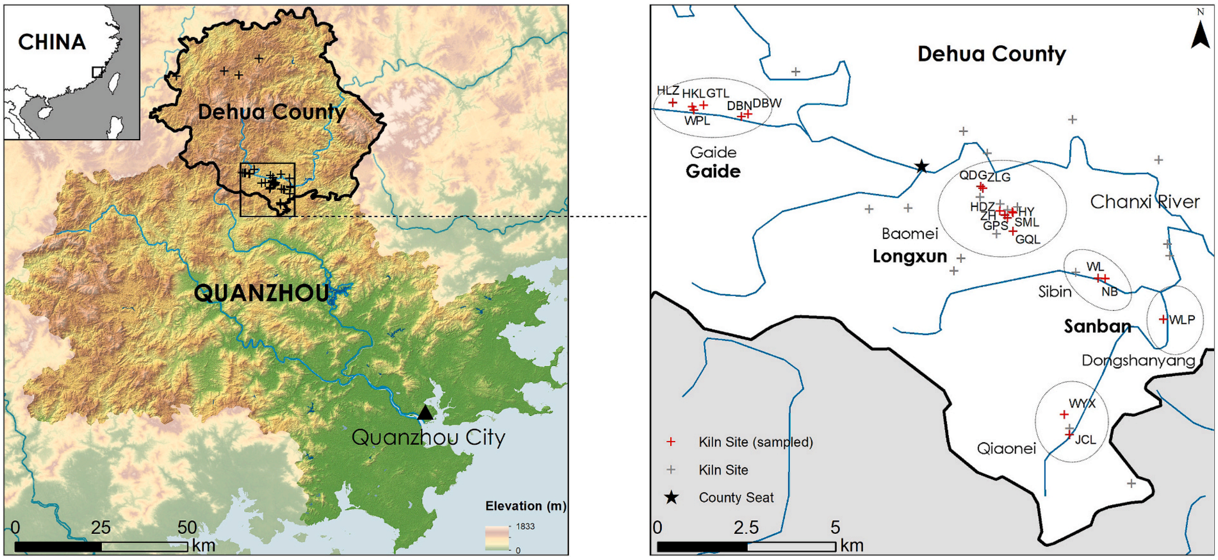

Fig. 1. Location of Dehua County in Quanzhou, Fujian, China, and locations of kiln sites of the Song and Yuan periods at Dehua. Base map of the left: ASTER Global Digital Elevation Model V003, distributed by NASA EOSDIS Land Processes DAAC. https://doi.org/10.5067/ASTER/ASTGTM.003.

Center of Underwater Cultural Heritage et al., 2017, 2018; Niziolek, 2018; Qin et al., 2017; Underwater Archaeology, 2005; Wong, 2006; Xu et al., 2019; Zhao, 2015).

The boom of porcelain production at Dehua suffered a tremendous blow during the early Ming dynasty due to the imposition of a series of *haijin* (sea ban) policies to forbid private trade (Chao, 2005; Li, 1990). Quanzhou's role as an international trade center quickly fell as its trade was restricted to the Ryukyu Islands only via official tribute trade (So, 2000:125–127). Archaeological surveys at Dehua revealed little information about porcelain production of the early Ming dynasty, suggesting that the Dehua porcelain industry, which heavily relied on the port of Quanzhou for overseas trading, witnessed a significant retraction. It was not until the removal of the *haijin* policies under the Longqing era (1567–1572 CE) that porcelain production at Dehua experienced a resurgence. High-quality white porcelains with the appearance of ivory white or milky white, widely known in the West as '*blanc de Chine*' ("white from China"), became the major products of this period and were exported in large quantities to Europe from the 17th and 18th centuries (Ayers and Kerr, 2002; Donnelly, 1969). Porcelain production at Dehua witnessed another considerable expansion during the Qing dynasty (1644–1911 CE), with the evidence of over 100 kilns being discovered throughout Dehua County (Xu, 1996). Currently, Dehua still has one of China's largest porcelain industries and exports its wares throughout the world.

Compositional analyses of Dehua ceramics have been conducted primarily using X-ray fluorescence (XRF) to analyze the major and minor elements in pastes and glazes to understand the chemical characterizations of ceramics from the few excavated kiln sites (Guo and Li, 1985; Huang et al., 2002; Leung et al., 2000a; Li et al., 2011). Strontium isotope compositions were also used to help identify the type of fluxes used in glaze recipes (Ma et al., 2016). Research has yet to be conducted to explore the possible compositional similarities and differences between kilns within the Dehua kiln complex.

3. Materials and methods

3.1. Samples

Samples used in this research come from archaeological excavations and surveys of 19 kiln sites dated to the Song and Yuan periods

Table 1
Number of ceramic samples used in LA-ICP-MS and pXRF analysis by kiln site.

Kiln ID	Kiln Site		Village	Town	Number of samples (LA-ICP-MS)	Number of samples (pXRF)
DBN	Dabannei	大坂内窑	Gaide	Gaide	7	30
DBW	Dabanwai	大坂外窑	Gaide	Gaide	6	31
GTL	Gongtianlun	公田仑窑	Gaide	Gaide	6	33
HKL	Houkenglong	后坑垄窑	Gaide	Gaide	6	22
HLZ	Honglongzai	后垄仔窑	Gaide	Gaide	4	26
WPL	Wanpinglun	碗坪仑窑	Gaide	Gaide	4	44
GPS	Gongposhan	公婆山窑	Baomei	Longxun	6	32
GQL	Gongqiaolong	拱桥垄窑	Baomei	Longxun	6	30
HDZ	Houdianzai	后店仔窑	Baomei	Longxun	6	29
HY	Houyao	后窑	Baomei	Longxun	6	30
QDG	Qudougong	屈斗宫窑	Baomei	Longxun	6	20
SML	Shimuling	虱母岭窑	Baomei	Longxun	6	31
ZH	Zhaihou	寨后窑	Baomei	Longxun	5	24
ZLG	Zulonggong	祖龙宫窑	Baomei	Longxun	7	19
NB	Neiban	内坂窑	Sibin	Sanban	10	37
WL	Weilin	尾林窑	Sibin	Weilin	4	30
WLP	Wuluping	乌鲁坪窑	Dongshanyang	Sanban	5	30
JCL	Jiachunling	佳春岭窑	Qiaonei	Sanban	6	32
WYX	Wanyaoxi	碗窑溪窑	Qiaonei	Sanban	7	35
Total					113	565

(960–1368 CE) at Dehua. The 19 kiln sites are distributed in five villages and three towns (Fig. 1 and Table 1). Due to the destruction of many kiln sites, it is not feasible to collect samples from all discovered kiln sites dated to the Song and Yuan periods. The 19 kilns analyzed in this research were widely spread in different production regions at Dehua, so analyzing ceramic samples from the 19 kilns can still provide a good picture of Song-Yuan porcelain production at Dehua.

The production dates of the 19 kiln sites overlap but are not exactly the same. It has long been suggested that the date of the Wanpinglun kiln in Gaide is earlier than that of the Qudougong kiln in Longxun (Fujian Museum, 1990), though precise dating of the two kiln sites is still under debate due to the very limited dateable materials available (Ho, 2001; Lin and Zhang, 1992; Meng, 2017; Zeng, 1990). Our stylistic analysis of ceramic products and stacking techniques of the 19 kiln sites shows that kilns in Longxun and Sanban might have different ceramic-making traditions compared to kilns in Gaide. Kilns in Gaide primarily used the pronged-ring stacking technique, referring to stacking ceramics on rings with five or six prongs, which would leave the inside bottom of the ceramics with five or six-prong marks (Fig. 2). Kilns in Longxun and Sanban, though still used the pronged-ring stacking technique on a small scale, primarily employed the new invert stacking technique, in which ceramics were placed upside down in ring setters to form a tall stack (Fig. 2). Porcelains fired using this technique are open-shaped, thin-walled vessels with unglazed rims. Despite the use of different stacking techniques, kilns at Dehua all produced some similar types of products, such as covered boxes and molded small bottles. These covered boxes and molded small bottles have been discovered in several ancient shipwrecks dated to the 12th-13th centuries, such as the Nanhai I shipwreck, the Huaguangjiao I shipwreck, and the Java Sea Shipwreck (Mathers and Flecker, 1997; National Center of Underwater Cultural Heritage et al., 2017, 2018; Niziolek et al., 2018; Underwater Archaeology, 2005; Xu et al., 2019). The Nanhai I shipwreck also carried a large number of Dehua plates with five or six-prong marks, which were produced in both Gaide and Longxun-Sanban. Thus, based on the stylistic analyses of ceramics from kiln sites and shipwrecks, we suggest that porcelain production in Gaide approximately spanned from the late 11th to 13th centuries and that kilns in Longxun and Sanban had a slightly later production date, roughly ranging from the 12th to 14th centuries.

