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Sourcing *qingbai* porcelains from the *Java Sea Shipwreck:* Compositional analysis using portable XRF

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ABSTRACT

This paper evaluates the use of portable x-ray fluorescence (pXRF) on glazes and pastes for sourcing Chinese porcelains from the 12th-13th century *Java Sea Shipwreck (JSW)* collection at the Field Museum. Three types of *qingbai* (bluish-white) wares from the *JSW* collection were chosen for pXRF analysis. Samples from four kiln complexes in China—Jingdezhen, Dehua, Huajiashan, and Minqing, hypothesized to be potential sources of the shipwreck's *qingbai* ceramics based on visual inspection—were also analyzed to establish reference groups. Results from kiln samples show that different kiln complexes can be clearly differentiated by pXRF analysis of glazes. Based on pXRF analysis of ceramic samples from the *JSW*, there appear to be four compositional groups, and each group closely matches one of the four kiln reference groups. These findings support the use of pXRF on glazes, especially when pastes are difficult to access, as a method for identifying the potential sources for overseas cargos found distant from production contexts for Chinese porcelains.

1. Introduction

1.1. Overview of project

As a non-invasive and non-destructive analytical technique, portable x-ray fluorescence (pXRF) is suited to research in a museum setting when the conservation of materials is a major concern in research design. Additionally, the ability to rapidly characterize artifacts at relatively low cost makes pXRF particularly attractive for the provenance study of ancient Chinese porcelains because the extremely large number of kilns in China requires a fairly large number of samples to build reference groups. In this paper, we evaluate the potential of using pXRF for differentiating Chinese *qingbai* porcelains from the *Java Sea Shipwreck (JSW)* that are visually similar to one another, as well as for identifying the potential sources of these porcelains. To build reference groups, we first analyzed porcelain samples from four different kiln complexes in China, which are hypothesized to be potential sources of the *JSW qingbai* porcelains based on stylistic similarities. Then, we compared the compositions of porcelain samples from the *Java Sea Shipwreck* to samples from kiln sites to identify their potential sources. Because glazes tend to be more easily accessed than pastes for pXRF analysis, this paper also compares the results of pXRF analysis on glazes and pastes to assess the effectiveness of using pXRF on glazes for differentiating and sourcing Chinese porcelains.

1.2. Historical background

Ancient maritime trade in the South China Sea and Indian Ocean areas prospered during the late first millennium CE to mid-second millennium CE, connecting a large number of regions ranging from China in the east, Southeast and South Asia in the center, and the Middle East and the east coast of Africa in the west (Abu-Lughod, 1989; Chaudhuri, 1985; Clark, 1991; Hall, 2011; Park, 2012; Tampoe, 1989; Wade, 2009; G. Wang, 1958). Through these extensive trade networks, raw materials, agricultural products, and manufactured goods were widely circulated in the pre-modern world system. Among them, high-fired Chinese ceramics (many classified as porcelains) were some of the most prominent trading products, which have been widely found in ancient polities from Japan all the way to East Africa (Addis, 1968–69; Brown, 1989; Cremin, 2007; Ding and Qin, 2016; Guy, 1986; Harrisson, 1970; Lam, 1985; Mikami, 1988, 1990; Miksic, 2009, 2017; Qin, 2013; Yuba, 2011–12; Zhao, 2012, 2015). There is also abundant evidence from shipwreck sites dated to the 9th to 14th centuries in Asian waters. Chinese high-fired ceramics are usually the most common cargo at these shipwreck sites, due to both the sheer number of pieces traded and their ability to survive in archaeological contexts (Brown, 2009; Flecker, 2002; Goddio, 1997; National Center of Underwater Cultural Heritage et al., 2017, 2018; Liebner, 2014; Mathers and Flecker, 1997; Ridho and McKinnon, 1998; Underwater Archaeology, 2005).

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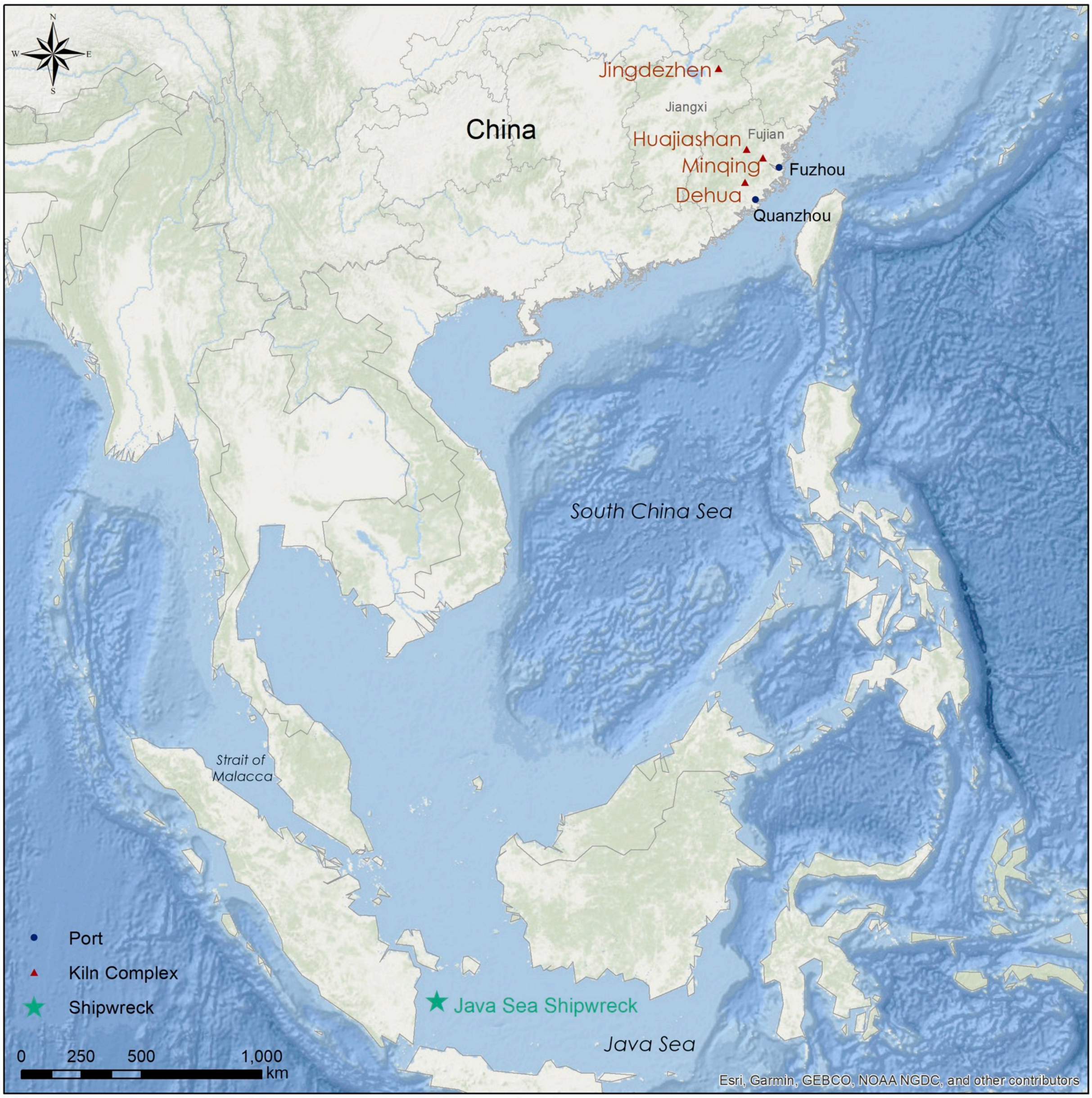

Fig. 1. Kiln complexes in China where samples were collected and the location of the *Java Sea Shipwreck*.

The *Java Sea Shipwreck* collection housed at the Field Museum in Chicago comprises one of these important shipwreck discoveries in Southeast Asia. The shipwreck was first found by fishermen in Indonesian waters between Sumatra and Java in the late 1980s (Fig. 1), and it was later systematically salvaged and recovered by Pacific Sea Resources (Flecker, 2005–2006; Mathers and Flecker, 1997). The date of the wreck was originally estimated to be the mid- to late 13th century (Brown, 1997; Flecker, 1997), but based on recent research, Niziolek et al. (2018) suggest an earlier mid- to late 12th century date be considered (but see Flecker, 2018). The ship was probably sailing from Quanzhou in southeastern China to Tuban on the island of Java, carrying an estimated 100,000 ceramic vessels and 200 tons of iron as well as some other items in smaller quantities, such as ivory, resin, and tin ingots (Flecker, 2003; 2005–2006). The majority of the ceramics found at the *Java Sea Shipwreck* site are Southern Song dynasty high-fired wares from south China, including *qingbai* (bluish-white), celadon, brown-glazed, black-glazed, and painted wares (Brown, 1997). Among them, *qingbai* wares are one of the largest categories of ceramics from the shipwreck.

