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Clay resources at early Chinese kiln sites: The search for a reliable INAA fingerprint

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29.1. Introduction

The primary foci of this paper are: (i) a discussion of an important group of ten stoneware sherds (Fig. 29.1) which, though recovered during excavation of the Islamic port of Siraf on the eastern side of The Gulf (Whitehouse 1973), assuredly were made at the Tongguan kilns (See-Yiu Lam 1991) which are situated on the eastern bank of the Xiang tributary of the Yangzi River, about 27 kilometres north of the Changsha (Hunan province, Fig. 29.2); and, to put those Tongguan analyses in context, (ii) a statistical overview of the now quite extensive literature (particularly in the proceedings of three recent Shanghai Conferences: see SICAS 1986; Li Jiazhi & Chen Xianqiu 1989; Li Jiazhi & Chen Xianqiu 1992: also Li Huhou 1988) on the composition of early stonewares (6th–12th centuries AD) recovered from several other kiln sites in China.

The chemical composition of the clay body of each of the Tongguan wares was determined by instrumental neutron activation analysis (INAA) at the SLOWPOKE Reactor Facility of the University of Toronto, using a flux of $\leq 2\times10^{11}$ neutrons. cm^{-2} s^{-1}. (See Hancock 1978, for fuller details of the experimental procedure and elemental detection limits.) Fragments were cut from each sherd, then abraded free of the surface glaze, to leave a sample typically weighing about 300mg.

29.2. Tongguan stoneware

The major and minor elements of the Tongguan stoneware from Siraf are presented in Table 29.1. Here our INAA data are presented first as measured as elemental contents, then after they have been converted to oxide equivalents. In that way readers familiar with the general literature on Chinese kiln products can make a rapid comparison with the many ways in which compositional data, both tabular and graphic, are presented. In the later sections of this paper we have used only our elemental contents, so as to be consistent with other INAA studies of this kind.

It is clear from Table 29.1 that the concentrations of magnesium and calcium are very low and quite variable in these samples. In fact they also exhibit quite poor reproducibility within individual sherds, with their measured contents varying a factor of two in several instances. The high standard deviations recorded here suggest that the minor levels of magnesium and calcium which *do* occur in these stoneware bodies are caused by dispersed mineral contaminants, which may not be helpful in the characterisation of the primary clay stock used at the site. Similar levels of data scatter were reported by Chinese scholars at the three Shanghai symposia mentioned above, for almost every kiln site excavated in recent years.

Thus, with INAA, there are really only four major elements—aluminium, potassium, iron, and titanium—which are reproducible enough to hold a reasonable potential for kiln site differentiation. For Tongguan, we were able to compare our data with those obtained by Chen Xianqiu and his colleagues, for a variety of ware types also dating to the Tang Dynasty (AD 618–906), and establish relatively good agreement (see Table 29.2, left and centre). As a consequence, the Tongguan data displayed later in Figs. 29.3a–d are a combination of these two studies. We assume that the raw material for these Tongguan wares was the kaolinitic clay known as China stone which occurs widely throughout south central and southern China. It contains potassium-rich micas and fine-grained quartz as its main mineralogical constituents (Hurlbut 1971). The moderate, consistent level of iron (about 1.6%) in this clay type prevented the Tongguan kilns from producing the near-white fabrics upon which some northern kilns of the time built their reputation.

We were also able to compare these Tongguan data to those obtained for other Tan Dynasty wares from the Chengguan kilns which lie only a few kilometres north west of Tongguan (Li Jiazhi and Chen Xianqiu 1992: A-18). The clay used at these sites seem to be very similar, except that Chengguan wares have a slightly lower iron content (see Table 29.2, right), and so a somewhat lighter body colour.

At this point, we would have accepted the often stated view (see, for example, several contributions in SICAS 1986) that the stoneware products of south central China (Yangzi River basin) and north central China (Yellow River basin)—together with sites even further north (such as Changbo, in Liaoning province: see Fig. 29.1)—could be differentiated by a rule-of-thumb akin to "moderate titanium, moderate iron" *versus* "high titanium, low iron" (Fleming & Swann 1992). When we reviewed compositional data presented at the two earlier Shanghai conferences, however, we were surprised to find that two of the primary elements—titanium and potassium—varied sufficiently at almost *all* Chinese kiln sites, northern and southern, as to make compositional differentiation between them quite ambiguous (Figs. 29.3a–b). There may even be a confusing bimodal behaviour in the potassium contents for wares from both the Gongxian and the Lincheng kiln sites, each of which produced prestigious whitewares for