A total of 113 samples were analyzed using LA-ICP-MS at the Elemental Analysis Facility (EAF) at the Field Museum in Chicago. A total of 565 samples were analyzed using pXRF at Dehua. Table 1 lists the locations of the 19 kiln sites and the number of samples analyzed using two analytical techniques (LA-ICP-MS and pXRF) for each site. We selected samples representing the typical types of products produced at each kiln site and excluded overfired and underfired sherds that may potentially affect the glaze compositions.

3.2. LA-ICP-MS and pXRF analysis of glazes

Compositional analysis of ceramics often uses paste to identify groups. In the case of Chinese porcelain production, glaze recipes might display greater compositional variability compared to paste recipes

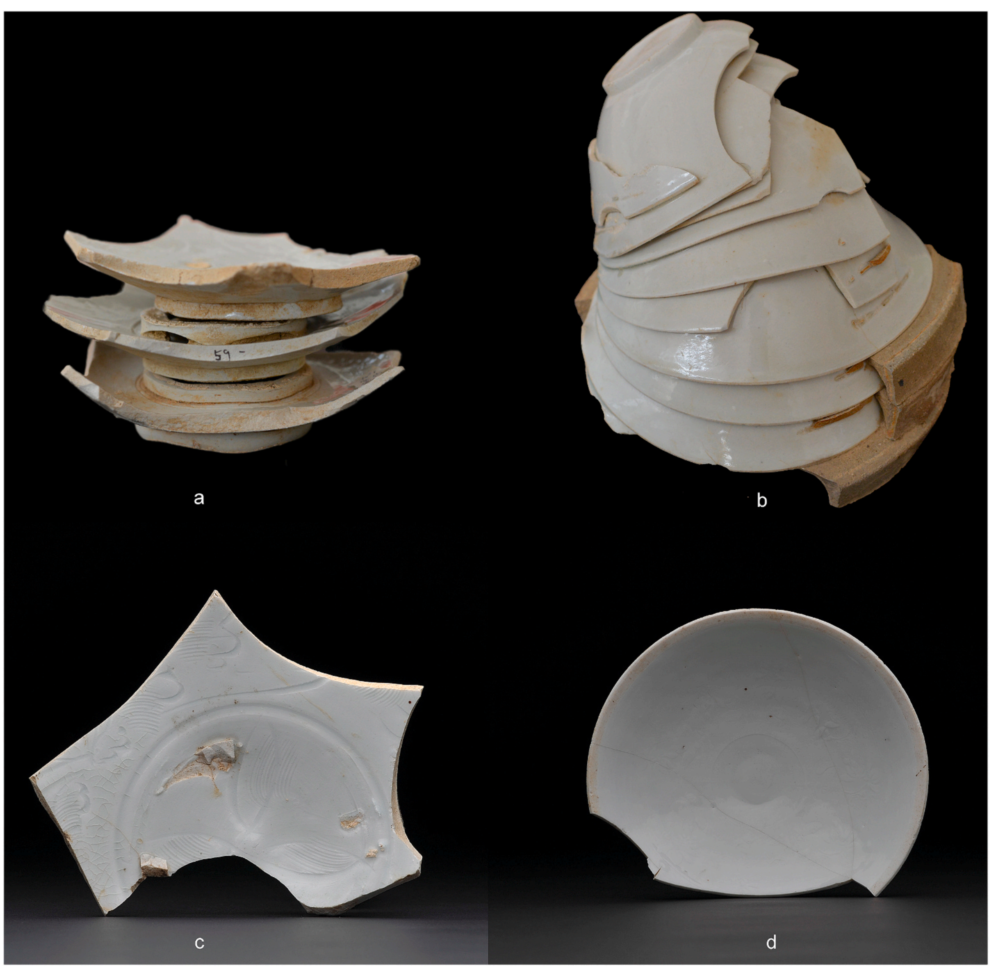

Fig. 2. Two different stacking techniques and examples of Dehua porcelains analyzed in this research. (a) The pronged-ring stacking technique. (b) The invert stacking technique. (c) *Qingbai*/white porcelain plate with five-prong marks (fired using the pronged-ring stacking technique). (d) *Qingbai*/white porcelain bowl with an unglazed rim (fired using the invert stacking technique).

because fluxes, such as burnt limestone and plant ash, were mixed with porcelain clay in the glaze-making process. In contrast, pastes were typically made from porcelain clay without adding additional materials. Glazes were usually made in small batches, often by potters themselves (Kitamura, 1908). Even if different kilns at Dehua used clay from the same source, each kiln might still use its unique combinations of clay and fluxes in making glazes. Therefore, this research prefers analyzing glazes to understand the patterns of compositional similarities and differences between kiln sites at Dehua.

The examination of the inter-kiln compositional similarities and differences requires both a sensitive analytical technique and a relatively large number of samples. LA-ICP-MS is considered as one of the best analytic techniques for examining the compositional patterns within a small region of ceramic production because of its excellent sensitivity, precision, and accuracy as well as low detection limits: it is capable of measuring the concentrations of more than 50 elements and reading elements present in the parts per billion (Dussubieux et al., 2007). LA-ICP-MS has been increasingly used in archaeological studies of materials (e.g., Arnold, et al., 2007; Dussubieux et al., 2016; Neff, 2003; Niziolek, 2013; Riebe and Niziolek, 2015; Sharratt et al., 2009, 2015; Speakman et al., 2007; Speakman and Neff, 2002, 2005) and has also been applied to compositional analysis of Chinese porcelains with success (e.g., Li, et al., 2003, 2005, 2006; Niziolek, 2015, 2017; Oka et al., 2009; Wang et al., 2018, 2020; Zhu et al., 2012, 2015). However, it is often not feasible to analyze a very large number of samples using LA-ICP-MS because it is semi-nondestructive, expensive, and time-consuming, although LA-ICP-MS is considered to be faster and cheaper than INAA or even ICP-MS and other technique that requires dissolution. On the other hand, portable XRF, as a rapid and non-destructive technique, allows for the analysis of a large number of samples *in situ*. Although the elemental concentrations measured with pXRF might not be as accurate as other high-sensitivity techniques, studies have shown that pXRF is capable of identifying compositional groups that closely correlate to those indicated by NAA or ICP-MS (Adlington et al., 2020; Forster et al., 2011; Holmqvist, 2017; Hunt and Speakman, 2015; Johnson, 2014; LeMoine and Halperin, 2021; Mitchell et al., 2012; Speakman et al., 2011; Li et al., 2021). Recent research has also shown that pXRF can effectively distinguish Chinese porcelains from different production regions (Cui et al., 2017; Cui et al., 2016; Fischer and Hsieh, 2017; Xu et al., 2019). Considering the advantages and disadvantages of LA-ICP-MS and pXRF, this research combines the two analytical techniques to examine the inter-kiln compositional similarities and differences.