The term *qingbai* typically refers to a style of high-fired ceramics with a thin body, fine, white paste, and an evenly applied transparent bluish glaze that were produced at Jingdezhen in Jiangxi province under the Song and Yuan dynasties (960–1368 CE) (Jiangxi Provincial Institute of Cultural Relics and Archaeology and Jingdezhen Museum of Civilian Kiln, 2007; Pierson, 2002). Meanwhile, *qingbai* is also used to broadly refer to imitations of Jingdezhen *qingbai* wares that were produced at kilns all over south China. A notable production region of the imitation of *qingbai* wares during the Song and Yuan period is Fujian province in southeastern China. In Fujian, the development of ceramic production corresponded to the rapid growth of maritime trading networks. Archaeological surveys and excavations have revealed dozens of kiln complexes producing *qingbai*-style porcelains dated to the Song and Yuan periods (J.-a. Li, 2008, 2010; Meng, 2017; Zeng, 2001). In general, *qingbai* ceramics from Fujian are considered of lower quality compared to their Jingdezhen counterparts and are characterized by fine to medium textured, white or grayish-white paste, a more opaque bluish-white or grayish-white glaze that is often unevenly applied, and casual and scratchy decoration. While there is little doubt that *qingbai*

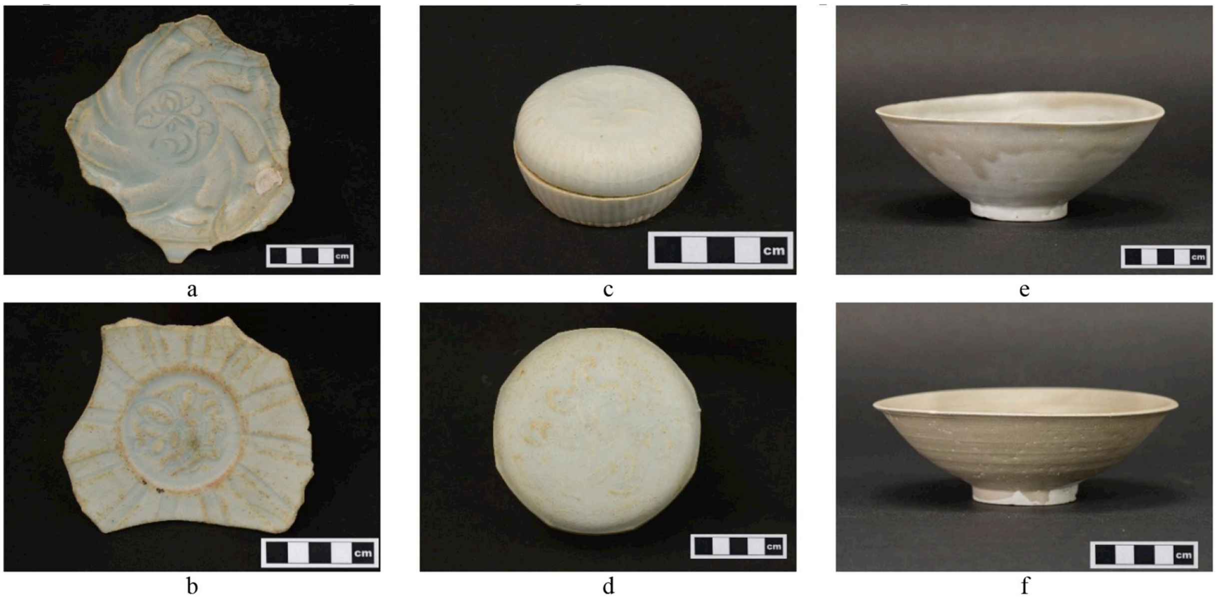

Fig. 2. Samples of *JSW qingbai* wares analyzed in this research. a: Type I dish, Cat. No. 345839. b: Type I dish, Cat. No. 345865. c: Type II box, Cat. No. 344280. d: Type II box, Cat. No. 344300. e: Type III bowl, Cat. No. 346757. f: Type III bowl, Cat. No. 347440. Photos © Field Museum.

Table 1
Frequencies of ceramic glaze and paste samples from the *Java Sea Shipwreck* collection analyzed using pXRF.

	Qingbai (Type I)	*Qingbai* (Type II)	*Qingbai* (Type III)	Total
Glaze	15	22	23	60
Paste	6	19	21	46

Table 2
Frequencies of ceramic glaze and paste samples from kiln sites analyzed using pXRF.

	Jingdezhen	Dehua	Huajiashan	Minqing	Total
Glaze	16	14	26	13	69
Paste	3	2	2	10	17

Table 3
Average elemental concentrations of ten measurements on Ohio Red Clay over the course of analysis (ppm).

	Average	SD	%RSD
Al	84941	4296	5%
Si	278090	7255	3%
S	956	195	20%
K	33284	294	1%
Ca	2507	821	33%
Ti	6736	133	2%
Mn	311	29	9%
Fe	52564	293	1%
Zn	160	68	42%
Rb	73	1	2%
Sr	71	1	2%
Y	38	2	5%
Zr	288	6	2%
Nb	19	1	4%
Pb	14	2	15%
Th	14	1	10%

ceramics from the *Java Sea Shipwreck* originated in China, pinpointing the exact provenance based on visual inspection is not an easy task because of the large number of kiln complexes producing stylistically similar products, the glaze color of which also varies significantly from bluish-white to grayish-white to grayish-green. Glazes of many *JSW qingbai* wares were also affected by postdepositional processes in a marine environment, which altered the color and condition of the glazes over time. Additionally, some *qingbai* wares from the *Java Sea Shipwreck* are small, broken fragments, making it hard to source these ceramics based on stylistic analysis.

1.3. Compositional analysis of Chinese porcelains

Compositional analysis of ceramic materials has emerged as a key methodology for archaeologists to investigate the production and movement of ceramics in the past (Bishop et al., 1982; Cecil, 2004; Descantes et al., 2001; Eerkens et al., 2002; Glascock, 1992; Hein et al., 1999; Hill et al., 2004; Kennett et al., 2002; Mirti et al., 2004; Neff et al., 1988; Sharratt et al., 2009; Skoglund et al., 2006). Several commonly used geochemical techniques for identifying the compositional signatures of ceramic samples are instrumental neutron activation analysis (INAA), inductively coupled plasma atomic emission spectroscopy or mass spectrometry (ICP-AES, ICP-MS), and X-ray fluorescence (XRF). Recently, these techniques have been increasingly applied to the elemental characterization of Chinese porcelains. Most studies analyzed ceramics from a single kiln complex to understand their elemental features (Cui et al., 2012; Duan et al., 2016; Li et al., 2012; Xie et al., 2009; Xu et al., 2017), or compared ceramic samples from different kiln complexes to identify the compositional differences between stylistically similar products from different kiln sites (He et al., 2016; Leung and Luo, 2000; Leung et al., 2000a; Li et al., 2005; Li et al., 2006; Ma et al., 2012; Wang et al., 2018; Yan et al., 2015; Yu and Miao, 1998; Zhu et al., 2010). Several geochemical studies have also been undertaken to trace the provenance of Chinese porcelains found overseas (Chen et al., 2016; Dias et al., 2013; Oka et al., 2009; Yap, 1991).

More recently, researchers have begun to use portable XRF (pXRF) to source Chinese blue-and-white porcelains found in East Africa and Southeast Asia. Cui et al. (2016) used pXRF to analyze the glaze compositions of 16^{th}-17^{th} century Kraak porcelains unearthed at Fort Jesus, Mombasa, Kenya. The compositional groups created based on pXRF data corresponded well to groups identified through stylistic analysis and confirmed that these Kraak porcelains came from three different sources—Jingdezhen (China), Zhangzhou (China), and Arita (Japan). This research also indicated that two elements found in glazes, Zr and

Table 4
Average elemental concentrations of ceramic samples from kiln complexes based on glaze composition (ppm).

	Jingdezhen (n = 16)			Dehua (n = 14)			Huajiashan (n = 26)			Minqing (n = 13)		
	Average	SD	%RSD	Average	SD	%RSD	Average	SD	%RSD	Average	SD	%RSD
Si	299381	20065	7%	290154	20138	7%	287757	22198	8%	314782	9580	3%
Al	47634	4027	8%	62381	6666	11%	64576	8968	14%	65661	8561	13%
K	17838	4649	26%	25518	5840	23%	29170	6007	21%	30341	4883	16%
Fe	6061	2182	36%	2489	751	30%	3438	728	21%	4588	1227	27%
Mn	400	121	30%	761	311	41%	533	199	37%	1305	245	19%
Ti	284	99	35%	346	82	24%	113	37	32%	771	114	15%
Rb	114	25	22%	72	10	14%	457	173	38%	71	9	12%
Sr	118	16	14%	107	38	36%	113	43	38%	216	51	23%
Y	13	4	30%	186	107	58%	35	8	23%	71	38	54%
Zr	51	19	37%	158	29	18%	36	3	7%	70	5	6%
Nb	16	5	31%	50	18	36%	67	3	5%	25	3	12%
Pb	21	7	32%	51	31	62%	56	27	47%	73	32	45%
Th	7	2	28%	35	9	26%	16	2	15%	42	2	4%

