# The Introduction of Celadon Production in North China: Technological Characteristics and Diversity of the Earliest Wares

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### **Abstract**

Celadon, technically a stoneware with a lime-rich glaze, had been produced in South China for more than two millennia before it was first made in the North in the second half of the sixth century. It appears to have been an immediate precursor to white porcelain, which was first produced by northern kilns. The compositions and microstructures of early northern celadons from kilns, residential sites and tombs in Shandong, Hebei and Henan provinces, and dated 550s-618 CE, have been determined by SEM-EDS. The majority of the vessels were made using a low-iron kaolinitic clay, with high alumina (20-29%), as anticipated for northern clays. A small number of celadon vessels from a kiln at Caocun, which produced mainly lead-glazed wares, have lower alumina contents and appear to have originated in the South. It seems possible that these imported vessels were being used by the potters as models on which Caocun wares were based. Consistent differences in major element composition are observed between the products of kilns at Anyang, Xing, Luoyang and Zhaili. Unlike southern celadon glazes, which were prepared as two-component mixtures of vegetal ash and body clay, the northern celadon glazes are three-component, and typically contained an additional siliceous component, probably loess. An exception is the glazes of the Xing celadons, which present no evidence for loess but which are rich in Na<sub>2</sub>O. The source of the soda is unclear, common salt and albitic feldspar are discussed as possibilities. Based upon micromorphological characteristics such as the relative size and abundance of remnant quartz and the extent of observable mullite, as well as the position of the glazes in the CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> phase diagram, the Xing bodies are more mature and they appear to have been fired to higher temperatures than the products of other kilns. These results suggest that celadon technology was not directly transferred to the North from the South, but that the northern potters adopted their own strategies to make high-fired glazes. Furthermore, each kiln appears to have had its own preferred recipe, to suit the available raw materials. The products of Xing kiln were exceptional and it appears that here the trajectory towards white porcelain was already apparent, perhaps reflecting the creativity of the Xing potters who were among the first to make a successful white porcelain.

Keywords: Celadon, North China, Technology, Characteristic, Diversity

## **1. Introduction**

The sixth-seventh centuries CE were a period of important developments in the evolution of ceramic production in North China. After more than two thousand years of production in kilns such as Yue in the South, the manufacture of celadon was introduced to the North for the first time in the second half of the sixth century. Furthermore, archaeological evidence indicates that white porcelain was produced soon afterward, at least from the last decades of the sixth century<sup>1</sup>. In this paper we explore the material characteristics of the northern celadons from the earliest confirmed kilns and other archaeological sites, the nature of the technological transfer from the South and the position of northern celadon on the trajectory towards white wares.

Celadon is, in the strict sense, a stoneware with a bluish-green or grey-green glaze fluxed with a lime-rich material. The origin of this term is European, and describes the colour of the glaze, regardless of its raw material or manufacture technology (Gompertz1980). In Chinese, it is called Qing Ci(青瓷), which is why "greenware" has also been used to refer to this product (for example: Pollard and Hatcher 1986), as literally "Qing" means green and "Ci" means porcelain<sup>2</sup>. However, in modern ceramic practice the term greenware is also used to refer to the unfired body of a ceramic while, in ancient Chinese, "Qing" can refer to a variety of different colours including green, blue and even black. It is a fact that some of the most admired celadons, for instance, examples of Guan Ware(官窑), Ru Ware(汝窑) and Jun Ware(钧窑), if described precisely, are blue or grey rather than green. For these reasons, despite its ambiguities, celadon has been accepted in a common context as the translation of Qing Ci.

The origin of celadon may be traced back to the 17<sup>th</sup> century BCE, when the so-called protoporcelain was first made in the Bronze Age lower Yangtze River Delta (Yin *et al.* 2011). Although it is suggested that the first ceramics recognised as celadon, in that they are characterised by a smooth glaze and a well-vitrified body, were developed in the Yue kiln in Zhejiang province in the late Eastern Han dynasty around 200 CE (Li 1978), the difference between the "proto" and "mature" celadons is quite blurred, as they were made using essentially the same materials and recipes. By the fifth century in the Southern Dynasties (420-589CE), celadon had been widely produced across South China. The main production centres are shown as circles in Figure 1, where it should be noted that each centre typically covers several kilometres and includes a number of individual kiln sites.