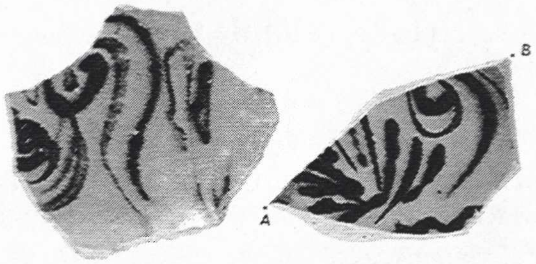

Figure 29.1: Two sherds (section AB, 9.1 cm) of underglaze-painted stoneware from the Chinese kiln site of Tonnguan, though excavated in an early 9th century AD *context at the Islamic port site of Siraf. Royal Ontario Museum, Toronto: inv. #986.407.1215 and 1229.*

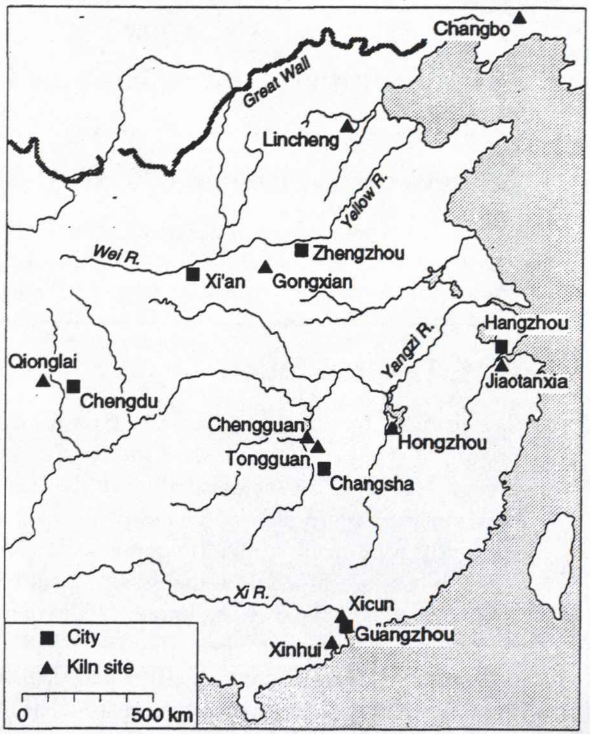

Figure 29.2: Map of China showing some of the primary kiln sites of the latter half of the 1st millennium AD *that are discussed in the main text. Changbo is the most southerly of a group of kiln sites producing stonewares during the 10th century* AD *in Liaoning province (see data of Figures 29.3a-d). Graphic: B Scaife and P. Zimmerman, MASCA.*

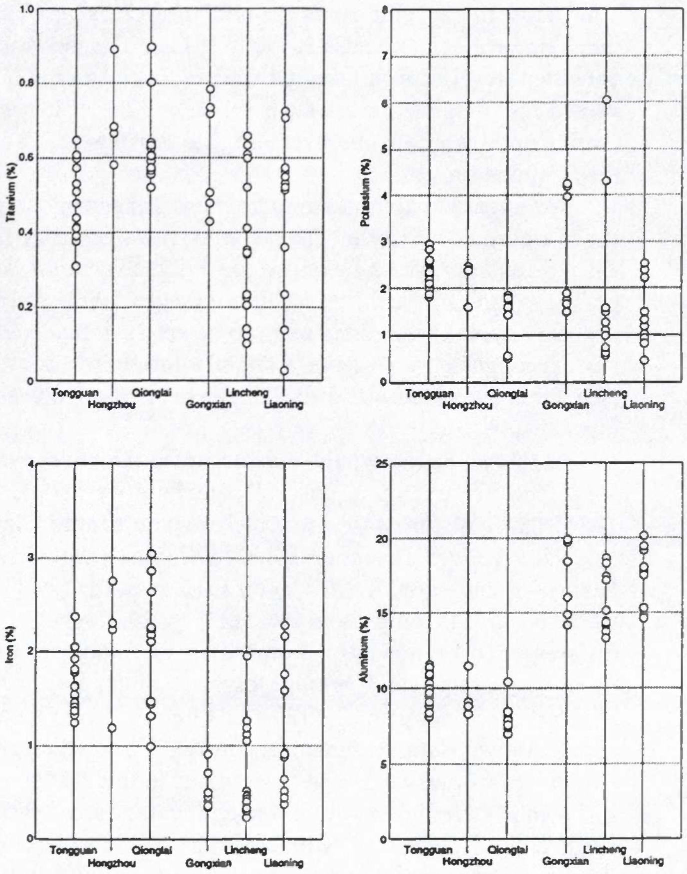

Figure 29.3: Summary of the primary elements in wares from various early Chinese kiln sites. Graphic: B. Scaife, MASCA.