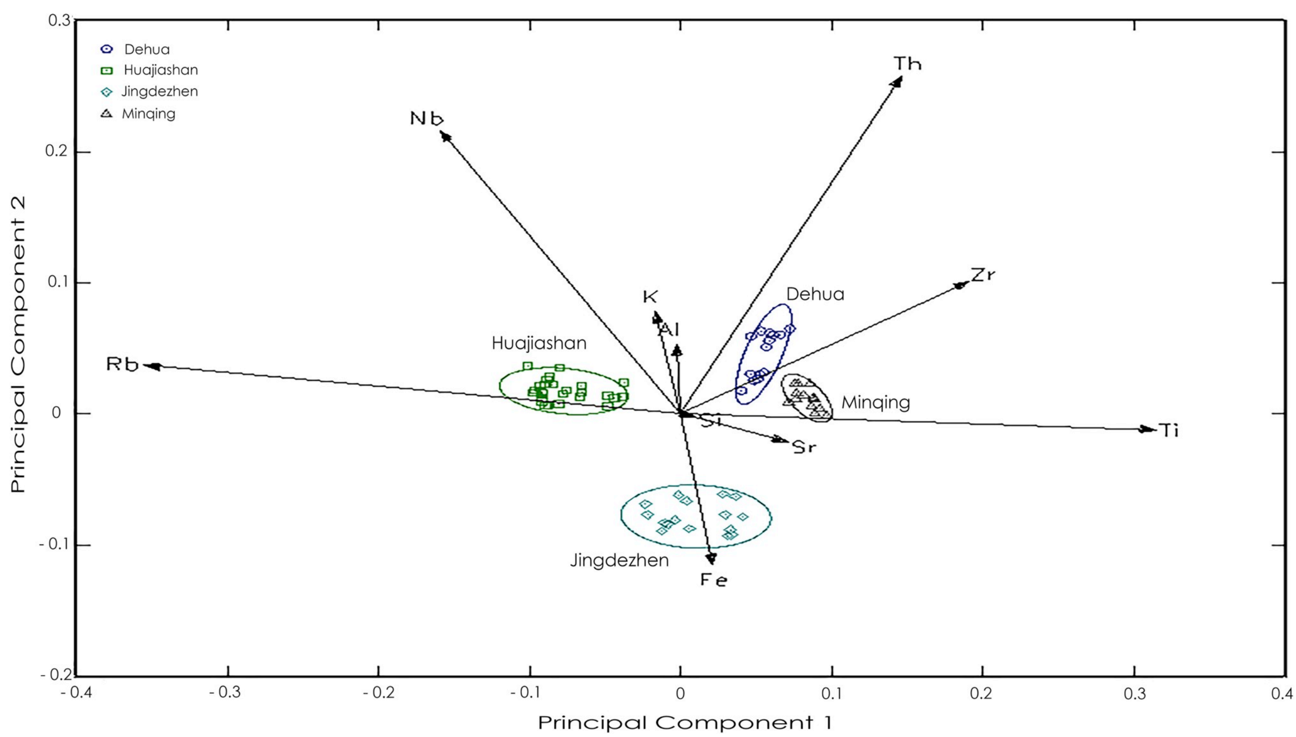

Fig. 3. R-Q mode biplot of principal components 1 and 2 based on glaze compositions of kiln samples. Ellipses represent 90% confidence intervals. PC1 summarizes 53.8% of the total variability in the data, and PC2 25.3%.

Th, can be used as key discriminators to distinguish Kraak porcelains from different kilns. Fischer and Hsieh (2017) used pXRF to analyze the pastes, glazes, and blue pigments of late 16th to early 17th century blue-and-white porcelains from the Philippines and the island of Java, and they successfully differentiated between Jingdezhen and Zhangzhou products. Although the glaze results for heavier elements such as Rb, Sr, Zr, and Th inevitably include a contribution from the vessel's body, they argued that "the key point here is that from a simple measurement on the white glazed surface, Jingdezhen and Zhangzhou blue-and-white export productions can be unambiguously differentiated with pXRF" (Fischer and Hsieh, 2017:22). These studies demonstrated that pXRF might be promising for sourcing Song and Yuan dynasties *qingbai* porcelains found overseas.

For the Chinese ceramics from the *Java Sea Shipwreck*, some compositional analysis has been undertaken on selected ceramics—including *qingbai*, green-glazed, and painted wares—using LA-ICP-MS. Niziolek (2015), for example, has successfully differentiated Jingdezhen *qingbai* wares from Fujian *qingbai* wares. Although LA-ICP-MS is preferred by many archaeologists for quantitative compositional studies of ceramic materials for its ability to determine concentrations of 50–60 elements and its low detection limits (Dussubieux et al., 2007), it has some drawbacks due to its semi-nondestructive nature and higher cost. Compared to LA-ICP-MS, pXRF is rapid, non-destructive, and low-cost; however, pXRF has high detection limits and relatively low accuracy. Preliminary investigations using a small selection of ceramic boxes from the *Java Sea Shipwreck* have shown that pXRF might be promising for differentiating between different types of *qingbai* wares based on paste composition (Niziolek, 2018). For this project, we expanded on this research to: (1) test whether pXRF is an effective method for distinguishing stylistically similar products from different kiln complexes and (2) identify the sources of some of the Chinese ceramics from the *Java Sea Shipwreck* using reference materials from kiln sites. Because the elemental compositions of distinctive styles of ceramics (e.g., *qingbai*, celadon, and black-glazed wares) are hypothesized to be significantly different (Li, 1998), this paper will focus on just one style of ceramics—*qingbai*.

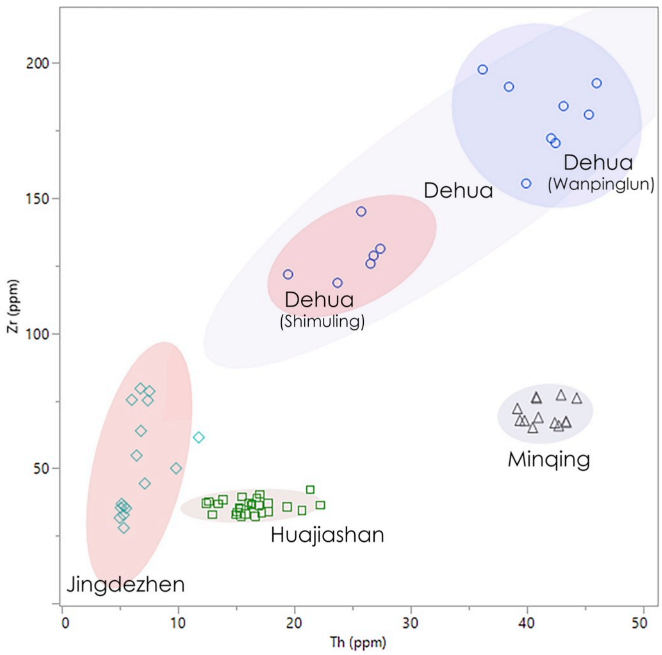

Fig. 4. Th-Zr biplot of ceramic glaze samples from Jingdezhen, Dehua, Huajiashan, and Minqing kiln complexes. Dehua samples form two groups representing two kiln sites. (Ellipses represent 90% confidence intervals.)

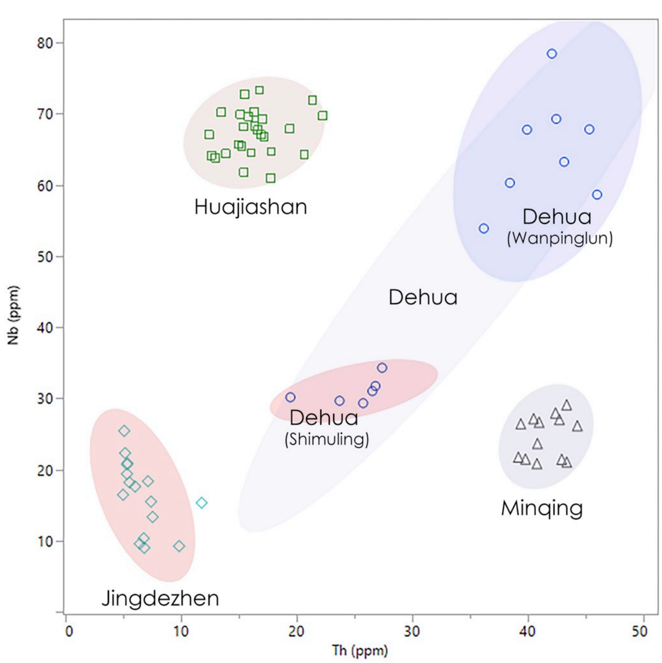

Fig. 5. Th-Nb biplot of ceramic glaze samples from Jingdezhen, Dehua, Huajiashan, and Minqing kiln complexes. Dehua samples form two groups, representing two kiln sites. (Ellipses represent 90% confidence intervals.)

2. Materials and methods

2.1. Samples

Samples analyzed for this project come from two sources—the *Java Sea Shipwreck* collection housed at the Field Museum in Chicago and kiln sites in China. A total of 60 ceramic samples from the *Java Sea Shipwreck* were selected for pXRF analysis and 69 ceramic samples from kiln complexes were used to create compositional reference groups (Appendix A).

We classified *JSW qingbai* wares into three types based on visual inspection. *Qingbai* Type I ceramics are the finest pieces, characterized by their fine, white paste, thin body, and translucent bluish glaze (Fig. 2-a and 2-b). The common form is a molded bowl or dish with foliated design. As mentioned above, the finest *qingbai* wares during the Song and Yuan periods were produced at Jingdezhen in Jiangxi province (Jiangxi Provincial Institute of Cultural Relics and Archaeology and Jingdezhen Museum of Civilian Kiln, 2007). We hypothesized Type I pieces to be authentic *qingbai* porcelains made at Jingdezhen. *Qingbai* Type II ceramics feature sugary white paste and light bluish-white or grayish-white glaze (Fig. 2-c and 2-d). A majority are molded boxes and small bottles. Archaeological work in Fujian has revealed several kiln complexes that produced *qingbai* boxes and bottles, such as Dehua, Huajiashan, Nan'an, and Pucheng (Fujian Museum, 1990; Lin and Zhang, 1993; Liu, 2013). It is difficult to determine the specific provenance of Type II ceramics based on stylistic analysis because of the great similarities between *qingbai* boxes and bottles from different kilns in Fujian, however, during a 2017 summer field season, one of the authors, Xu, found almost identical boxes and bottles at Dehua and Huajiashan kiln complexes. We hypothesized that the potential provenance of Type II ceramics might be Dehua and Huajiashan. *Qingbai* Type III ceramics are characterized by their grayish-white paste and glazes ranging from grayish-white to grayish-green (Fig. 2-e and 2-f). Most of the Type III pieces are plain bowls or ones decorated with a casually incised and combed floral or cloud pattern. Recent archaeological surveys at Minqing Yi kiln complex in Fujian revealed more than 100 kiln sites producing these type of ceramics (Yang, 2016). In 2017, Xu examined Minqing samples at the Fujian Museum and found that some of them are almost identical to Type III wares from the *Java Sea Shipwreck*. We hypothesized that the most likely provenance for Type III *qingbai* wares is Minqing Yi kiln.