Made first in South China, celadon production did not emerge until more than two thousand years later in the North. Although celadon wares were found in northern sites as early as the Bronze Age, their origin has been always under debate (An 1960, Zhou *et al.* 1961, Chen 1997 & 2016, Zhu *et al.* 2004, Xia *et al.* 2009). Until at least the second half of the sixth century, the celadons uncovered in the North were closely similar to southern burial objects and kiln products (Yabe 1981, Hasebe 1982, Hsieh 1994, Liu 2015). Furthermore, no contemporaneous kiln site with similar products could be confirmed in the North. Scholars therefore generally agree that the celadon wares found in the North preceding the second half

<sup>&</sup>lt;sup>1</sup> As exemplified by vessels recovered from Sui Dynasty tombs such as the tomb of Lv Wu in Xi'an of Shaanxi Province (吕武墓, 592 CE, IoA 1966) and tomb of Zhang Sheng in Anyang of Henan province (张盛墓, 595 CE, IoA 1959)

<sup>&</sup>lt;sup>2</sup> The definition of porcelain in Chinese is much more flexible than in the Western context, thereafter, "ware" is an appropriate word to translate "Ci" under this circumstance.

of the sixth century are likely to have been made in and exported from the South (Hsieh 1994, Guo and Zhang 1997, Liu and Yuan 1999, Liu 2015). Recent archaeological work has unearthed celadons displaying clear "northern attributes" from burials mainly dating from the last quarter of the sixth century, across the North China Plain in Shandong, Hebei and Henan provinces (for details see the description of sites below). Furthermore, a number of contemporaneous kiln sites yielding sherds of similar celadon wares were also uncovered within this area (Fig.1). Given the assumption that the formation of a distinctive style might take some time, we therefore are inclined to suggest that the production of celadon in North China may have started by the second half of the sixth century CE and became active from around the 570s. In contrast to southern celadon, its abrupt emergence and limited manufacturing scale has meant that little has been known regarding early northern celadon until recently.

Below we explore the introduction of celadon production in the North through the analysis of material from the earliest manufacturing centres so far published. We have analysed bodies and glazes of 35 sherds from kiln sites, residential sites and tombs in Shandong, Hebei and Henan provinces in North China, dating from the late Northern Dynasties to the Sui Dynasty (approximately 550s-618 CE). These contribute an overview of northern celadon technology in its early stages, highlight the changes that occurred in the transfer of the technology from the South and point to the special features of the Xing kiln, which is known to have produced some of the earliest translucent white porcelain.

## 2. Materials and Methods

## 2.1 Sites sampled for this study

From the last quarter of the 6<sup>th</sup> century CE, celadon with northern features started to appear in Northern Qi (550-577 CE) and Sui Dynasty (581-618 CE) burials in Shandong, Hebei and Henan provinces. This included the half glazed coarse bowls with black "tear-like" glaze drops, the rope-shaped looped jars, and the high stem plate which had not been seen in the South. Correspondingly, celadon kiln sites yielding similar sherds were discovered at Zibo (淄博), Zaozhuang (枣庄), Xingtai (邢台), Cixian (磁县), Linzhang (临漳), Handan (邯郸市 区), Anyang (安阳) and Luoyang (洛阳) in the same three provinces. The samples analysed here were collected from five archaeological sites and are summarised below and in Table 1. Locations are shown in Figure 1.

### 2.1.1. Zhaili kiln site in Zibo, Shandong Province (山东淄博寨里窑)

Located in a hilly region in central Shandong province, this kiln site is unknown from any historical record. A trial excavation of this site (Fig. 1, site 1) was undertaken in the 1970s, discovering large numbers of sherds of celadon and yellow glazed ceramics. As some of the vessels are identical to burial objects from local late Northern Dynasties burials, for instance, bowls similar to the ones from the tombs of Dao Gui (道贵, 571 CE, Jinan Museum 1985) and Cui Bo(崔博, 573 CE, Shandong IoA 1984), four-looped jar similar to the objects from the tombs of Cui Fen (崔芬, 551CE, Shandong IoA et al. 2002), this kiln site is dated no later than the Northern Qi dynasty (550-577CE, ZCHWR 1984:352-359) and continued to the Sui

and Tang Dynasties. Eight pieces of early celadon were collected from Zhaili kiln site for analysis.