The glazes of the 113 porcelain samples were analyzed using a Thermo Scientific iCAP Q quadrupole ICP-MS with a New Wave UP213 laser at the Elemental Analysis Facility (EAF) at the Field Museum in Chicago. Analytical procedures and parameters followed the well-established approach (e.g, Duwe and Neff, 2007; Greer et al., 2021; Klesner et al., 2019) and the standard protocol of the EAF (Dussubieux et al., 2007; Niziolek, 2015; Oka et al., 2009). The spectrometer was set to measure 60 elemental isotopes for each sample: ^{7}Li, ^{9}Be, ^{11}B, ^{23}Na, ^{24}Mg, ^{27}Al, ^{29}Si, ^{31}P, ^{35}Cl, ^{39}K, ^{44}Ca, ^{45}Sc, ^{49}Ti, ^{51}V, ^{52}Cr, ^{55}Mn, ^{57}Fe, ^{59}Co, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{82}Se, ^{85}Rb, ^{88}Sr, ^{89}Y, ^{90}Zr, ^{93}Nb, ^{95}Mo, ^{107}Ag, ^{111}Cd, ^{115}In, ^{118}Sn, ^{121}Sb, ^{133}Cs, ^{137}Ba, ^{139}La, ^{140}Ce, ^{141}Pr, ^{146}Nd, ^{147}Sm, ^{153}Eu, ^{157}Gd, ^{159}Tb, ^{163}Dy, ^{165}Ho, ^{166}Er, ^{169}Tm, ^{172}Yb, ^{175}Lu, ^{178}Hf, ^{181}Ta, ^{182}W, ^{197}Au, $^{206, 207, 208}$Pb, ^{209}Bi, ^{232}Th, and ^{238}U. For the glaze analysis, a total of 4 locations on each sample were chosen for ablation. Visible inclusions in the ceramics were avoided as much as possible. A pre-ablation pass line with a laser spot size of 110 μm and a pass speed of 70 μm/s was first used to remove potential surface contamination and to eliminate the transient part of the signal. An ablation pass line with a spot size of 100 μm, at 10 μm/s, and a dwell time of 60 s was then performed on samples to ablate glaze, using a 213 nm wavelength laser operating at 80% energy and a pulse frequency of 10 Hz. A total of five samples were placed into the ablation chamber each time. Glass standard materials and controls—NIST SRM 610, Corning B, and Corning D—were run every five samples to correct for instrument drift over time and to calculate element concentrations. Standards and controls were also ablated four times each in different locations. Results of the LA-ICP-MS analysis can be found in Appendix A. The average of the measurements for Corning B and Corning D as well as the relative standard deviation and accuracy can be found in Appendix C.

Element concentrations for each sample and control were corrected using silicon (^{29}Si) as an internal standard to improve the reproducibility of measurements (Dussubieux et al., 2007). To decrease the effects of potential outlying values associated with instrument drift or surface contamination, aberrant readings for each element were eliminated for samples and standards using the mean, standard deviation, and relative standard deviation of the four readings of each sample with a threshold of 15% of variation. Usually, no more than one outlying value was removed per element. The relative standard deviation (RSD) of the remaining three or four measurements of each element is quite low (generally less than 10% or even 5%), suggesting that there is not much compositional variability within samples. Of the original 58 elements measured by LA-ICP-MS, four elements with readings consistently below the limit of detection were excluded from further statistical analysis: Cl, Se, Cd, and Au.

The glazes of 565 samples were analyzed at Dehua using a Thermo Niton XL3t GOLDD + portable X-ray fluorescence spectrometer with a silver (Ag) anode tube. The main filter operates at voltage of 40 kV and current of 100 μA. Compositional data were collected with acquisition times set to 120s. Test All Geo in the soils and minerals mode was used. Results of the pXRF analysis can be found in Appendix B. NIST SRM 610 was analyzed to examine the accuracy of the pXRF device (Appendix D). After removing elements with readings consistently below the limit of detection (LOD) of the pXRF analyzer, 15 elements were retained for the glaze analyses: Si, Al, K, Ca, Fe, S, Mn, Sc, Rb, Sr, Zr, Nb, Pb, Bi, and Th.

It is worth noting that the purpose of using pXRF in this research is not to measure the absolute concentrations of elements in the glaze, as the accuracy of pXRF itself is not as good as LA-ICP-MS, but to explore whether pXRF can reveal similar compositional patterns suggested by LA-ICP-MS. Since the measurements of some elements, especially those with high energy lines, in the glaze by pXRF might also reflect the contribution of both the glaze and body (Bezur and Casadio, 2012), it is important to explore how the glaze thickness impacts analysis results. The glaze thickness of the ceramic samples used in this research ranges from 150 to 350 μm (Fig. 3). For pXRF analysis, the infinite thickness for Mg, Al, Si, P, K, Ca, Ti, Mn, and Fe in a clear glaze layer are all below 140 μm (Bezur and Casadio, 2012:262), so the measurements of these elements are mostly likely restricted to the glaze layer. For clear glazes that are 150–350 μm thick, Rb, Sr, Y, Zr, Pb, and Th are very likely contributed by both the glaze and the porcelain body (Bezur and Casadio, 2012:262). To examine the degree of compositional variability within samples, which might be contributed by the uneven glaze thickness and insufficiently homogenized melts, we also analyzed the same sample in ten different spots. As shown in Appendix E, the relative standard deviations of the ten measurements for all measured elements are quite low. Although the readings for some elements (such as Rb, Sr,

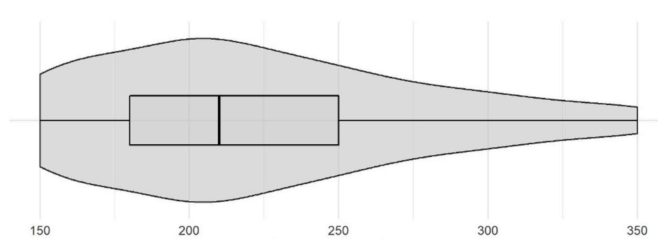

Fig. 3. Violin plot of the glaze thickness of the porcelain samples analyzed in this research.

Zr, and Th) very likely reflect the contribution of both the glaze and the body, the relatively low %RSD suggests that there is not much variation within samples.

3.3. Multivariate statistical analysis and social network analysis

Unsupervised learning techniques—principal component analysis (PCA) and hierarchical cluster analysis (HCA)—were first used to discover compositional patterning in the data and identify compositional groups. Log base-10 values of the elements were used in these analyses to minimize overall differences between major, minor, and trace elements (Baxter, 2006; Bishop and Neff, 1989; Glascock, 1992).

To better examine the relationship between kiln sites at Dehua in terms of glaze compositions, this paper also combines multivariate statistical results with a social network analysis (SNA) approach. SNA has gained increasing popularity in archaeological research for examining patterns of interactions and relationships, such as the exchange networks, the movement of ideas and people, and social boundaries (e.g., Brughmans, 2010; Donnellan, 2020; Golitko and Feinman, 2015; Golitko et al., 2012; Hart and Engelbrecht, 2012; Hart, 2012; Knappett, 2013; Knappett et al., 2008; Mills et al., 2013; Mills et al., 2015; Mizoguchi, 2009; Östborn and Gerding, 2014; Ownby et al., 2014; Peeples, 2019). Archaeological research usually treats material culture similarity as a proxy for measuring the strength of relationships between sites. Whether similarities are produced through direct contact or indirect influences, greater similarities between two nodes can be defined as stronger ties. The underlying supposition is that greater homogeneity between nodes is expected when they have relatively frequent contact and less homogeneity between nodes with less frequent contact (Friedkin, 1984). In the case of porcelain production, kiln sites are considered as actors ("nodes"), and the connections ("edges") between them are measured based on similarities of chemical compositions of porcelains between kilns.