Based on the hypotheses developed through stylistic analysis of *qingbai* ceramics from the *Java Sea Shipwreck*, we collected 69 ceramic samples from four different kiln complexes in China—Jingdezhen in Jiangxi Province and Dehua, Huajiashan, and Minqing in Fujian Province—for pXRF analysis (Fig. 1 and Appendix B). Because there were dozens of individual kiln *sites* producing similar products within each kiln *complex*, the ideal case would be to collect samples of different ware types from all kiln sites in order to more accurately determine the elemental signatures of various ware types from each kiln complex as a whole. However, the extremely large number of kiln sites in Jingdezhen and Fujian makes it difficult to access materials from all kiln sites at the current stage of the project. For this project, kiln samples were obtained from several kiln sites within each kiln complex. We think including samples from multiple kiln sites provides a more accurate general chemical signature for each complex represented.

2.2. Methods

Initially we wanted to analyze both the glazes and pastes of all the samples, however, not all pastes were accessible for pXRF analysis. Because most surfaces of porcelains are covered by glazes, pastes can only be analyzed by a pXRF analyzer if a significant and relatively flat, unglazed portion of the piece exists. In the case of the *JSW* samples, ceramics are generally more complete and often include a base or partial base. Nevertheless, due to the marine growth on the surface of some samples, not all pastes of samples with bases could be analyzed. In the case of kiln samples, ceramics are typically broken sherds. The exposed paste area is often very narrow and does not provide sufficient surface area for pXRF analysis. Thus, the glazes of all 129 samples were analyzed, while the pastes of only 63 samples were tested (Table 1 and Table 2).

Elemental analysis was conducted using a Thermo Niton XL3t GOLDD + portable X-ray fluorescence spectrometer with a silver (Ag) anode tube housed at the Elemental Analysis Facility (EAF) of the Field Museum. The main filter operates at voltage of 40 kV and current of

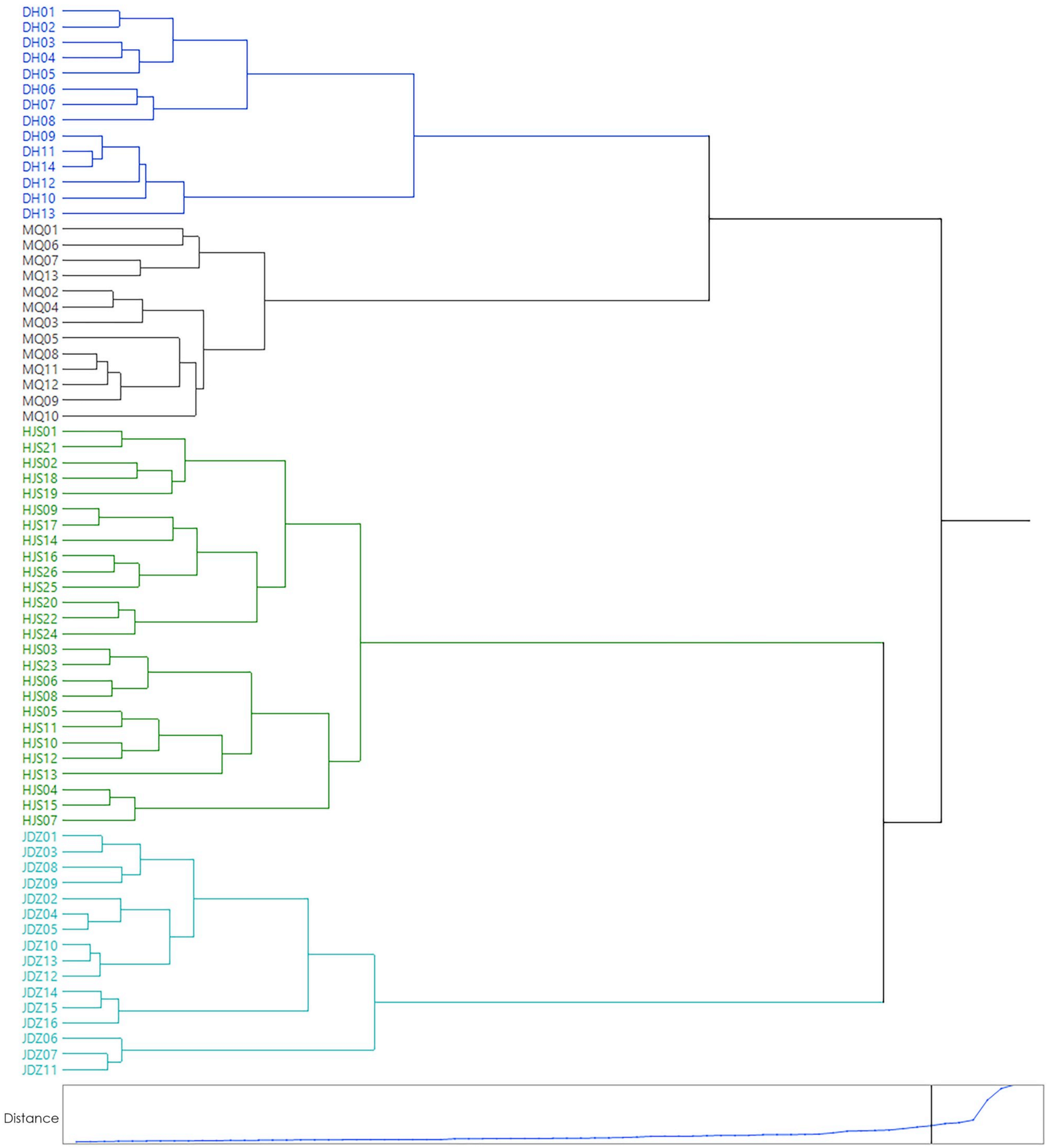

Fig. 6. Dendrogram from the hierarchical cluster analysis (Ward's method) using parts-per-millinon values of glaze compositions of kiln samples. (DH = Dehua, MQ = Minqing, HJS = Huajiashan, JDZ = Jingdezhen).

100 μA. Compositional data were collected with acquisition times set to 120s. Ohio Red Clay served as a measure of quality control. Test All Geo in the soils and minerals mode was used to measure 44 elements: magnesium (Mg), aluminum (Al), silicon (Si), phosphorus (P), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), scandium (Sc), titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), selenium (Se), rubidium (Rb), strontium (Sr), yttrium (Y), zirconium (Zr), niobium (Nb), molybdenum (Mo), lead (Pb), silver (Ag), cadmium (Cd), tin (Sn), antimony (Sb), tellurium (Te), cesium (Cs), barium (Ba), hafnium (Hf), tantalum (Ta), tungsten (W), rhenium (Re), gold (Au), mercury (Hg), lead (Pb), bismuth (Bi), thorium (Th), and uranium (U).

To prepare the data for statistical analysis, 28 elements with readings consistently below the limit of detection (LOD) of the pXRF analyzer were excluded from further analysis: Mg, P, Cl, Sc, V, Cr, Co, Ni, Cu, As, Se, Mo, Pb, Ag, Cd, Sn, Sb, Te, Cs, Ba, Hf, Ta, W, Re, Au, Hg, Bi, and U. Another three elements (Ca, S, and Zn) were also removed because the relative standard deviations (RSDs) obtained from ten measurements of Ohio Red Clay were high ($\geq$20%) (Table 3). After the initial data processing, 13 elements were retained for the glaze and paste analyses: Si, Al, K, Fe, Mn, Ti, Rb, Sr, Y, Zr, Nb, Pb, and Th. Statistical analysis was performed on the dataset using JMP Pro 14 and

Table 5
Average elemental concentrations of ceramic samples from kiln complexes based on paste composition (ppm).

	Jingdezhen (n = 3)			Dehua (n = 2)			Huajiashan (n = 2)			Minqing (n = 10)		
	Average	SD	%RSD	Average	SD	%RSD	Average	SD	%RSD	Average	SD	%RSD
Si	295243	19557	7%	267475	27872	10%	353683	89473	25%	265511	18784	7%
Al	79871	9059	11%	91205	177	0%	88052	11283	13%	78594	11529	15%
K	17741	1433	8%	18661	1336	7%	36943	213	1%	20959	4072	19%
Fe	11031	1367	12%	4189	1379	33%	4547	1728	38%	10570	3240	31%
Mn	400	105	26%	303	115	38%	226	60	27%	259	73	28%
Ti	1082	589	54%	670	292	44%	187	127	68%	984	279	28%
Rb	139	25	18%	62	19	31%	609	106	17%	68	9	14%
Sr	34	4	12%	12	1	13%	13	8	61%	26	12	47%
Y	19	5	28%	224	220	98%	25	3	11%	62	34	55%
Zr	41	11	26%	192	14	7%	41	4	11%	79	10	12%
Nb	23	6	26%	68	12	17%	73	6	8%	25	2	7%
Pb	38	12	31%	31	16	53%	121	3	3%	86	40	47%
Th	7	3	35%	45	4	8%	22	1	4%	49	4	9%