	Elemental content (%, by wt.)						Oxide content (%, by wt.)						
Inv. no.	Al	Ti	Fe	K	Ca	Mg	SiO_2	Al_2O_3	TiO_2	Fe_2O_3	K_2O	CaO	MgO
Underglaze-painted dishes													
Sr 0005	11.3	0.48	1.78	2.48	0.19	0.70	70.9	21.2	0.80	2.55	2.98	0.27	1.16
Sr 1160	8.9	0.44	1.34	2.44	0.60	0.30	76.3	16.7	0.73	1.92	2.93	0.84	0.50
Sr 1214	9.5	0.42	1.64	1.84	0.08	0.39	76.0	17.9	0.69	2.35	2.21	0.11	0.65
Sr 1215	9.7	0.42	1.92	2.60	0.17	0.27	74.3	18.3	0.70	2.75	3.12	0.23	0.45
Sr 1222	11.6	0.60	2.38	2.06	0.56	0.23	75.2	16.9	0.73	3.40	2.47	0.78	0.38
Sr 1229	8.2	0.41	1.54	2.98	0.26	0.37	77.1	15.3	0.69	2.20	3.58	0.36	0.61
Sr 1231	8.9	0.31	1.25	2.62	1.04	0.40	75.5	16.7	0.52	1.79	3.14	1.46	0.66
Sr 1233	8.5	0.39	1.48	2.86	0.47	0.35	76.5	15.9	0.65	2.12	3.43	0.66	0.58
Straw-glazed jars													
Sr 1223	10.8	0.51	1.52	2.19	0.25	0.46	72.8	20.3	0.85	2.17	2.63	0.35	0.76
Sr 1230	9.8	0.38	1.78	2.30	0.47	0.17	74.5	18.5	0.63	2.55	2.76	0.65	0.28
mean n=10	9.7	0.44	1.66	2.44	0.41	0.38	75.0	17.8	0.70	2.38	2.92	0.57	0.60
Std. devi.	1.2	0.08	0.33	0.35	0.28	0.15	1.9	1.9	0.09	0.47	0.42	0.40	0.24

Table 29.1. The body composition of a group of Tongguan dishes and jars excavated from early 9th century contexts at the Islamic site of Siraf. Since the calcium and magnesium contents are relatively low, there is little likelihood of carbonates in these wares, so the silica content reported here has been calculated by mass balance. In all instances the Na_2O content was less than 0.17%. Each inventory number quoted here is an abbreviation, the prefix Sr replacing that used by the Royal Ontario Museum (Toronto)— #986.407., for Siraf.

	Tongguan u-g. ptd.(n=10)		Tongguan various (n=12)		Chengguan various (n=19)	
Al	9.7	± 1.20	10.30	± 0.80	9.30	± 0.80
Ti	0.44	± 0.08	0.59	± 0.04	0.44	± 0.04
Fe	1.66	± 0.32	1.63	± 0.26	1.27	± 0.21
K	2.44	± 0.35	2.18	± 0.20	2.35	± 0.32

Table 29.2. Comparison of the body compositions for wares from kiln sites close to Changsha. Mean content (%, by wt.) based on compositional data given in Table 29.1, and the work of Chen Xianqiu et al. *in Li Jiazhi and Chen Xianqiu (1989), pp.309-316; and Chen Shiping* et al. *in Li Jiazhi and Chen Xianqiu (1992), pp.157-167.*
Uncertainty in each mean value is quoted as one standard deviation.

the Tang capital of Xian over the period of the 7th to 9th centuries.

A similar situation seems to hold true for iron. Though the iron content of wares from south central sites is, *on average*, about twice that of north central and northern wares, the *scatter* of data for these sites allows for appreciable overlap (Fig. 29.3c). Even at Lincheng, the white-bodied Xing wares with an iron content of about 0.4% occur with some darker bodied wares with an iron content of about 1.1% or more. It is only in aluminium contents (Fig. 29.3d) that a broad regional differentiation becomes convincing.