To facility social network analysis, the Brainerd-Robinson (BR) coefficient of similarity was calculated based on the identified compositional groups from HCA. The Brainerd-Robinson coefficient, which creates weighted network data to allow for more nuanced interpretations of network characteristics, serves as a proxy for exploring the inter-kiln compositional similarities (Brainerd, 1951; Cowgill, 1990; Peeples and Roberts, 2013; Robinson, 1951). The result of the BR coefficient of similarity, which is a similarity matrix of the 19 kiln sites ranging between 0 (indicating complete dissimilarity) and 200 (indicating complete similarity), was used to measure the edge weight among kiln sites. We also scaled the BR similarity coefficient to range between 0 and 1 to facilitate network metrics calculation (Peeples and Roberts, 2013). Node-level centrality (degree and eigenvector) measures were calculated to examine the importance of kilns in the network. Degree centrality, a simple measure of the number of ties a node has, can reflect the level of participation or connection of kilns in the network. Eigenvector centrality, which counts the number of nodes adjacent to a given node but weights each adjacent node by its centrality, can provide a good characterization of a kiln's overall position within the network. These centrality measures were calculated based on weighted measures, which treat higher link weights as less costly to traverse (Opsahl et al., 2010; Peeples and Roberts, 2013), to examine which kiln was more influential and was in a better position to control technological information flows through the network. Node-level centrality measures were also used to calculate network-level centralization indices to examine the extent to which entire nodes are concentrated (Freeman, 1978; Peeples and Roberts, 2013).

All of the analyses presented in this research were conducted in R, version 3.6.3 (R Core Team, 2020). HCA and PCA were conducted using the dendextend package (Galili, 2015) and the stats package (R Core Team, 2020), respectively. The Brainerd-Robinson coefficient of similarity was calculated using the script written by Peeples (2011). Network measures were calculated using the igraph package (Csardi and Nepusz, 2006), the sna package (Butts, 2008), the tnet package (Opsahl et al., 2010), and the script written by Peeples (2017).

4. Results and discussion

4.1. Results from LA-ICP-MS analysis

Principal component analysis (PCA) was first conducted on the LA-ICP-MS data to examine the compositional patterning. The biplot of the first two principal components shows that porcelain samples from the Gaide town can be unambiguously differentiated from those from Longxun and Sanban (Fig. 4). Within the Gaide group, the elemental signatures of the six kilns (DBN, DBW, GTL, HKL, HLZ, and WPL) highly overlap and cannot be separated. Within the Longxun-Sanban group, there are significant overlaps between kilns within the same village, whereas different villages do not entirely overlap. The Baomei village only partially overlaps with the other three villages—Dongshanyang, Qiaonei, and Sibin. The three kilns (WLP, JCL, and WYX) in the Dongshanyang and Qiaonei villages highly overlap, but they only partially overlap with the Baomei and Sibin villages. The two kilns (NB and WL) in the Sibin village partially overlap with the other three villages—Baomei, Dongshanyang, and Qiaonei.

Hierarchical cluster analysis was performed using Euclidean distances as the dissimilarity measure and Ward's minimum variance as the clustering method to examine the compositional patterning of the LA-ICP-MS data. As shown in the dendrogram plot arising from HCA (Fig. 5), there are clearly two main clusters: samples from the Gaide subregion join into one main cluster and samples in the Longxun-Sanban subregion join into another main cluster. Although there are overlaps in the dendrogram between different villages in the Longxun-Sanban cluster, samples from Baomei largely group together, while samples from Dongshanyang and Qiaonei largely join into one small cluster. Samples from Sibin are scattered into different groups. This result is consistent with the above grouping patterns revealed from PCA.

There are clear differences in the concentrations of many trace elements between the two main production groups—Gaide and Longxun-Sanban. Compared to those from the Longxun-Sanban group, glazes of porcelains from the Gaide group are characterized by substantially higher concentrations of heavy mineral-related trace elements, such as Zr, Nb, Hf, Ta, and Th (Fig. 6). The inclusion of trace elements in ceramics is mostly accidental, and their concentrations are less susceptible to modification by human activity (Glascock, 1992; Wilson and Pollard, 2001). In addition, glazes of porcelains from the Gaide region are characterized by significantly higher amounts of Fe (Fig. 6). The concentration of iron in ceramics also reflects the characteristics of the clay used in the production because the process of washing clay does not necessarily reduce the iron content in ceramics (Guo, 1987:5). These elements—Fe, Zr, Nb, Hf, Ta, and Th—are critical for the identification of the two main groups. PCA conducted only using the six elements shows that the two main groups—Gaide and Longxun-Sanban—can be clearly discriminated (Fig. 7). The four villages in Longxun-Sanban cannot be separated using the six elements.

Principal component analysis was also conducted on the lanthanide rare earth elements (REEs) to examine whether the REEs contribute to the discrimination of the groups. The PCA biplot shows that the REEs to some extent contribute to the differentiation of the two main compositional groups (Gaide and Longxun-Sanban) as well as to the partial separation of different villages in Longxun-Sanban (Fig. 8). In addition, there are some differences in the content of flux-related elements between different villages. Glazes of porcelains in Baomei and Sibin have relatively higher concentrations of Ca and K, as well as other flux-related elements—such as Sr and Ba (Fig. 9). However, these differences are not absolute because there are significant overlaps in the amount of flux between different villages.

The identification of two compositional groups—Gaide and Longxun-Sanban—is in line with our stylistic analysis. Differences in stacking

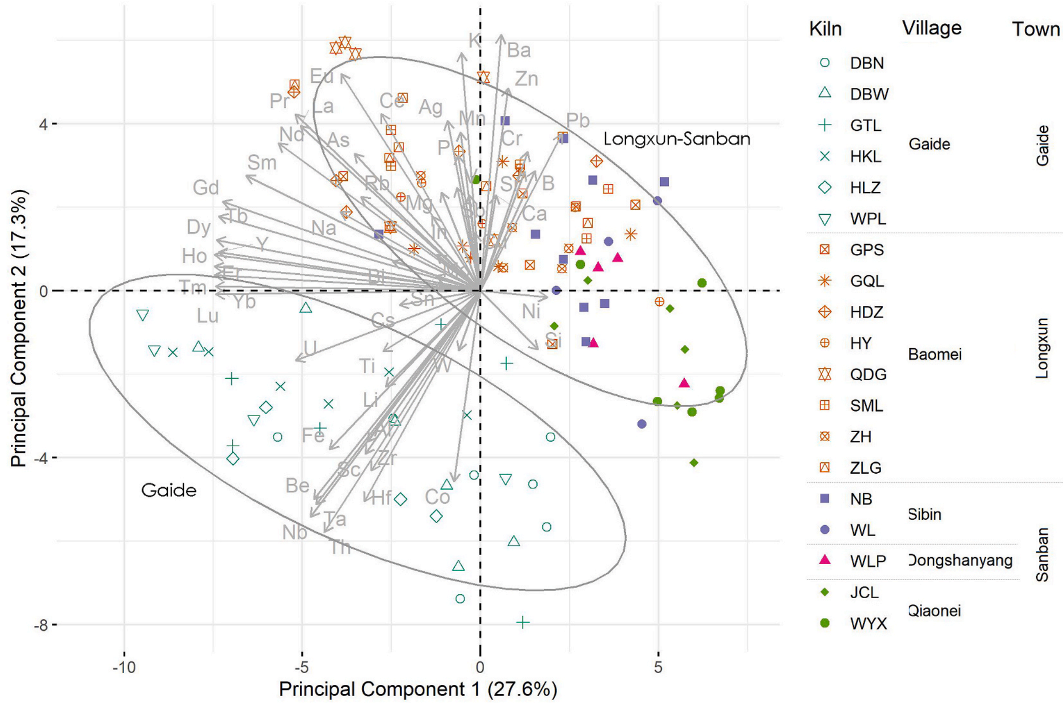

Fig. 4. Biplot of principal components 1 and 2 based on glaze compositions measured by LA-ICP-MS, with samples shaped by kilns and colored by villages. The ellipses represent 90% confidence intervals.