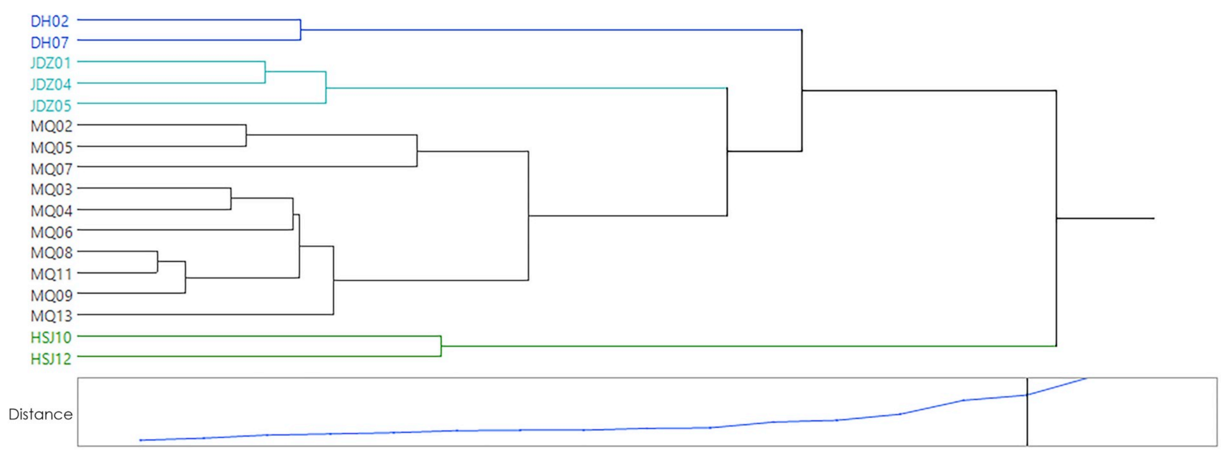

Fig. 7. Dendrogram from the hierarchical cluster analysis (Ward's method) using parts-per-million values of ceramic paste compositions of kiln samples. (DH = Dehua, MQ = Minqing, HJS = Huajiashan, JDZ = Jingdezhen).

GAUSS Runtime statistical routines, an Aptech Systems, Inc. program with routines developed by Hector Neff and Michael Glascock at the University of Missouri Research Reactor Center (MURR).

It is worth noting that when using pXRF to analyze the clear glaze layer of porcelains it may be possible that high atomic elements reflect the contribution of both the glaze and the porcelain body (Bezur and Casadio, 2012). Clear glaze layers of Chinese porcelains are typically 200–500 microns thick (Leung et al., 2000; Yu and Miao, 1996). For the heavier elements (such as Rb, Sr, Y, and Zr) the analysis depth of the clear glaze ranges from 382 to 645 microns (Bezur and Casadio, 2012:262), so the interference from the underlying body cannot be totally avoided. Nevertheless, surface analysis of the clear glaze layers of porcelains with pXRF can still be considered a valid method because the goal of using pXRF on glazes is to distinguish porcelains from different kilns rather than quantification analysis of the glaze compositions. Additionally, studies showed that although the absolute elemental concentrations generated with pXRF might not be as accurate as other high-sensitivity techniques, such as NAA and ICP-MS, pXRF can still be used to identify geochemical groups that closely correlate to those indicated by other methods (Forster et al., 2011; Holmqvist, 2017; Hunt and Speakman, 2015; Johnson, 2014; Mitchell et al., 2012; Speakman et al., 2011). Thus, the purpose of using pXRF for this project was to examine whether pXRF can be used to effectively differentiate Chinese porcelains from different kiln complexes and to source *qingbai* porcelains found in the *Java Sea Shipwreck*.

3. Results and discussion

3.1. Ceramic compositional differences between kiln complexes

Ceramic samples from kiln complexes in China were analyzed first to determine whether pXRF data can be used to differentiate between sherds from different production areas based on glaze and paste compositions.

3.1.1. Glazes

As mentioned above, 13 elements were retained for the glaze analysis: Si, Al, K, Fe, Mn, Ti, Rb, Sr, Y, Zr, Nb, Pb, and Th. Table 4 lists the average elemental concentrations of the glazes on ceramics from different kiln complexes. As we can see from the table, the standard deviations (SDs) and relative standard deviations (RSDs) are generally high for the four kiln complexes. Compositional variation within a kiln complex could be explained by differences in elemental signatures of kiln sites within that complex. However, three elements (Mn, Y, and Pb) have rather high %RSDs (often > 30%). These three elements were excluded from further data analysis. The remaining data with ten elements (Si, Al, K, Fe, Ti, Rb, Sr, Zr, Nb, and Th) were processed using well-established statistical routines, including principal component (PC) analysis, hierarchical cluster analysis, and group membership probabilities based on Mahalanobis distance calculations (Baxter, 1992, 2001; Baxter and Heyworth, 1989; Bishop and Neff, 1989; Dussubieux et al., 2007; Glascock, 1992; Neff, 1994).

Principal components were first calculated using the ten elements as a means of rapidly examining multivariate patterning in the data. A

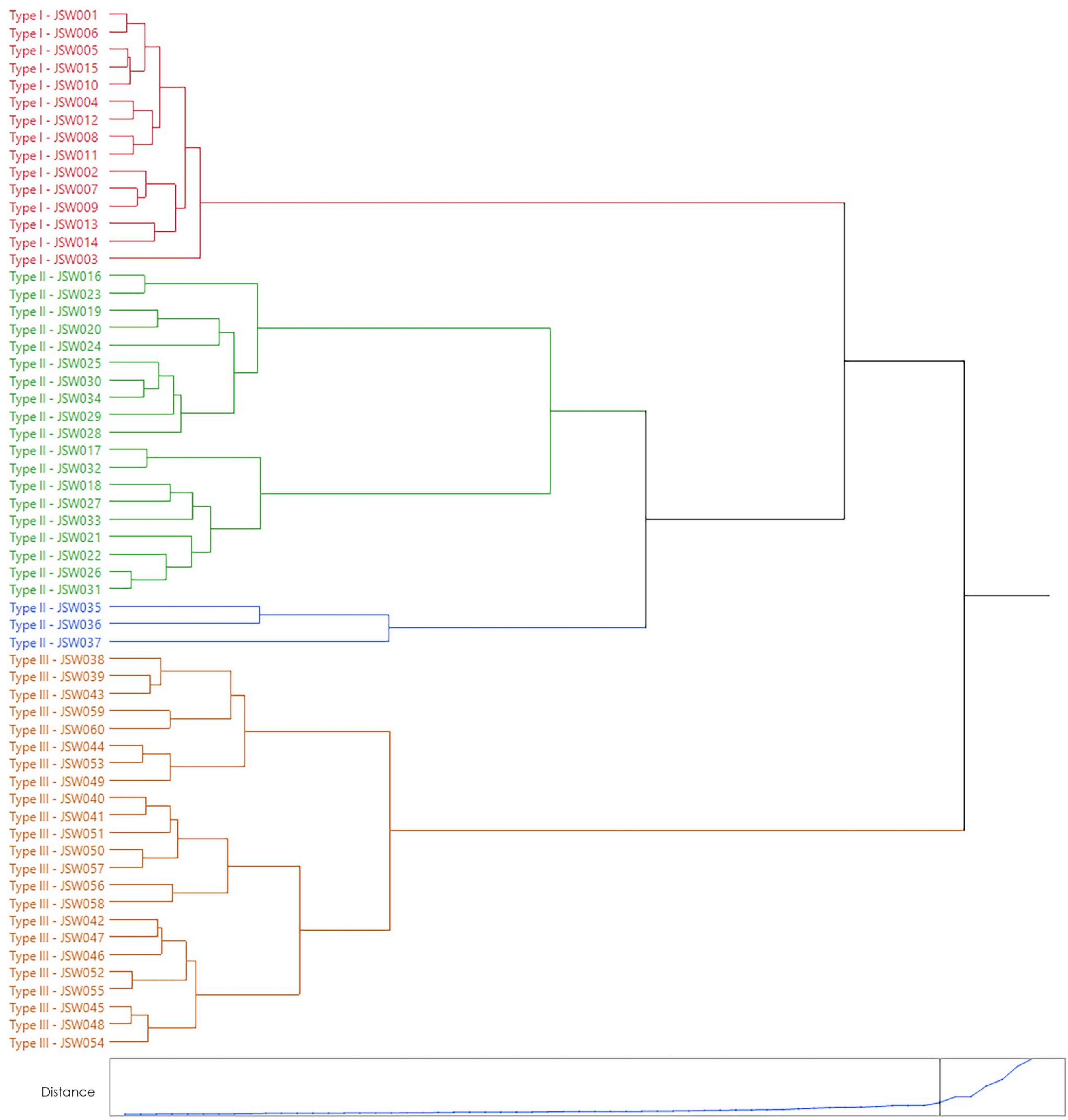

Fig. 8. Dendrogram from the hierarchical cluster analysis (Ward's method) of glaze compositions of *JSW* ceramic samples.

biplot of the first two principal components, which account for 79.1% of the total variance in the data, shows clear differences between *qingbai* wares from the four different kiln complexes (Fig. 3). *Qingbai* glazes of porcelains produced at Jingdezhen are characterized by higher concentrations of Fe, and lower concentrations of Nb and Th. Dehua samples have higher amounts of Zr and Th, and lower amounts of Fe and Rb. Huajiashan pieces have higher concentrations of Rb and Nb, and lower concentrations of Ti and Zr. Minqing *qingbai* glazes are characterized by higher concentrations of Ti and Sr, and lower quantities of Rb. The different *qingbai* glaze compositional groups can clearly be seen on the biplot of principal components 1 and 2.

Mahalanobis distance probabilities of group membership were calculated for all kiln samples using the first four principal components, which account for more than 90% of the elemental variance in the dataset. Results show that probabilities of samples from one kiln complex falling into other groups are extremely low (< 1%), confirming that the four kiln complexes are four valid reference groups.