Data obtained for two ware types—celadon and black glazed—produced at the kiln site of Xinhui, just south of Guangzhou (Guandong province), should also be noted here (Li Jiazhi & Chen Xianqiu 1992: A-19). The titanium content of the black wares exhibits bimodalism (Fig. 29.4, left); and the two ware types have significantly different chemical "fingerprints" in terms of their iron, potassium and aluminium contents (see Fig. 29.4, right and caption). Both ware types have aluminium contents—typically, about 15.5%—that fall midway between what is usual for south central and north central China (cf. Fig. 29.3d).

So we are left with the situation that: (i) *no* major element differentiates between some of the most active pottery producing areas on or north of the Yellow River; and (ii) *no* major element differentiates between at least three of the primary pottery producing areas along the Yangzi River. Such matters are important in the general interpretation of Chinese ceramics for two reasons. First, it is known that underglaze-painted wares were produced at other sites along the Yangzi River (Fig. 29.2). In this respect, Qionglai is of particular interest, since excavations of its kilns have yielded underglaze-painted wares which, though aesthetically said to be somewhat cruder in finish, are stylistically quite close to those from Tongguan (Li Jiazhi & Chen Xianqiu 1989). This is also true for Hongzhou, where kiln excavations have yielded wares with a transparent greenish-yellow glaze which is very similar to the one usually used at Tongguan. The distinctive technology of a Tongguan stoneware's glaze—described nowadays as a *phase-separated, glass emulsion*—is shared by a number of Tang Dynasty production sites (Woods 1992). Once wares of this kind became "blended" together in depots at Hangzhou and other ports at the mouth of the Yangzi River, prior to their shipment westward, stylistic differences would become blurred and ambiguous.

Second, the kilns at Tongguan, and indeed those at all the other production centres along the Yangzi River's length, produced many other wares that are far less distinctive than the underglaze-painted wares discussed here (See-Yiu Lam 1991). Many of these also were exported to the West, and now turn up in the excavations of the same Islamic contexts that yielded the underglaze-painted wares (Whitehouse 1972; Rawson et al. 1987). Such wares were also the subject of Islamic mimicry (Mason & Keall 1990). A typical example is the green-and-white glazed Bowl illustrated in Fig. 29.5 here, that was also excavated at Siraf. It is definitely Chinese in origin (Fleming & Swann 1992), but stylistic parallels can be found not only at Tongguan, but also

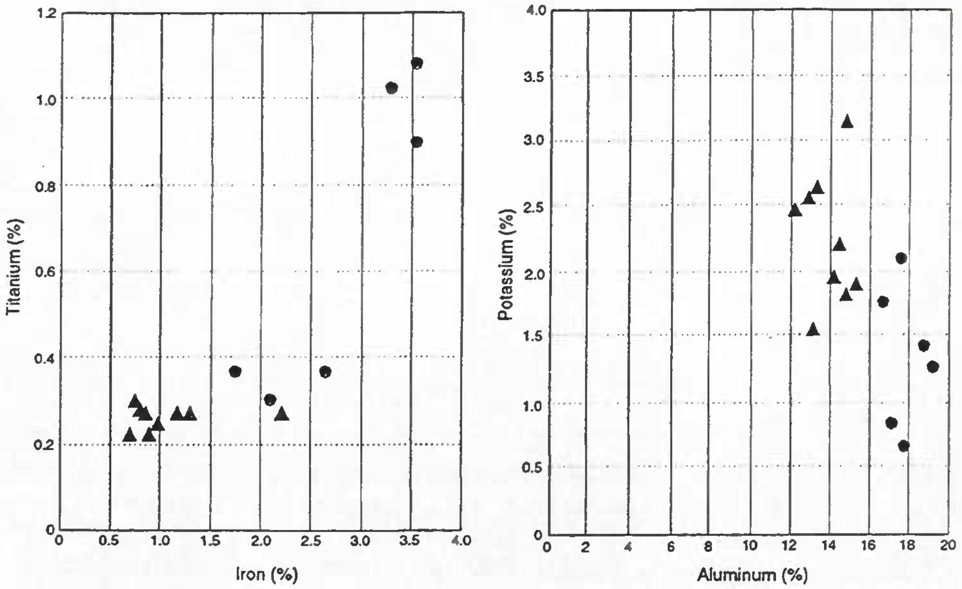

Figure 29.4: Summary of the primary elements in celadon and black-glazed wares (solid triangles and circles, respectively) from the kiln site of Xinhui (Guandong province). Mean values for these two ware types are as follows: Celadon: Fe% 1.08 ± 0.47; K% 2.21 ± 0.49; Al% 13.9 ± 1.1. Black glazed: Fe% 2.82 ± 0.78; K% 1.33 ± 0.54; Al% 17.9 ± 0.9. Graphic: P. Zimmerman, MASCA.

among tomb goods in the luxurious burials near Xian (Caroselli 1987); and in southern China, at various sites in Guangdong province (Lam 1986). The most recent part of our research program has therefore focused on the question of whether *trace* element analysis offered at least a means of differentiating the products of various kiln sites located in the Yangzi River basin.