## 2.1.2 Xing kiln site in Xingtai, Hebei Province(河北邢合邢窑)

The Xing kilns are well known for the production of the white porcelain, Xing Ware, at least as early as in the eighth century CE. As Li Zhao (李肇 713-805CE) wrote in Guo Shi Bu (国 史补, A Supplement to the History of the Country) : "White porcelain bowls from Neiqiu and purple ink stone from Duanxi are popular throughout the country and commonly used by people of all the classes." ("内丘白瓷瓯,端溪紫石砚,天下无贵贱通用之。") Neiqiu is one of the production centres of the Xing kilns which are mainly located in Xingtai City (邢 合市), Hebei province (Fig.1, site 2). Archaeological survey and trial excavation from 1980s revealed that celadon was also produced in the Xing kilns, likely started from late Northern Dynasties or Sui Dynasty (approximately 570s to 618 CE; Yang and Zhi 2011). The products are exemplified by the half glazed deep bowl unearthed from Xijiangu kiln site(西 坚固窑) in Xingtai which is in close similarity with the bowls discovered from the tombs of Gao Run (高润, 576 CE, Cixian Cultural Centre 1979) Gao Tan (高潭, 582 CE, Hebei Culture Relics Management Office 1979) in Hebei. Eight celadon samples were collected from the Neiqiu site for analysis.

## 2.1.3 Caocun kiln site in Linzhang, Hebei Province(河北临漳曹村窑)

Caocun kiln site was discovered in 2009 and the excavation has been undertaken since 2010 (see Fig.1 kiln site 3). Located within Yecheng (邺城, City of Ye) which was the capital city of the latter half of the Northern Dynasties from 534 to 577 AD (CASS IoA et al. 2014), its products are identical in appearance to the burial objects from the Northern Qi noble tombs in Anyang(安阳) and Cixian(磁县), such as the Wanzhang tomb (湾漳大墓, believed to be the mausoleum of Gao Yang (高洋), the first emperor of the Northern Qi Dynasty, 559 CE (IoA et al. 2003), the tomb of Yuan Hu (元祜墓, by 537 CE, Zhu et al. 2008) and etc., which have been confirmed as lead-glazed ceramic with pXRF by the authors. Furthermore, the city of Ye was burnt down during warfare in 580 CE, which has contributed to its chronological significance by providing a *terminus ante quem*. Glazed ceramic sherds unearthed from this site are mostly lead-glazed (nearly 3000 sherds, making up 87% of the total excavation), together with a small amount of 85 pieces of celadon (pXRF result supplied by the Yecheng Archaeology Team), four of which were selected for analysis.

## 2.1.4 Xiangzhou kiln site in Anyang, Henan Province(河南安阳相州窑)

Discovered in 1970s, the Anyang Xiangzhou kiln site (subsequently referred to by the more familiar name Anyang in this paper) has been excavated three times since 2006 (Kong 2014). Anyang replaced Yecheng as "New Ye City" after the latter had been burnt down in 580 CE, functioning as a regional capital from the Sui Dynasty (see Fig. 1, kiln site 4). Celadon is the dominant product of this kiln, and is similar to the local Sui Dynasty burialobjects such as the celadon wares from the tombs of Song Xun (宋循, 584 CE, ABCE 1973), Han Yong (韩邕, 587 CE, Anyang Museum 1986) and Bu Ren(卜仁, 603 CE, Song 1958), therefore mostly dated to Sui dynasty. Seven sherds were collected, plus another two taken from local Sui Dynasty burials.

## 2.1.5 Han-Wei city site in Luoyang, Henan Province(河南汉魏洛阳城遗址)

The Han-Wei city site is the ruin of the capital of the Northern Wei Dynasty from 495 to 534 CE, which was then continuously occupied till at least the Sui Dynasty (581-618 CE, see Fig.1, site 5). Due to the long-term occupation, a wide variety of ceramics have been unearthed, including almost all types of lead-glazed ceramic, celadon and white porcelain, among which are some of the finest early celadons dated to the late sixth and early seventh centuries. The celadon was probably supplied by nearby kilns in Gongyi (Henan IoA et al. 2009 & 2011, see Figure1 the site marked as hollowed star to the east of site 5), and it has been suggested that celadon production at the Gongyi kiln sites could hardly be dated before the Sui Dynasty (Mori 2009,Kobayashi 2009 ). Six celadons unearthed from the Han-Wei Luoyang city site were analysed in this study.