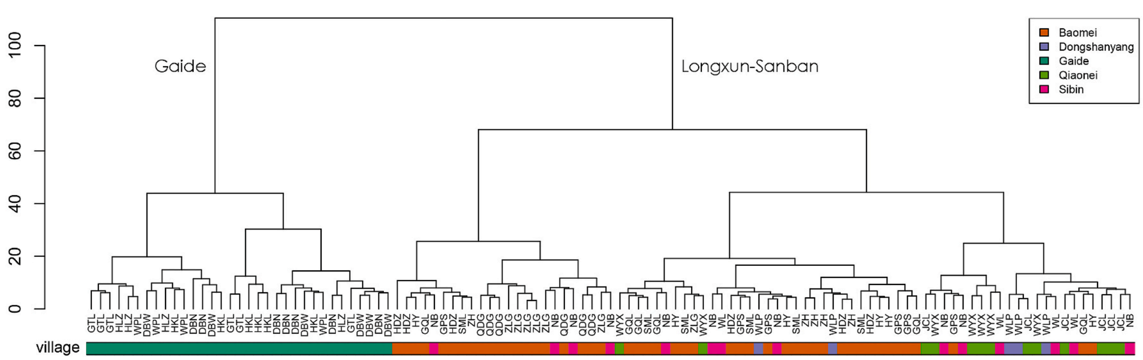

Fig. 5. Dendrogram arising from the hierarchical cluster analysis (Ward's method) of glaze compositions measured by LA-ICP-MS.

techniques, porcelain types, and approaches to fluxing all suggest that the two subregions might have different porcelain-making traditions and technologies. The clear differences in trace elements between Gaide and Longxun-Sanban also suggest that the two subregions have different underlying geologies. Potters in each production subregion probably used local clay for porcelain production without exchange of clay.

4.2. Results from pXRF analysis

Principal component analysis of the pXRF data reveals a pattern that is highly similar to LA-ICP-MS. As shown in the biplot of the first two principal components (Fig. 10), there are two distinct compositional groups—Gaide and Longxun-Sanban. Elemental signatures of kilns in the same village highly overlap, but different villages in Longxun-Sanban do not entirely overlap. Samples from the Baomei and Sibin villages largely overlap, but they only partially overlap with the Dongshanyang and Qiaonei villages.

Hierarchical cluster analysis of the pXRF data also shows that there are two main compositional clusters—Gaide and Longxun-Sanban (Fig. 11). In the Longxun-Sanban cluster, a large percentage of samples from Dongshanyang and Qiaonei and a small number of samples from Baomei and Sibin join into one subcluster, while most of the samples from Baomei-Sibin and some samples from Dongshanyang-Qiaonei join into another subcluster. This pattern is consistent with that revealed by LA-ICP-MS.

pXRF results show that the two groups (Gaide and Longxun-Sanban) can be unambiguously distinguished through a bivariate plot of Zr–Th or Nb–Th, although the measurements of these elements in the glaze by pXRF are not as accurate as LA-ICP-MS (Fig. 12). The compositional pattern of the flux-related elements (Ca and K) measured by pXRF is also highly similar to that measured by LA-ICP-MS (Fig. 13). Porcelains from the Baomei and Sibin villages have relatively higher concentrations of Ca and K, but there are also significant overlaps in the amount of flux between different villages. Results from pXRF also suggest that there is great variation in the amount of flux in glazes within kilns (Fig. 13). Further studies on a larger number of samples are needed to better examine the possible factors that contribute to the variation.

In summary, the compositional patterns revealed by pXRF are highly consistent with those revealed by LA-ICP-MS. Although the absolute elemental concentrations measured by pXRF are not as accurate as LA-ICP-MS, the highly comparable results suggest that pXRF can identify geochemical groups that closely correlate to those indicated by the high-

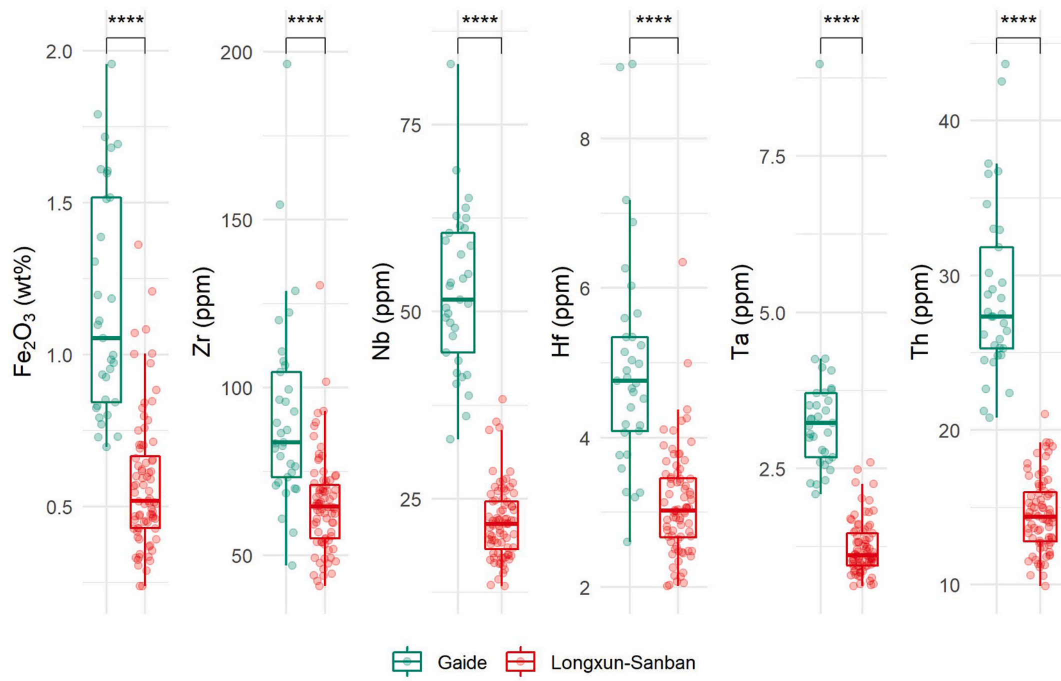

Fig. 6. Boxplots of the concentrations of Fe_2O_3, Zr, Nb, Hf, Ta, and Th in glazes measured by LA-ICP-MS for each subregion. Symbols indicate the statistical significance of the Welch's *t*-test: ****: $p \leq 0.0001$.

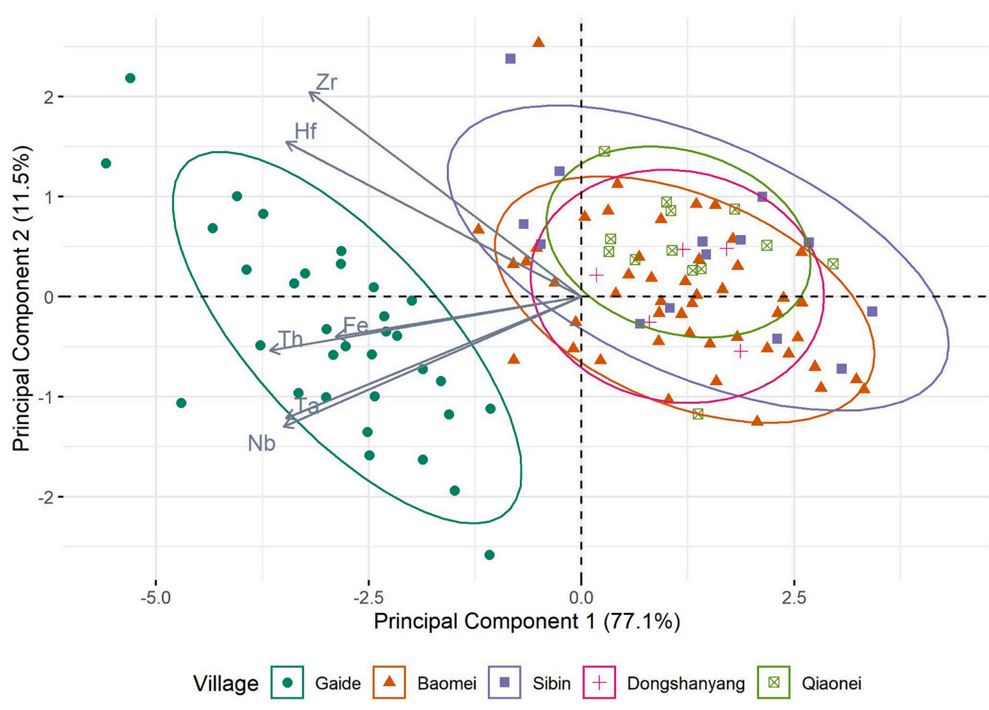

Fig. 7. Biplot of principal components 1 and 2 from PCA conducted using the six elements—Fe, Zr, Nb, Hf, Ta, and Th. The ellipses represent 90% confidence intervals.

sensitivity technique—LA-ICP-MS.