The biplots of Th—Zr and Th—Nb also clearly display the compositional differences between pieces from different kiln complexes (Figs. 4 and 5). Furthermore, based on the Th—Zr and Th—Nb biplots, we suggest that there are two compositional groups within the Dehua kiln complex represented. These groups correspond to two different kiln sites at Dehua. Samples in the upper right compositional group on Figs. 4 and 5 all came from the Wanpinglun kiln site in Gaide village, and samples in the middle group all came from the Shimuling kiln site in Baomei village. The two kiln sites are about 8 km apart. Because of the formation of these subgroups, we suggest that pXRF also might be an effective method for differentiating porcelains from different

Table 6
Group membership probabilities of glaze samples from *Java Sea Shipwreck* ceramics based on Mahalanobis distance calculations using the first five principal component values.

Sample #	Catalog #	Stylistic Group	Jingdezhen	Dehua	Huajiashan	Minqing	Best Group
JSW001	344476	Type I	59.088	0.000	0.000	0.000	Jingdezhen
JSW002	345810	Type I	11.124	0.000	0.000	0.000	Jingdezhen
JSW003	345816	Type I	91.638	0.000	0.000	0.000	Jingdezhen
JSW004	345819	Type I	51.000	0.000	0.000	0.000	Jingdezhen
JSW005	345838	Type I	49.335	0.000	0.000	0.000	Jingdezhen
JSW006	345839	Type I	60.962	0.000	0.000	0.000	Jingdezhen
JSW007	345840	Type I	38.008	0.000	0.000	0.000	Jingdezhen
JSW008	345842	Type I	50.713	0.000	0.000	0.000	Jingdezhen
JSW009	345859	Type I	38.101	0.000	0.000	0.000	Jingdezhen
JSW010	345864	Type I	57.828	0.000	0.000	0.000	Jingdezhen
JSW011	345865	Type I	48.268	0.000	0.000	0.000	Jingdezhen
JSW012	350400	Type I	72.813	0.000	0.000	0.000	Jingdezhen
JSW013	350407	Type I	15.028	0.000	0.000	0.000	Jingdezhen
JSW014	350409	Type I	9.457	0.000	0.000	0.000	Jingdezhen
JSW015	351111	Type I	31.916	0.000	0.000	0.000	Jingdezhen
JSW016	344263	Type II	0.000	21.080	0.000	0.122	Dehua
JSW017	344280	Type II	0.000	4.110	0.001	0.010	Dehua
JSW018	344287	Type II	0.000	17.667	0.000	0.370	Dehua
JSW019	344300	Type II	0.000	27.232	0.002	0.046	Dehua
JSW020	344378	Type II	0.000	3.080	0.003	0.061	Dehua
JSW021	344385	Type II	0.000	15.786	0.000	0.240	Dehua
JSW022	344629	Type II	0.000	64.429	0.003	0.036	Dehua
JSW023	344877	Type II	0.000	35.150	0.001	0.139	Dehua
JSW024	344898	Type II	0.000	2.721	0.001	0.010	Dehua
JSW025	344908	Type II	0.000	56.843	0.012	0.002	Dehua
JSW026	345069	Type II	0.000	25.481	0.001	0.010	Dehua
JSW027	345159	Type II	0.000	21.281	0.001	0.056	Dehua
JSW028	345262	Type II	0.000	39.726	0.007	0.002	Dehua
JSW029	350324	Type II	0.000	64.373	0.001	0.008	Dehua
JSW030	350339	Type II	0.000	32.116	0.002	0.007	Dehua
JSW031	350343	Type II	0.000	55.276	0.001	0.014	Dehua
JSW032	350351	Type II	0.000	1.225	0.000	0.007	Dehua
JSW033	350352	Type II	0.000	70.867	0.000	0.023	Dehua
JSW034	350365	Type II	0.000	12.392	0.003	0.002	Dehua
JSW035	344284	Type II	0.001	0.000	12.876	0.000	Huajiashan
JSW036	344710	Type II	0.001	0.000	6.954	0.000	Huajiashan
JSW037	344964	Type II	0.004	0.000	25.180	0.000	Huajiashan
JSW038	345402	Type III	0.002	0.067	0.000	90.379	Minqing
JSW039	345715	Type III	0.002	0.058	0.000	63.269	Minqing
JSW040	346108	Type III	0.001	0.364	0.000	7.527	Minqing
JSW041	346109	Type III	0.001	0.730	0.001	4.840	Minqing
JSW042	346110	Type III	0.001	1.556	0.000	14.382	Minqing
JSW043	346325	Type III	0.002	0.046	0.000	88.964	Minqing
JSW044	346757	Type III	0.001	0.061	0.000	74.250	Minqing
JSW045	346758	Type III	0.002	0.045	0.000	94.220	Minqing
JSW046	347067	Type III	0.001	0.633	0.000	23.052	Minqing
JSW047	347350	Type III	0.002	0.132	0.000	32.499	Minqing
JSW048	347440	Type III	0.001	0.079	0.000	89.952	Minqing
JSW049	347441	Type III	0.001	0.185	0.000	11.341	Minqing
JSW050	347442	Type III	0.001	0.131	0.000	13.194	Minqing
JSW051	347464	Type III	0.000	0.252	0.000	1.064	Minqing
JSW052	347465	Type III	0.001	0.804	0.000	18.504	Minqing
JSW053	347494	Type III	0.001	0.200	0.000	42.904	Minqing
JSW054	347495	Type III	0.002	0.131	0.000	40.484	Minqing
JSW055	347496	Type III	0.001	0.497	0.000	36.763	Minqing
JSW056	348712	Type III	0.001	0.014	0.000	3.937	Minqing
JSW057	348713	Type III	0.001	0.096	0.000	9.234	Minqing
JSW058	348714	Type III	0.001	0.013	0.000	18.034	Minqing
JSW059	349877	Type III	0.001	0.016	0.000	50.510	Minqing
JSW060	349878	Type III	0.002	0.006	0.000	50.944	Minqing

production locales at a smaller scale, that of the kiln site. This can serve as the basis for future investigations.

A hierarchical cluster analysis of the 69 glaze samples based on the concentrations of all 16 elements provides an even more straightforward visualization of the observations discussed above. The resulting dendrogram clearly reveals four compositional groups, with samples from each kiln complex grouping together (Fig. 6). Additionally, the two clusters within the Dehua kiln complex on the dendrogram correspond well to kiln sites at Dehua, with samples (DH01–DH08) all from the Wanpinglun kiln and samples (DH09–DH14) all from the Shimuling kiln site. Although the clusters within other three kiln complexes do not perfectly correspond to different kiln sites, this result again demonstrates the potential of using pXRF to further detect compositional groups within a kiln complex.

a b c

Fig. 9. *Qingbai* Type II wares that were assigned to the Huajiashan kiln complex based on glaze composition. a: JSW035, box, Cat. No. 344284. b: JSW036, box, Cat. No. 344710. c: JSW037, box, Cat. No. 344964. Photo © Field Museum.

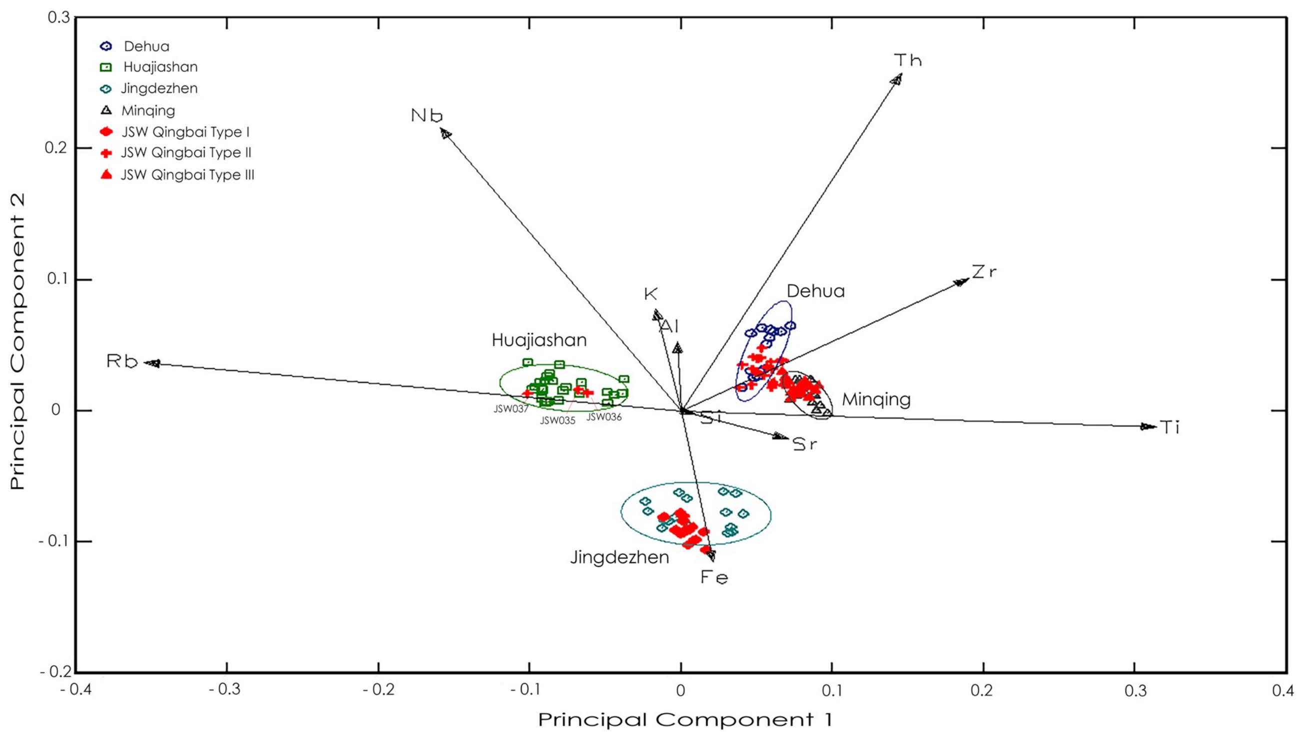

Fig. 10. R-Q mode biplot of principal components 1 and 2 based on glaze compositions of kiln samples and *JSW* ceramic samples. Ellipses represent 90% confidence intervals. PC1 summarizes 53.8% of the total variability in the data, and PC2 25.3%.