### 2.1.6 Putative Northern Dynasties kilns not included

There are several more sites which have been claimed to be Northern Dynasties kiln sites in Shandong, Hebei and Henan provinces shown as hollowed stars in Figure 1. In summer 2016 and 2017, field surveys were undertaken by the authors of this paper to these sites, discovering they were either so poorly preserved that very little evidence could be collected, or that the typology of the ceramics was at best ambiguous with respect to a Northern Dynasties date. Therefore, although it remains possible that some of these sites produced early celadon the evidence is currently too limited for inclusion here. Nevertheless, their locations in Figure 1 are still valuable to illustrate the spatial distribution of ceramic production in the North China Plain by the end of the sixth century.

# 2.2 Sample Description

Although collected from different archaeological sites, the celadon samples share some common features. Their glaze is mainly transparent, yellowish grey, with various degrees of brown/black speckles and fine cracks. Their differences are in detailed properties, such as the tone of the colour, the reflectivity of the surface, or the thickness of the glaze. All the bodies are grey or pale grey and dense in texture, mostly with visible pores and/or black speckles (see Figure 2 and Table 2).

# 2.3 Analysis

The Munsell Rock Color Book was applied to record the colour of the samples. In addition, sherds were photographed in RAW format with white balance set using an X-RITE Mini Color-Checker neutral grey balance card before shooting. ISO was between 50 to 200, and exposure confirmed using the X-RITE greyscale target.

Ceramic samples were cut and embedded in epoxy resin blocks, which were ground and polished to 1 µm with diamond paste. Carbon-coated polished cross sections were examined in a Philips XL 30 ESEM at 20 kV with 10 mm working distance using back-scattered electrons. Compositions were determined using an Oxford Instruments INCA energy dispersive X-ray spectrometer. The composition of each sample represents the average of at least three different areas. The data were produced as oxide percentages and normalised to 100%, due to low totals which reflected the porosities of the bodies. The analysis of Columbia River Basalt (BCR-2) reference standard is given in in-line table A1 and indicates relative accuracies of around 5% or better for the elements of interest except for MnO and

 $TiO_2$  of about 10%. Detection limits were taken as three standard deviations on the background counts.

## 3. Results

#### 3.1 Microstructure

All samples show a similar microstructure in the SEM. A typical example, Figure 3a, shows a glaze layer, around 300  $\mu$ m thick, overlying a body with continuous vitrification and fine pores. At the glaze-body interface fine crystals of calcium-rich plagioclase (typically anorthite CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>) may be developed (Fig. 3b). Rounded grains of quartz, frequently showing cracks due to differential expansion, are common (Fig.3c). The rounded nature of the grains indicates that they are not crushed, and the fine size suggests they are intrinsic to the clay and were not added. Bright particles of TiO<sub>2</sub>, probably derived from rutile or anatase in the clay, may be seen. Iron oxides are typically completely absorbed into the matrix, although sometimes diffuse areas rich in iron may be observed. In back-scattered images fibrous crystals of mullite could be observed in the matrix of many of the samples (Fig.3d). The generally fine grain sizes of the bodies, their homogeneity, the absence of needle-like secondary mullite and their relatively constant compositions do not suggest the addition of materials in addition to kaolinitic clay, such as feldspar or dolomite, which have sometimes been suggested as additives to other Chinese stoneware bodies.

Different degrees of vitrification were observed as shown for example in Figs. 3c & 3d, which compare two samples collected from the Anyang kiln viewed at the same magnification. Low-fired samples have large numbers of relatively small, irregular pores, which are inherited from the original fabric of the clay, and relatively abundant sub-rounded quartz (Fig. 3c). Higher-fired samples have fewer, large rounded pores shaped by the pressure from trapped gases which are unable to escape from the highly vitrified matrix. The higher fired samples have fewer, finer quartz grains and fewer visible TiO<sub>2</sub> particles. Mullite crystals, just resolvable in the matrix, are disordered and have relatively low aspect ratios, indicating that they were primary mullite generated from the clay rather than secondary mullite from feldspar inclusions (Iqbal and Lee 1999, 2000).