4.3. Results from social network analysis

The above multivariate statistical analyses reveal some differences in the compositional similarities between different kiln sites within the same production subregion; however, it is cumbersome to use the PCA biplot or HCA dendrogram to examine the patterns of inter-kiln compositional similarities and differences similarities, such as whether kilns geographically close have higher compositional similarities and whether there are small production communities within each subregion. To better examine the strength of connections between kiln sites at Dehua in terms of glaze compositions, we calculated the Brainerd-Robinson (BR) coefficient of similarity based on the compositional subgroups identified through HCA, and then analyzed it using a social network analysis (SNA) approach. Although the results from LA-ICP-MS

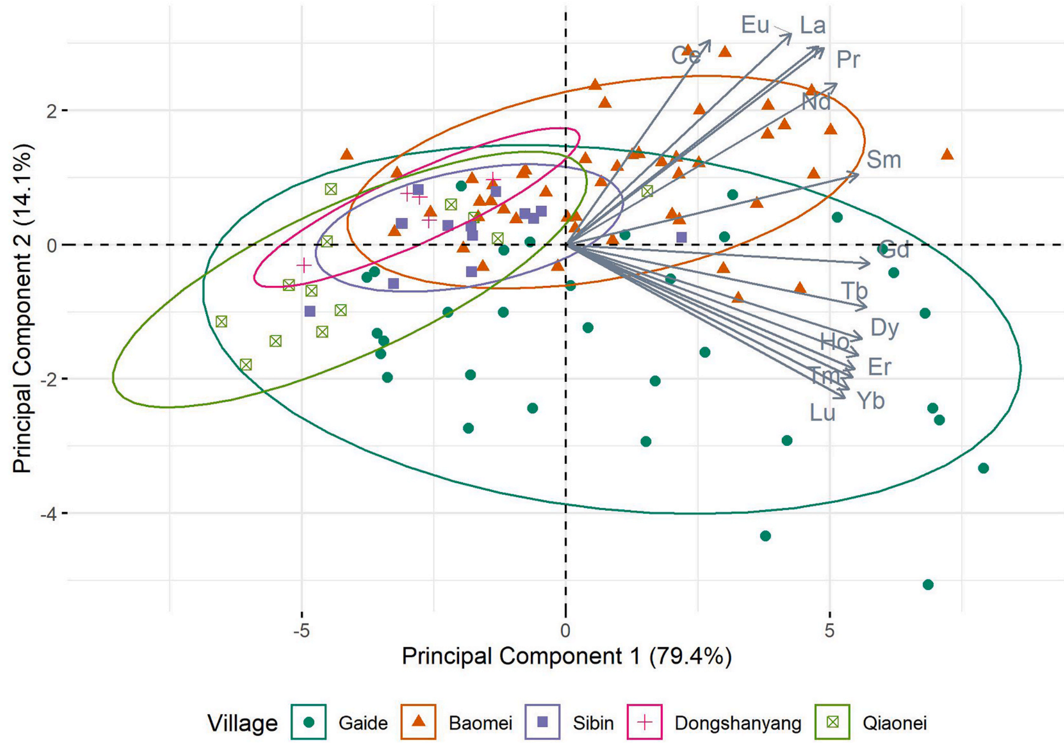

Fig. 8. Biplot of principal components 1 and 2 from PCA conducted using the REEs. The ellipses represent 90% confidence intervals.

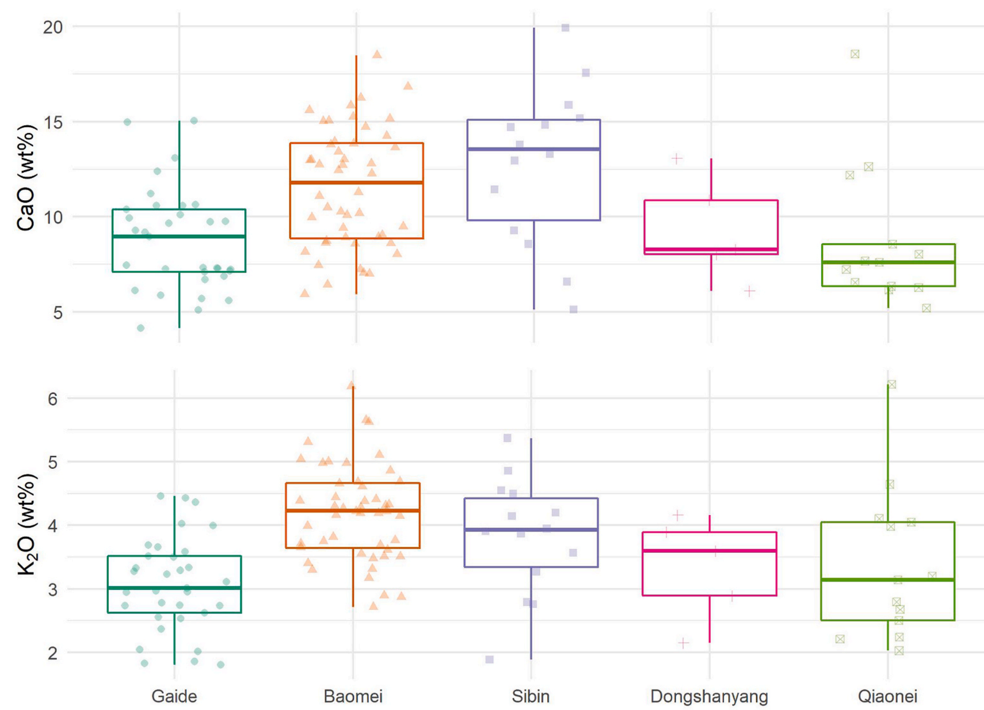

Fig. 9. Boxplots of the concentrations of Cao and K_2O in glazes measured by LA-ICP-MS for each village.

and pXRF analyses are highly consistent, the relatively small number of samples analyzed using LA-ICP-MS might introduce a bias for analyzing the inter-kiln compositional similarities and differences. It is more reasonable to conduct SNA based on the pXRF results. As there are generally no rules of deciding how many clusters to read out from HCA results (Baxter, 2006; Drennan, 2010), the decision devolves primarily on what is meaningful. In this study, cutting the dendrogram into 6, 12, or 18 compositional groups all looks reasonable (Fig. 11). The smaller the number of groups identified, the more likely it is that samples from different kilns would join together. In other words, cutting the dendrogram into a larger number of groups would provide a more refined picture of the inter-kiln compositional similarities and differences. In this research, we used 18 compositional subgroups to calculate the Brainerd-Robinson coefficient of similarity. The results of compositional