3.1.2. Pastes

As mentioned above, 13 elements were retained initially for the paste analysis: Si, Al, K, Fe, Mn, Ti, Rb, Sr, Y, Zr, Nb, Pb, and Th. Table 5 lists the average elemental concentrations of ceramic pastes from the different kiln complexes. Again, the standard deviations (SDs) and relative standard deviations (RSDs) are generally high, which might be caused by the compositional variation within a kiln complex. Four elements (Ti, Sr, Y, and Pb) with very high %RSDs (often > 30%) were excluded from further data analysis.

The remaining data with nine elements (Si, Al, K, Fe, Mn, Rb, Zr, Nb, and Th) were processed using similar statistical procedures used for the glaze data. However, due to the limited number of samples in the Jingdezhen, Dehua, and Huajiashan groups compared to elemental variables, principal components analysis and group membership probabilities could not be performed. The dendrogram plot of the hierarchical cluster analysis of all 17 paste samples based on the concentrations of the nine elements reveals four compositional groups, with samples from each kiln complex grouping together (Fig. 7). Although the results show that products from the four different kiln complexes can be differentiated through paste analysis, any interpretations must be tentative because of the small number of samples in each group.

3.2. Sourcing qingbai ceramic samples from the Java Sea Shipwreck

After the four kiln reference groups were established, pXRF data from *qingbai* samples from the *Java Sea Shipwreck* were analyzed using similar statistical procedures to identify the potential sources of these high-fired pieces.

3.2.1. Glazes

The dendrogram plot of the hierarchical cluster analysis of all 60 glaze samples from the *Java Sea Shipwreck* based on the concentrations of ten elements (Si, Al, K, Fe, Ti, Rb, Sr, Zr, Nb, and Th) shows that there might be at least four different compositional groups (Fig. 8). These groups correspond well to stylistic differences. Samples that were visually classified as Type I were grouped together in hierarchical cluster analysis. Samples classified as Type II *qingbai* wares were broken down into two (or possibly three) sub-groups. Type III samples grouped together for the most part, although they might be further broken into two sub-groups.

Group membership probabilities were then calculated using the

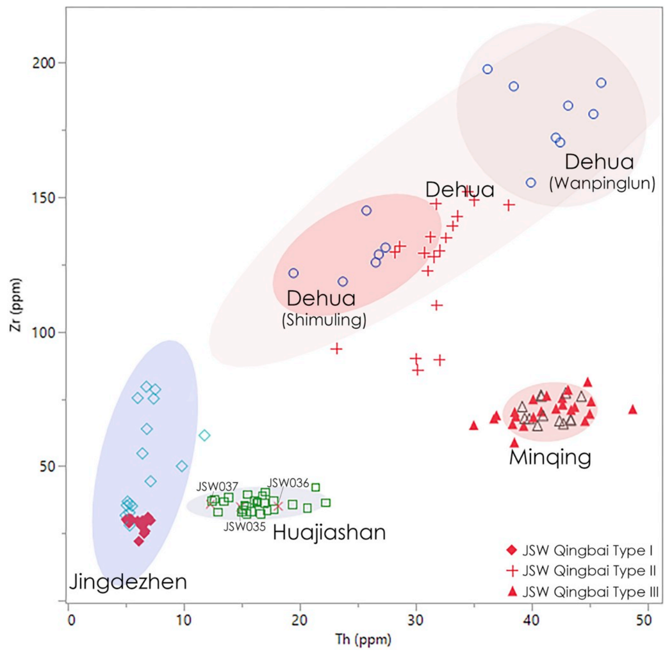

Fig. 11. Th-Zr biplot of glaze compositions of kiln samples and *JSW* ceramic samples. Dehua samples form two groups, representing two kiln sites. (Ellipses represent 90% confidence intervals.)

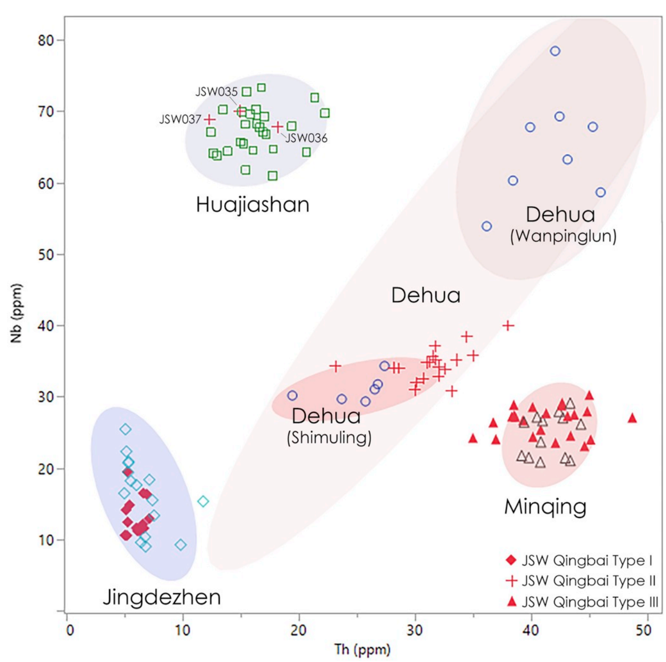

Fig. 12. Th-Nb biplot of glaze compositions of kiln samples and *JSW* ceramic samples. Dehua samples form two groups, representing two kiln sites. (Ellipses represent 90% confidence intervals.)

results of the glaze analysis of kiln samples as four compositional reference groups and *JSW* glaze samples as an unknown dataset. Results of group membership probabilities are consistent with the hierarchical cluster analysis (Table 6). Samples in the *JSW* Type I group had the highest probability of being from the Jingdezhen kiln complex. *JSW* Type II samples were assigned to two different kiln complexes, Dehua and Huajiashan, based on group membership probabilities. Three samples—JSW035, JSW036, and JSW037 (Fig. 9), which appear as a sub-group in the hierarchical cluster analysis, were assigned to the Huajiashan kiln complex. All other samples in the *JSW* Type II group were assigned to the Dehua kiln complex. *JSW* Type III samples were all assigned to the Minqing kiln complex.

Assigning *qingbai* porcelains from the *Java Sea Shipwreck* to potential kiln complexes can also be done by examining biplots of the first two principal components, as well as biplots of Th−Zr and Th−Nb (Figs. 10 and 11, and 12). Different types of *qingbai* ceramics form separate groups and match well with the reference groups. The three samples (JSW035, JSW036, and JSW037) in Type II fall into the Huajiashan group, which is consistent with assignments made using Mahalanobis distance calculations. All other samples generally fall within the 90 percent confidence intervals of their expected reference groups, with Type I samples in the Jingdezhen group, other Type II samples in the Dehua group, and Type III samples in the Minqing group. Because kiln samples analyzed here represent only some of the kiln sites in each kiln complex, it makes sense that some samples fall slightly out of the confidence intervals. These samples probably came from other kiln sites in the assigned kiln complex instead of from other kiln complexes.

Additionally, the Th−Zr and Th−Nb biplots demonstrate the ability of further assigning *JSW qingbai* samples to a specific kiln site within the kiln complex. Based on Figs. 11 and 12, we think that Type II (Dehua) samples probably came from kilns near the Shimuling kiln instead of the Wanpinglun kiln site.

3.2.2. Pastes

The dendrogram plot of the hierarchical cluster analysis of all 46 paste samples from the *Java Sea Shipwreck* ceramics based on the concentrations of nine elements (Si, Al, K, Fe, Mn, Rb, Zr, Nb, and Th) shows patterns similar to classifications based on stylistic and glaze compositional analyses (Fig. 13). Samples from each stylistic type grouped together. One sample (JSW035) that was assigned to the Huajiashan group based on the glaze composition was placed with other samples from the Type II group based on paste composition, although it formed a sub-group by itself.

Principal components of *JSW* paste samples were then calculated using the nine retained elements (Si, Al, K, Fe, Mn, Rb, Zr, Nb, and Th). A biplot of the first two principal components, which account for 75% of the total variability in the data, illustrates the difference between the types of *qingbai* wares (Fig. 14). One sample (JSW035) fell out of the 90% confidence interval of the *JSW* Type II group, but it is near the Huajiashan kiln samples. This result is consistent with the previous assumption that the provenance of this sample may be the Huajiashan complex. The two Dehua samples fell on or near the confidence interval of the Type II group, suggesting that the provenance of other Type II samples is Dehua. Three Jingdezhen samples fell into or near the Type I group. The provenance of the Type I group is probably Jingdezhen. The majority of Minqing samples are within the 90% confidence interval of the Type III group, which again shows that the provenance of Type III is most likely Minqing.

Although the results are generally consistent with the provenance assignments based on glaze compositions, the compositional groupings based on pastes are not quite as clear as those generated through the glaze analysis. Sourcing *qingbai* porcelains from the *Java Sea Shipwreck* based on paste compositions is more challenging than using glaze compositions. Not all the kiln reference groups have enough samples for which the pastes are accessible for pXRF analysis to represent the paste compositions of each kiln complex.