The micromorphological observations for all samples are summarised in Table 2. An empirical scale of vitrification stages was developed based upon the number, shape and size of pores, the size and abundance of quartz and mullite formed in the body, and the development of crystals at the interface between body and glaze. Vitrification stage 1 is least vitrified, stage 2 refers to continuous vitrification with large numbers of irregular pores, stage 3 is continuous vitrification with both irregular pores and some rounded pores, stage 4 is continuous vitrification with fine spherical pores, stage 5 is continuous vitrification with medium spherical pores, and stage 6 is continuous vitrification with coarse spherical pores. The size of the relict quartz is described as coarse, medium and fine and its abundance was visually estimated with petrographic comparator charts (see Quinn 2013:82). The mullite in the body is described as absent, present, or abundant; similarly, the crystals at the glaze-body interface are described as none, sparse or extensive.

As seen from Table 2, there are few systematic differences between the ware groups, with most samples showing vitrification stages 3 (18 out of 35) or 2 (10 out of 35). However, the Xing sherds analysed showed a relatively high vitrification stage at 3-4, and include almost all of the stage 4 samples observed. It is also observed that relict quartz grains are more abundant in wares from Zhaili, Anyang and Caocun Group 1, less abundant in the Luoyang samples and least abundant in the Xing wares. Mullite was observed least frequently in the Zhaili wares and most frequently in those from the Xing kiln but it should be borne in mind that the numbers of samples in each group are small and that the atomic number contrast between mullite and the vitreous matrix is low in the SEM.

#### 3.2 Body compositions

The bulk compositions of the bodies were measured by averaging 3 arbitrarily selected areas at low magnifications ( $\times$ 50), results are presented in Table 3, and the relationship between major components silica and alumina shown in Figure 4. Generally, the total concentration of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> is around 95%, irrespective of source, while the balance is composed of the oxides of K, Fe, Ti, Ca, Mg and Na. MgO and Na<sub>2</sub>O do not exceed 0.5%. There is some clustering of sites in Fig. 4. Two samples from Caocun kiln site plot as outliers, with particularly low Al<sub>2</sub>O<sub>3</sub> (<17%) and high in SiO<sub>2</sub> (>76%) and are designated as Caocun Group 1, while the remaining two, with high alumina, form Caocun Group 2 (Table 3). All samples other than Caocun Group 1 are relatively rich in Al<sub>2</sub>O<sub>3</sub> (>20%), and consequently the SiO<sub>2</sub> is typically below 74%. It is also observed from Fig. 4 that Caocun Group 2 and the samples from Xing and Zhaili kiln sites are distributed evenly with intermediate Al<sub>2</sub>O<sub>3</sub> contents (20%-24%). The Luoyang celadons have a range of Al<sub>2</sub>O<sub>3</sub> contents but these are high (>26%).

Differences between the production sites may also be observed in other components, for example Xing celadons tend to have high MgO and low  $TiO_2$  relative to other sites, Anyang has high K<sub>2</sub>O, Luoyang has low FeO and so on (Table 3). While our sample size and analytical method were not selected to form the basis of a robust provenance study, it appears that compositional analysis involving major elements, perhaps with a few selected traces, is likely to offer a way forward in sourcing northern celadons.

#### 3.3 Glaze compositions

To the eye, the glazes are typically yellowish and transparent and the body can be seen through the glaze (Fig. 2). Away from the glaze-body interface, few crystalline inclusions or inhomogeneities are observed in the SEM (Figs 3a & b), although a layer of bubbles occurs at the base of the glaze. In spite of the 1500 years since deposition, no corrosion layer is observed, even in backscattered mode in the SEM (Fig. 3a).

The chemical composition of each glaze (Table 4) was measured by averaging 3 arbitrarily selected areas in the centre, away from the body, at low magnifications. All glazes are fluxed with high lime at 11-22%, and the concentrations of the major elements of most samples are similar, with a total of 70-80 wt. % Al<sub>2</sub>O<sub>3</sub> plus SiO<sub>2</sub> and most samples containing between 12-17% Al<sub>2</sub>O<sub>3</sub> and 55-63% SiO<sub>2</sub>. Xing celadon glaze is relatively distinctive, with high Al<sub>2</sub>O<sub>3</sub> (c. 17%) and low SiO<sub>2</sub> (c. 56%), while Caocun Group 1 has the lowest Al<sub>2</sub>O<sub>3</sub> in the glaze (12%). These two groups are also distinctive in other aspects of their composition; Xing celadon glaze has notably higher Na<sub>2</sub>O than all other wares, averaging 3% as opposed to around 0.5%, while Caocun Group 1 has notably higher MgO, P<sub>2</sub>O<sub>5</sub> and MnO than other

types, and is especially low in Na<sub>2</sub>O (less than 0.2%, below the detection limit) (Table 4). As might be expected, CaO is negatively correlated with the concentrations of  $Al_2O_3$  and  $SiO_2$ . FeO is present at around 1.5% in most of the wares analysed, although the Zhaili glazes have an average of 3% FeO with a relatively wide dispersion.