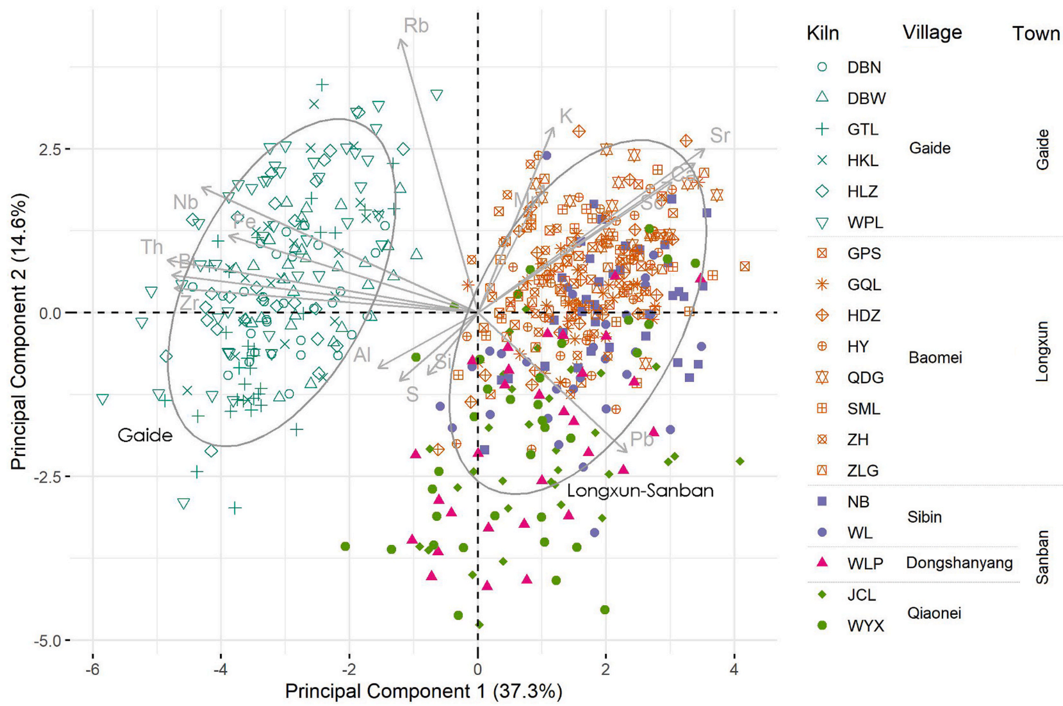

Fig. 10. Biplot of principal components 1 and 2 based on glaze compositions measured by pXRF, with samples shaped by kilns and colored by villages. The ellipses represent 90% confidence intervals.

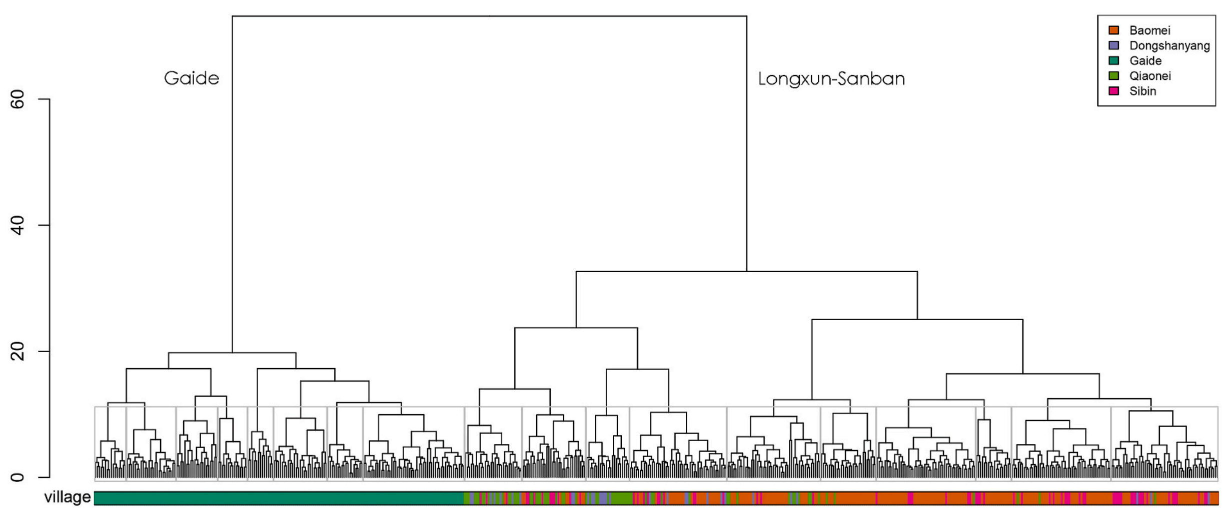

Fig. 11. Dendrogram arising from the hierarchical cluster analysis (Ward's method) of glaze compositions measured by pXRF. The gray rectangles represent the 18 compositional groups used for network analysis.

grouping and BR coefficient can be found in Appendix F and G.

As shown in the network graphs (Fig. 14), the 19 kilns at Dehua formed two distinct clusters/production communities—the six kilns in Gaide formed one production community, whereas the remaining 13 kilns in Longxun and Sanban formed another production community. Although this patterning is expected as PCA and HCA both clearly show that there are two distinct groups, SNA reveals a more clear picture of the production networks within each production subregion. Kilns in the same village are more closely connected than those outside the village. The six kilns in Gaide are highly connected in terms of glaze compositions, although the tie between HKL to other kilns are relatively weak. The ties between the eight kilns (GPS, GQL, HDZ, HY, QDG, SML, ZH, and ZLG) in Baomei are quite strong, so are the three kiln sites (WLP, JCL, and WYX) in Dongshanyang and Qiaonei. The connections between the Baomei and Dongshanyang-Qiaonei groups are relatively weak. The two kiln sites (NB and WL) in the Sibin village, located between Baomei and Dongshanyang-Qiaonei, are moderately connected to both the Baomei and Dongshanyang-Qiaonei groups.

Researchers have argued that kilns at Dehua might be jointly owned by multiple families or workshops bopt_ecause different surnames inscribed or written on porcelains oopt_ kiln furniture were found at the same kiln site (Ho, 1999; So, 2000). Wang (1936) recorded that kilns at Dehua of the early twentieth century were usually joint ventures of several workshops or families: those with larger capital owned two or three chambers of a kiln, while those with small capital had only one chamber, or even shared a chamber with other families. The intra-kiln compositional variability revealed in this research suggests that porcelains from each kiln site might be produced using multiple recipes,

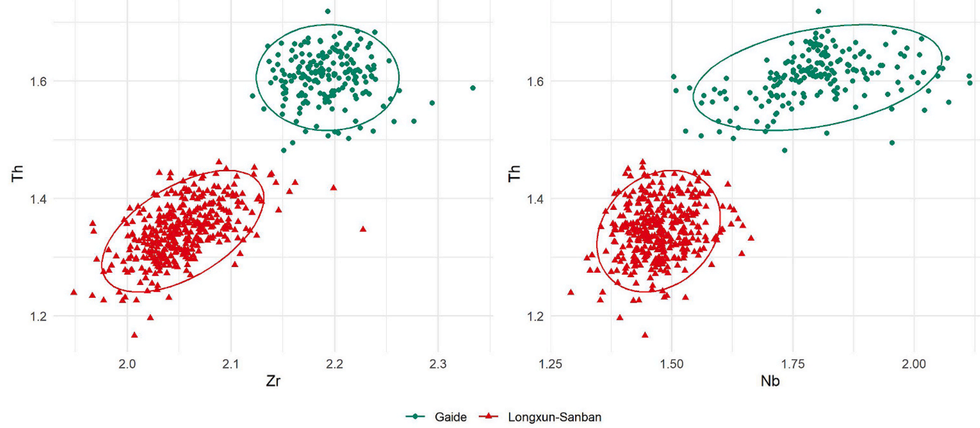

Fig. 12. Bivariate plots of log-base 10 values for Zr–Th and Nb–Th measured by pXRF. The ellipses represent 90% confidence intervals.