29.3. Trace element analysis

The data that we have obtained for the Tongguan stonewares from Siraf are given in Table 29.3, ordered according to the degree of scatter those data displayed. Note that the occurrences are given in ppm, as appropriate for trace elements — not in the percentages which have been given previously for the major elements. The rare earth element, lanthanum, correlates significantly to the body's aluminium content (with an R-value of +0.896). Since we already know that aluminium will not differentiate kiln sites along the Yangzi river, we cannot expect lanthanum to do so either. The same cannot be said of hafnium; and, to a lesser extent, of thorium, caesium, and rubidium. (Scandium's behaviour in this study is unusual in that is shows no significant correlation to iron [R-value of +0.121]—cf., the usually very high linkage of these elements in pottery from the Near East, as reported by Rothman and Blackman [1992], among others.) The low correlation to aluminium content that these elements display suggests that they are carried by mineral inclusions *not* immediately related to the main kaolinitic clay constituent of the stoneware. This idea is consistent with a recent petrographic study which showed that the bodies of these Tongguan wares are heavily tempered not only with quartz sand, but also with significant amounts of volcanic glass and associated feldspar (see Li Jiazhi & Chen Xiangqiu 1992: A-23; Mason & Keall 1991). We would anticipate that there is much variability in the inclusion geology over the great length of the Yangzi river, and so expect characteristic patterning of these and other trace elements in the vicinity of each early kiln site.

We would like to have studied some excavated Qionglai and Hongzhou wares in this way, but we have not yet been able to obtain any representative samples from those sites. We have been able to take our studies one step further, however, because of the recent publication of INAA data for so-called Guan ware from some 12th century AD kilns at Jiaofangxia (Li Huhou 1988). While most of the trace elements occur at levels very similar to those we have found in Tongguan wares, the Guan wares have caesium and rubidium contents that are quite different (Table 29.4, Fig. 29.6). It is of some interest that the INAA trace element pattern for the green-and-white dish discussed in the last section (see Fig. 29.5), does not match that of either Tongguan or Jiaofangxia. A lower rubidium content (*c.* 89.3ppm) and a much higher cobalt content (*c.* 21.3ppm) sets the dish well apart, leaving its provenance of production still in doubt.

29.4. Conclusions

Neutron activation analysis (INAA) offers an effective way of characterising the constituents that will have influenced the behaviour of the body of Chinese ceramics during kiln firing. At the same time, this analytical technique provides much data about the trace element components of both the main clay stock of the body and of the coarse-grained minerals which are often introduced to improve that clay's working properties and robustness. Such trace elements are likely to cluster into concentration patterns which are diagnostic of individual kiln sites. That point has been demonstrated here for underglaze-painted wares from Tongguan, and we have little doubt that such studies would be equally effective for many other ware types of importance in the study of early trading relationships between China and the Occident.

Acknowledgements

We wish to thank Rob Mason of the Royal Ontario Museum (Toronto) for his background collaboration in this project.

Element	Mean (ppm)	Std. devn.	Scatter	R-value (*v.* Al)
Al	9.70	± 0.30	3.29%	1.000
Hf	9.36	± 0.65	6.94%	− 0.198
La	59.90	± 5.00	8.35%	+ 0.896
Lu	0.79	± 0.08	10.0%	+ 0.280
Yb	5.04	± 0.53	10.5%	+ 0.266
Th	26.30	± 2.80	10.7%	+ 0.401
Sm	10.10	± 1.10	11.2%	+ 0.889
Ce	120.00	± 13.0	12.2%	+ 0.664
Sc	15.60	± 1.90	12.2%	+ 0.595
Ta	2.62	± 0.33	12.7%	+ 0.434
Cr	102.00	± 15.0	14.4%	+ 0.601
Rb	199.00	± 29.0	14.7%	+ 0.375
V	91.00	± 14.0	15.6%	+ 0.581
U	6.80	± 1.10	15.8%	+ 0.094
Na	0.11	± 0.02	16.1%	− 0.284
Cs	33.30	± 5.40	16.2%	+ 0.303
Eu	1.90	± 0.30	17.5%	+ 0.771
Ti	0.44	± 0.08	18.2%	+ 0.593
Fe	1.66	± 0.33	19.9%	+ 0.192
Sb	2.25	± 0.56	24.9%	+ 0.521
Co	5.10	± 1.50	28.4%	− 0.228
As	5.40	± 1.90	34.3%	+ 0.290
Mn	109.00	± 39.0	35.8%	− 0.332
Mg	0.36	± 0.15	40.1%	+ 0.560
Sr	127.00	± 76.0	59.3%	− 0.096
Ca	0.41	± 0.28	68.3%	− 0.419