#### **4.Discussion**

#### 4.1 Regional affiliations

As is well established, the raw materials for porcelain and stoneware production in northern and southern China were very different (Sundius and Steger 1963; Pollard and Hatcher 1986, 1994, Guo 1987). This is due to the difference in geologies on either side of the Qinling-Dabieshan Belt and the Tanlu wrench fault system (see Fig. 1, Wood 2000). In the South, clays formed from altered igneous rocks were used, and southern bodies therefore typically have relatively low alumina/silica ratios. In the North, on the other hand, the bodies were prepared from the sedimentary kaolins associated with coal deposits. Northern bodies normally have correspondingly higher Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>. The present results are generally consistent with this pattern. The bodies of all of the samples analysed from those kilns known to have been firing high temperature celadons are of the high alumina type.

The exceptions come from the Caocun kiln site, where the pXRF survey indicated that the waste ceramics were overwhelmingly relatively low-fired lead-glazed ceramics which appear to have been emulating celadon. The two Caocun Group 1 celadon bodies are clearly of southern affinity, with low Al<sub>2</sub>O<sub>3</sub> contents resembling southern celadon types such as Yue ware, which is shown in Fig.4 for comparative purposes. Given that the majority of the ware produced at Caocun was lead-glazed, and the bodies of the local products are of the high alumina type, it appears that these sherds represent imported southern celadon. Glaze compositions are consistent with the interpretation of Caocun Group 1 as imported southern wares. They differ in composition from the other celadon glazes analysed in many respects, most notably in their Na<sub>2</sub>O, MgO, P<sub>2</sub>O<sub>5</sub> and MnO contents. It is probably diagnostic that the Caocun Group 1 glazes have much higher MnO than any of the northern celadon glazes analysed here, at around 0.7% as opposed to below the detection limit of 0.21% in all other wares. High MnO contents appear to be more typical of southern celadon glazes and this may well offer a parameter which would allow rapid and non-destructive identification of celadon vessels from the two regions, using pXRF.

The presence of southern celadon sherds on a northern kiln site which produced lead-glazed wares is intriguing. The dominant lead-glazed products of Caocun closely resemble true, high-fired celadon in appearance and we have speculated that these two sherds may represent models – southern celadons used by the northern potters so that they could emulate southern celadon as closely as possible. This raises an important question about the two Caocun Group 2 celadons. These are clearly *northern* bodies, yet the evidence for the manufacture of high-temperature celadon at Caocun is extremely limited. Were these also models, and were the potters at Caocun emulating the northern celadons, implying that these were already in production? At present the resolution of these issues awaits a complete analysis of the excavated materials at Caocun. The Caocun Group 2 ware glazes are consistent with the other northern products and in particular resemble the wares produced in the nearby celadon kilns

at Anyang, as might be expected if they were being copied by the lead-glaze potters but were not made at the site. Whatever the function of the celadon wares recovered from Caocun, their presence indicates that the potters there were familiar with celadon products and, whether intentionally or not, this is likely to have had an influence on their products.

#### 4.2 Glaze formulation

All the glazes are fluxed by CaO. P<sub>2</sub>O<sub>5</sub> contents of around 0.7% (Table 4), coupled with 1-2% MgO suggest that the lime was added as vegetal ash (e.g. of wood or bracken). The glazes contain significant Al<sub>2</sub>O<sub>3</sub>, and it therefore seems plausible that they were produced by mixing ash with the body clay. We compare the ashes used to flux the northern celadon glazes by normalising the ash-related fluxes to 100%, shown in Table 5. The fluxes show a limited range of variation, and all have very low MnO, below the detection limit. Several typical North China plant ash compositions (provided by Zhang 1984) were normalised in the same way, and it is seen (Table 5) that the ashes are very variable between species and frequently have a significant concentration of MnO, with the exception of the low manganese in poplar ash. We therefore infer that all the early northern celadon glazes analysed were prepared with the ashes of similar types of plant/wood. The minor variations of MgO, K2O and CaO can be attributed to minor variables such as different harvesting seasons, the underlying geomorphology or the parts harvested from the tree (twigs, trunks etc) (Jackson 2008).