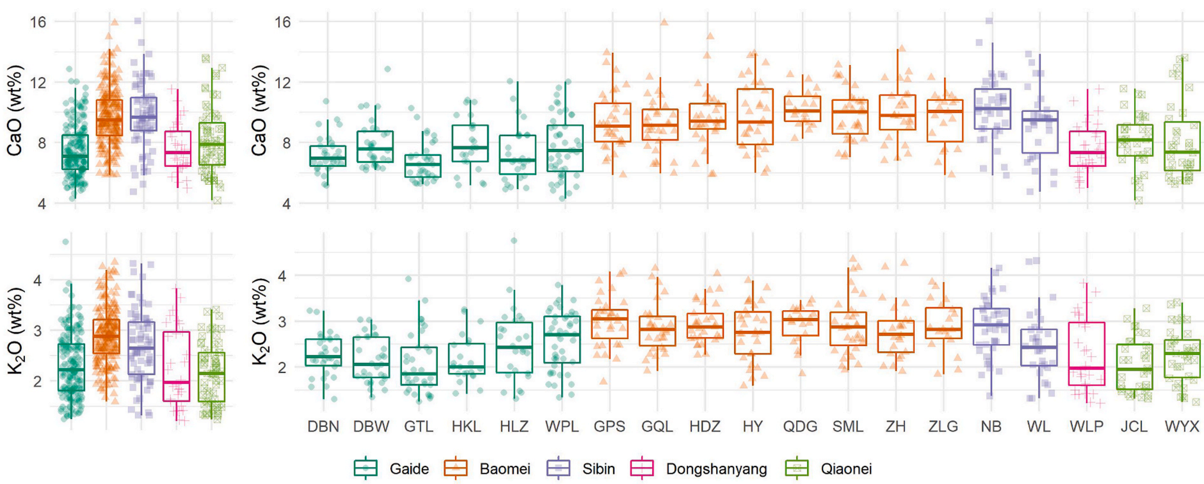

Fig. 13. Boxplots of the concentrations of Cao and K_2O measured by pXRF for each village and each kiln.

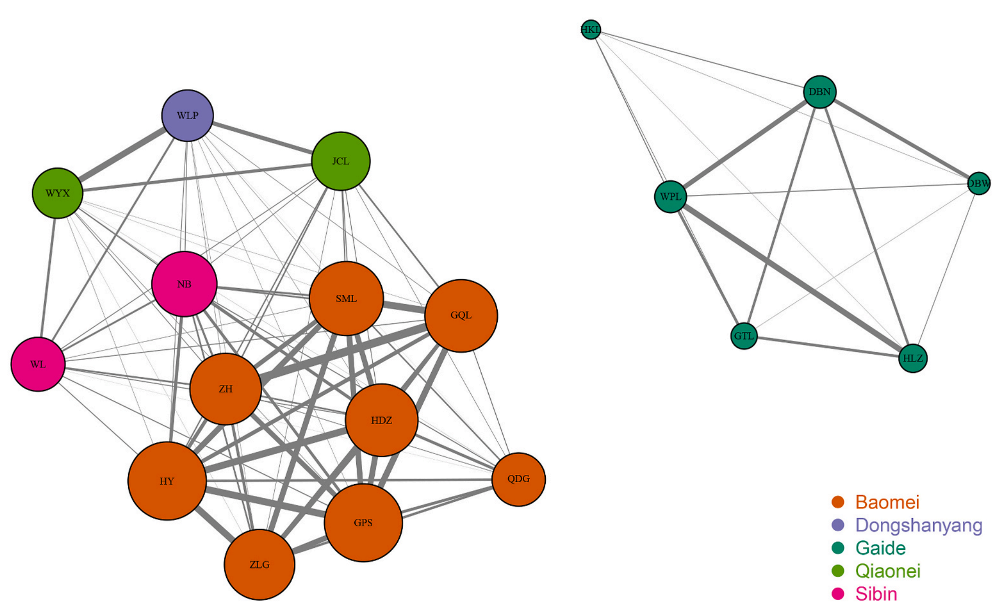

Fig. 14. Weighted force-directed network graph of the porcelain production at Dehua. Edge thickness represents the link strength between kiln sites in terms of glaze compositions. The relative sizes of nodes represent the weighted degree centrality scores for each kiln site.

which further indicates that multiple families or workshops might jointly own one kiln at Dehua during the Song and Yuan periods. It was also noted that during the late Qing dynasty (the late 19th to early 20th centuries), potters at Dehua obtained clay that was procured and prepared by specialists and then made glazes by themselves using recipes that were handed down from their ancestors or learned from their peers (Kitamura, 1908). Although no written record is available about Dehua porcelain production of the Song and Yuan periods, the highly connected networks suggest that the way of clay procurement and knowledge transmission in the Song and Yuan dynasties might parallel that in the late Qing dynasty. Raw materials and glaze recipes might be widely shared within each production subregion, but the interaction between potters in the same village was more frequent.

5. Conclusions

Using large-scale porcelain production of the Song and Yuan periods at Dehua as a case study, we illustrated that compositional analysis at the micro scale provides a fine-grained perspective on the production organization and patterns of interaction between different production loci below the level of the broader production region. The combination of multivariate statistical analysis with social network analysis also shows its effectiveness in revealing the inter-kiln compositional similarities and differences. Two main production groups/subregions at Dehua—Gaide and Longxun-Sanban—were identified. Geological differences and different ceramic-making traditions might both contribute to the distinct separation of the two compositional groups. The existence of intra-kiln compositional variability supports the argument that kilns at Dehua of the Song and Yuan periods might be jointly owned by multiple families or workshops. Potters associated with the same kiln might use different recipes or different batches of raw materials, resulting in compositional variability among porcelains fired at the same kiln. Within the same production subregion, there are strong inter-kiln compositional similarities, but kiln sites in the same village are more closely connected than those outside the village. Potters working at different kilns in the same subregion (especially in the same village) might use the same batch of raw materials or similar glaze recipes, causing compositional similarities between porcelains fired at different kiln sites.

The research demonstrates the value of conducting chemical composition analysis at the micro level (below the level of the broader production region). Future studies of craft production should consider investigating the possible compositional similarities and differences between individual production units beneath the level of broader production regions. Conducting compositional study at the micro level is particularly crucial for examining the large-scale porcelain production in southeast China, where hundreds or even thousands of huge kilns clustered into numerous production regions. After careful examination of all production regions at the micro level, a more comprehensive study at the supra-regional scale, such as the whole Quanzhou region or even the whole Fujian region, will further provide a much deeper understanding of the large-scale, complex porcelain production systems in southeast China.

Through comparing the results of two analytical techniques—LA-ICP-MS and pXRF, this research also demonstrated that pXRF is capable of finding compositional patterning within a small region of ceramic production, although the measurements of elements by pXRF are not as accurate as LA-ICP-MS. Considering that pXRF can rapidly and non-destructively analyze a large number of samples at a relatively low cost, the validation of the methodology of using pXRF for micro-level analysis will enhance our ability to examine the inter-kiln compositional similarities and differences at a much large scale. Comparison with other high-sensitivity techniques, such as LA-ICP-MS and NAA, is still needed before using pXRF to analyze ceramic products from other production regions.

The identification of two production groups/subregions at Dehua—Gaide and Longxun-Sanban—also provides criteria to help more precisely source Dehua wares from shipwrecks or land sites outside China. The promising results from pXRF also indicate that future research can use pXRF to rapidly identify the subregion of Song-Yuan Dehua wares found overseas. More refined sourcing of Dehua ceramics would reveal a better picture of the organization of the export-oriented production at Dehua and the complex maritime trade networks at the micro level.

Credit author statement

Wenpeng Xu: Conceptualization, Methodology, Funding acquisition, Software, Formal analysis, Investigation, Project administration, Resources, Data curation, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. Zelin Yang: Investigation, Resources, Data curation, Writing – review & editing. Lifang Chen: Investigation, Resources, Data curation. Jianfeng Cui: Methodology, Investigation, Writing – review & editing. Laure Dussubieux: Investigation, Methodology, Funding acquisition, Writing – review & editing. Wenjing Wang: Conceptualization, Methodology, Data curation, Investigation, Writing – review & editing.

Declaration of competing interest

We have no conflict of interest regarding the research or entities involved in this research.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jas.2021.105481.

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