Table 29.3: Summary of major, minor, and trace element data for Tongguan stoneware (n=10). ***NB****. Ni was also sought but not found above its INAA detection limit of about 50 ppm.*

	Jiaofanxia kilns		green/white glazed dish
Element	Mean (ppm)	Std. devn.	Content (ppm)
Hf	9.1	± 1.10	9.8
La	not measured		61.3
Th	23.5	± 1.00	23.4
Sc	16.7	± 2.20	17.2
Ta	1.82	± 0.17	2.51
Cr	73	± 12.00	79.0
Rb	141	± 19.00	89.0
V	not measured		110.0
Cs	9.9	± 1.60	6.5
Sb	1.23	± 0.75	2.12
Co	4.2	± 1.40	21.3
Mn	not measured		112.0
Sr	not measured		283.0

Table 29.4: Summary of trace element data for Guan wares from the kilns of Jiaofangxia. Ni was also sought but not found above its INAA detection limit of about 50 ppm. The green/white dish is Royal Ontario Museum, inv. #986.407.1205, see Figure 29.5.

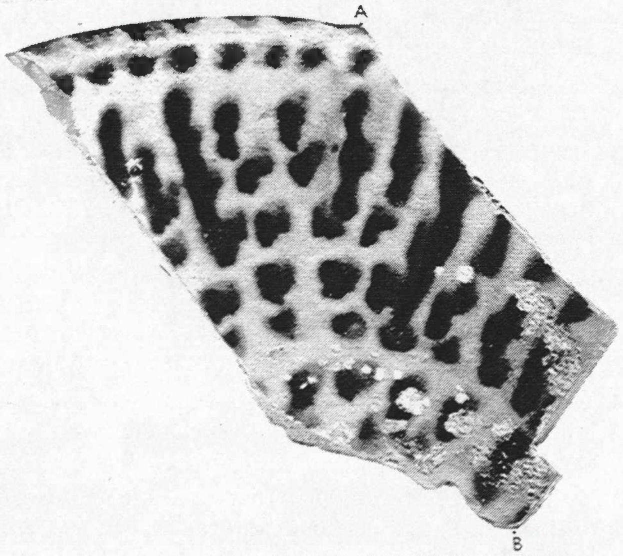

Figure 29.5: Sherd (section AB, 11.5 cm) for a Chinese green-and-white glazed dish excavated at the Islamic port site of Siraf. Royal Ontario Museum, Toronto: inv. #986.407.1205.

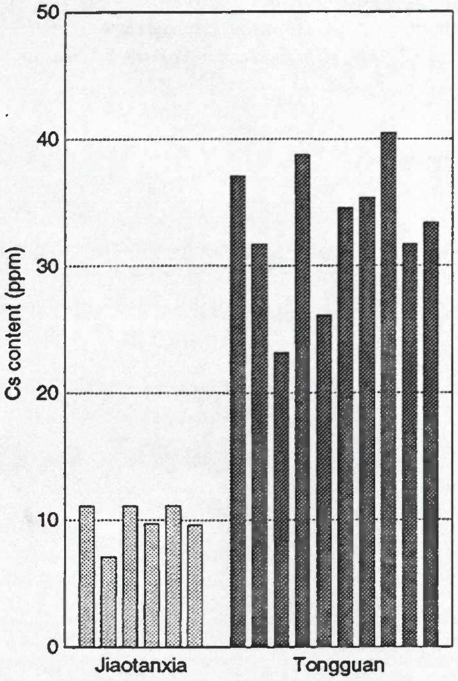

Figure 29.6: Comparison of caesium contents for the kiln sites of Jiafangxia (Zhejiang province) and Tongguan (Hunan province). Graphic: B Scaife, MASCA.

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