To investigate the formulation of the glaze, we have modified the approach originally proposed by Hurst and Freestone (1996) and Tite et al (1998) to determine the original constituents of lead glazes and which has been widely applied (see e.g. Walton and Tite 2010, Waksman et al 2017). According to this method, if the glaze comprises a mixture of ash flux and body clay, and the ash contained relatively minor quantities of clay components such as SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and FeO, then the ratios of these components should be similar in both glaze and body.

Figs.5 a-d compare the pertinent inter-element ratios. It is observed that only the Xing and the southern Caocun Group 1 wares lie close to the 1:1 line. This implies that the ash used to make the Xing glazes was low in clay components, as appears to have been the case for southern celadons. The alternative would be that clay components were present in the ash at relatively high concentrations and coincidentally were in precisely the same proportions as in the body clay, which seems unlikely. The other wares plot towards the body axis in the plot of Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>, (Fig. 5a) indicating that the glazes contained an additional component which was rich in silica and poor in alumina relative to the body. Similarly, departures from the 1:1 trend in terms of FeO/Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub>/FeO indicate that the added siliceous material contained FeO and TiO<sub>2</sub> in different proportions to their concentrations in the bodies. The FeO/TiO<sub>2</sub> and FeO/Al<sub>2</sub>O<sub>3</sub> ratios are higher in the glazes indicating that the added material was relatively high in iron oxide.

The conclusion that the glazes apart from the Xing ware and southern ware Caocun Group 1 contain a third component in addition to ash and body clay is, of course, dependent upon the assumption that the ashes in these glazes were, like those used in the Xing and southern wares low in SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and FeO. This assumption seems likely because, as discussed above, the glaze ashes were similar in composition to the Xing wares and likely to represent use of the same type of wood. Furthermore, as a general observation, senior trunks of wood such as poplar normally contribute little silica and alumina to their ash (Zhang 1984

and Jackson 2008). In addition, the low concentration of MnO in the glazes also suggests that the ash had low FeO, as we observe a general correlation between MnO and FeO in published Chinese wood ash compositions and their concentrations are of the same order (see Zhang 1984). On this basis, if an iron-rich ash had been used, one would expect that the addition of one percent iron to the glaze would typically (although not invariably) result in the addition of around one percent manganese and would mostly add more than 0.1% MnO. Therefore, we believe that the assumption that the wood ash contributed only minor iron oxide, alumina and silica to the glazes is justified.

Finally, we also note that if a highly aluminous and siliceous glaze ash had been used, which also had a high lime flux content, then when this ash was produced by burning wood, it would begin to sinter to a clinker at the temperatures of a large open fire, around 800°C, in a manner analogous to low-fired pottery made with a calcareous clay, and its use as a glaze flux would have required extensive crushing and grinding.

The variations in the ratios between Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub> and FeO suggest that the siliceous material added to the glazes was not pure silica but was an impure material. A likely candidate, which is ubiquitous in North China, is loess, which was widely used as a building material but also for pyrotechnological purposes, such as the production of bronze moulds (Freestone et al 1989, Liu et al 2013) and the pottery sculpture of the Terracotta Army (Quinn et al. 2017). Loess samples from northern China (Zhengzhou, Luoyang and Xi'an), as well as loess bronze moulds from Anyang were analysed by Freestone et al. (1989). They typically contain around 70% SiO<sub>2</sub>, 12% Al<sub>2</sub>O<sub>3</sub>, 3.5% FeO and 0.7% TiO<sub>2</sub>. Addition of such a component to the glaze mixture might be expected to result in the differences seen in Fig. 5.