4. Conclusions

The results of this project demonstrate the effectiveness of using pXRF for discriminating stylistically similar products from different kiln complexes and identifying the potential sources of ceramics found along ancient maritime trade routes. *Qingbai* porcelains from four kiln complexes in China—Jingdezhen, Dehua, Huajiashan, and Minqing—can be differentiated through both glaze and paste compositions. The findings also serve to demonstrate that pXRF can be

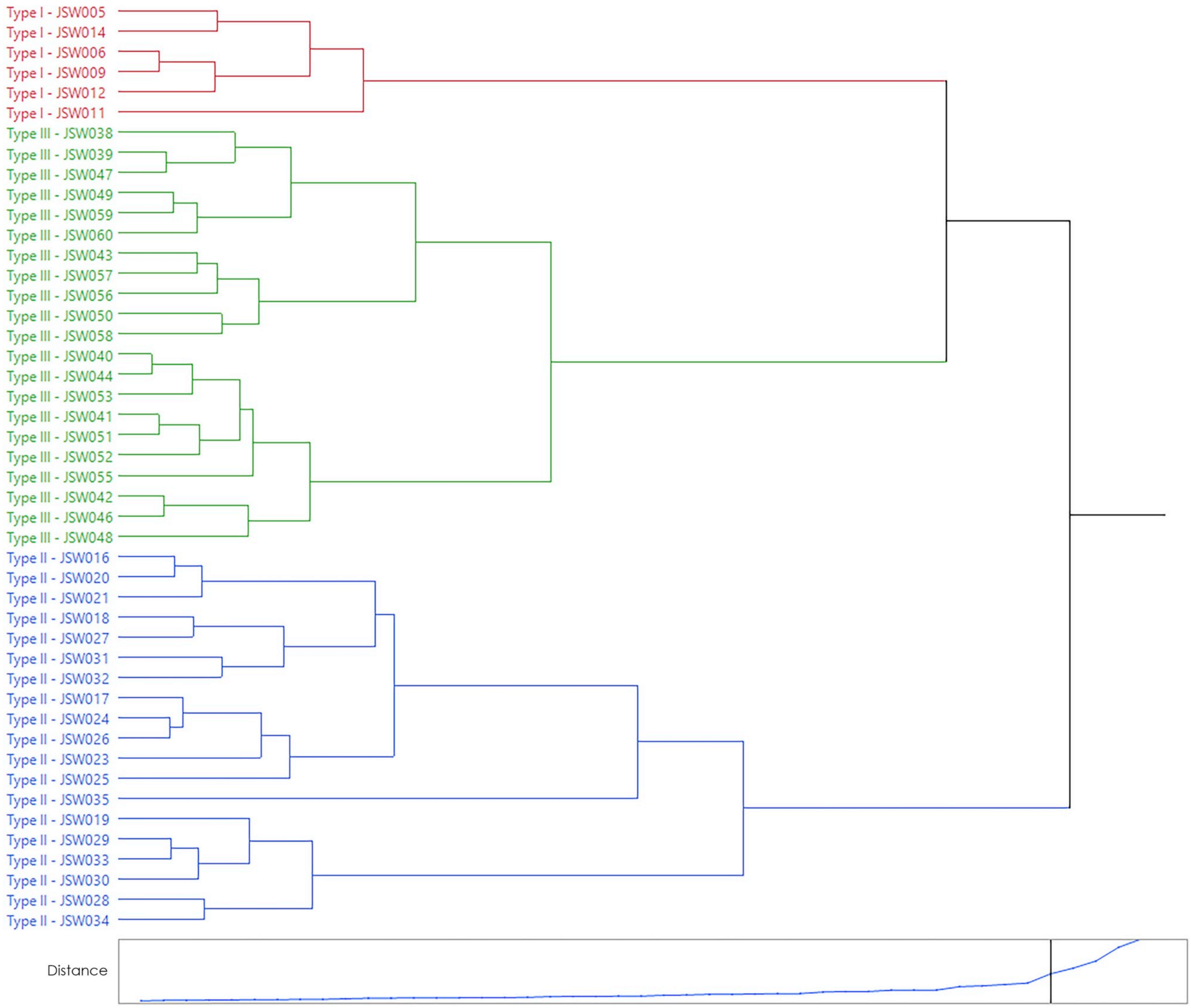

Fig. 13. Dendrogram from the hierarchical cluster analysis (Ward's method) of paste compositions of *JSW* ceramic samples.

employed to characterize the compositional differences between kiln sites within a kiln complex. Samples from two different kiln sites at Dehua—Wanpinglun and Shimuling—were clearly distinguished through glaze compositions.

Through pXRF analysis of *qingbai* samples from the *Java Sea Shipwreck*, we have confirmed and, in some instances, refined stylistic groups. Three stylistic types of *qingbai* porcelains from the shipwreck could be assigned to four compositional groups, and these groups corresponded well to reference groups established using porcelain samples from the four selected kiln complexes in China. All *JSW* Type I wares were sourced to the Jingdezhen kiln complex. Type II ceramics appear to be from two sources, with the majority being from the Dehua kiln complex (possibly near the Shimuling kiln site) and three samples from the Huajiashan kiln complex. Type III ceramics were unambiguously assigned to the Minqing kiln complex, which is near the port of Fuzhou. Minqing ceramics form one of the largest categories of ceramics in the *JSW* collection. In addition, potters and traders working in the Minqing, Huajiashan, and Jingdezhen areas would have had access to the Minjiang River and its tributaries for transporting goods to Fuzhou. Based on the results of this analysis we suggest that the route of the *Java Sea Shipwreck* vessel be re-evaluated (also see Niziolek et al., 2018; Flecker, 2018). Instead of Quanzhou being the initial port of lading as previously thought, we now think the ship probably sailed from the port of Fuzhou then on to Quanzhou to load porcelains from the Dehua kiln complex. To serve diverse markets overseas, merchants relied on access to a variety of ware types in terms of form, design, and quality. In this instance, finely made *qingbai* wares from Jingdezhen would have been most treasured. Similar pieces of lesser quality, such as those from Huajiashan, Minqing, and Dehua, were produced in larger quantities and fulfilled the needs of numerous societies in Southeast Asia and other parts of the Indian Ocean World.

Comparative analysis of glaze and paste compositions also finds that glaze compositions are sufficient to distinguish between products from different kiln complexes as well as to source *qingbai* porcelains from the *Java Sea Shipwreck*. Because pastes are not always accessible for the pXRF analysis of porcelains, it is easier to analyze glazes. Although readings of high atomic elements in the glazes might reflect the contribution of both the glaze and the porcelain body, pXRF analysis of the glaze surface can still be considered an effective method to source Chinese porcelains because the key point is to identify compositional groups rather than providing quantitative measurements of the glazes. In conclusion, considering the convenient, portable, non-destructive, and low-cost characteristics of pXRF, the validation of the methodology of using pXRF to source Chinese ceramics will allow for rapid identification of the potential sources of porcelains found in ancient maritime trade routes. These results might be further refined (e.g., specific kilns identified) using more sensitive and quantitative techniques such as LA-ICP-MS.

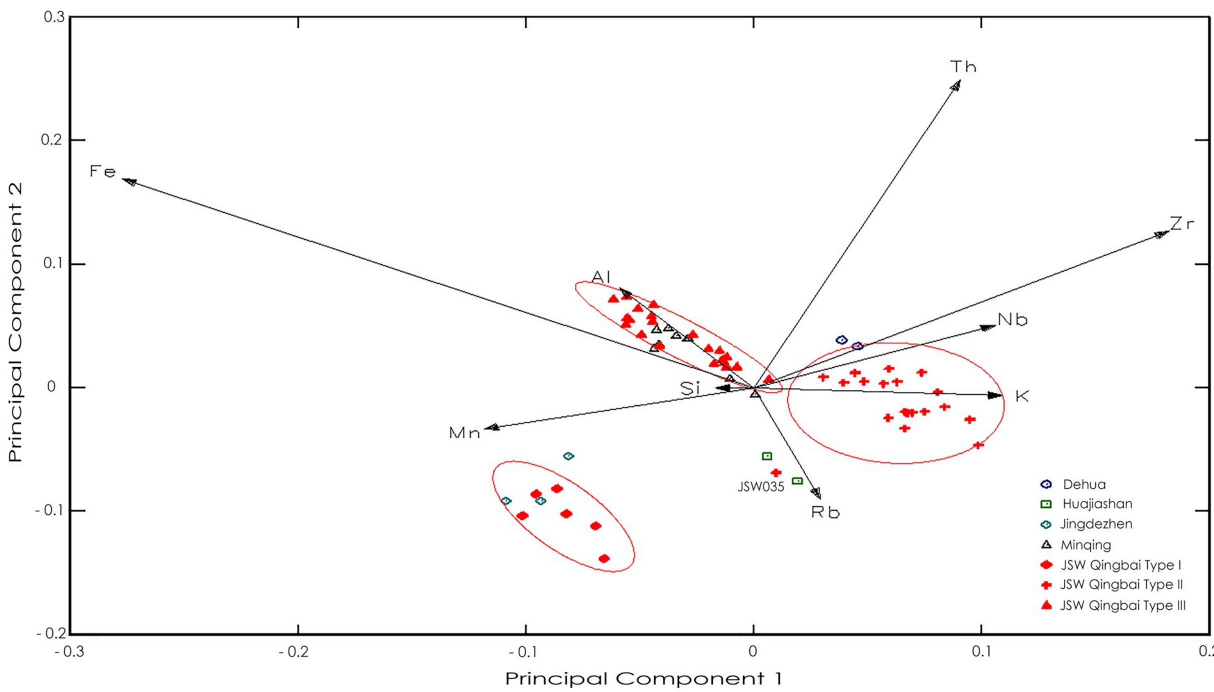

Fig. 14. R-Q mode biplot of principal components 1 and 2 based on paste compositions of *JSW* ceramic samples, with kiln samples added. Ellipses represent 90% confidence intervals. PC1 summarizes 42.1% of the total variability in the data, and PC2 32.9%.

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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.2018.12.010.

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