In order to test the hypothesis that loess was added as a component to the celadon glazes, we have calculated the *flux-free compositions* (FFCs) for glazes, bodies and loess, where an FFC is the composition less the fluxing components of vegetal ash (CaO, MgO, K<sub>2</sub>O, Na<sub>2</sub>O and P<sub>2</sub>O<sub>5</sub>), and these are compared in Fig. 6. If loess was a glaze component, then the FFC of the glaze should lie between the FFC of the ceramic body and the FFC of loess. Fig. 6 shows that the Zhaili data are fully consistent with a loess addition to the glaze. Most Luoyang samples also fit this model. However, the Anyang, some Luoyang and Caocun Group 2 glazes appear to require an addition which is higher in silica than typical loess. The precise identity of this additive is not clear, it appears to have contained some iron and may have been loess which had been refined by washing or by gravity settling in water. Other possibilities include siliceous sand, crushed sandstone or quartzite. Loess would have had practical benefits as a glaze additive, in that it is very fine grained (20-60  $\mu$ m; Freestone et al., op.cit.) and therefore reactive and is a very loose or friable material, and easy to mix. The Zhaili glazes appear to contain a loess richer in clay, which may have been easier to obtain in the region of the Zhaili kiln site which is distant from the others (Fig. 1).

Xing celadon is not only exceptional among the northern wares analysed in that its glaze composition does not require an addition of loess or similar quartz-rich material. It also has exceptionally high Na<sub>2</sub>O, at around 3%, relative to around 0.5% in the glazes of the other wares. Given that Na<sub>2</sub>O in the bodies is less than 0.5%, and that inland lime-rich wood ashes do not normally contain high Na<sub>2</sub>O, this implies an addition from some other source. It is unclear as to the form in which the excess Na<sub>2</sub>O was added to the glazes. While common salt, NaCl, would be a possibility, we have detected no chlorine under counting conditions favouring low detection (detection limit c.0.12% Cl in all cases, and for several samples at high count rates with a detection limit of 0.02%). Although we cannot confirm an NaCl

addition analytically, this does not mean that NaCl was not added to the glaze; for example, chlorine is not detected in the glazes of European salt-glazed stonewares which were fired to similar temperatures (Freestone and Tite 1997). Loess contains moderate soda, up to around 3% Na<sub>2</sub>O (Freestone et al 1989), but this is too low to produce the Na<sub>2</sub>O concentrations in the glazes, and it has in any case been observed above that Xing ware glazes appear to have contained no loess addition. Addition of a rock rich in the sodic feldspar albite (NaAlSi<sub>3</sub>O<sub>8</sub>) is another alternative, and feldspar-bearing porcelain and glaze stones were a raw material in the production of southern ceramics. It has been suggested that feldspathic rocks were added to early Xing white porcelain glazes by Kerr and Wood (2004: 156-157) and later Ding northern white porcelains by Cui et al. 2012, and their addition to Xing celadons would be another early example of such a practice. Around 20% albitic feldspar (NaAlSi<sub>3</sub>O<sub>8</sub>), which contains around 11% Na<sub>2</sub>O, would have had to be added to the Xing glazes to generate the 3% Na<sub>2</sub>O present in the glazes. As the Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> ratios of albite and Xing body are similar (c. 0.3), albite additions would not have significantly disrupted the relationships seen in Fig.5a, allowing the possibility of an albite addition. However, as albite contains only small amounts of FeO, the ratio FeO/Al<sub>2</sub>O<sub>3</sub> should be reduced in the glaze relative to the body. The absence of any perturbation of FeO/Al<sub>2</sub>O<sub>3</sub> in the Xing glazes in Fig. 5b appears to rule out albite as a source of sodium. Albite-rich rocks such as granite or pegmatite seem unlikely additives; they have lower Al<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub> ratios due to the presence of quartz, and the larger quantities required to provide the required amount of sodium, would have disrupted the relationship seen in Fig. 5b. Therefore, at the present time, we have no firm evidence as to the form in which the soda was added to the Xing glaze, and must leave this question open.

#### 4.3 Coloration

The colours of the ceramics result from the FeO and  $TiO_2$  concentrations in the bodies and glazes, as well as their oxidation states. At the present time, we do not have information about the relative oxidation states of the different northern celadons, and for present purposes we assume a common kiln technology and similar firing conditions.

The early northern celadon bodies are typically pale in colour, and usually appear yellowish/ivory or greyish which can be observed from the unglazed areas shown in Figs.2b and 2d. Their FeO concentrations distribute in a narrow range around 1% with Zhaili being an exception around 1.6%, yet generally low. The different iron oxide concentrations in celadon bodies from various sites are shown in the box-and-whisker plot, where total iron in all wares is expressed as FeO (Fig.7). FeO contents of the bodies of well-known later northern celadons such as Yaozhou, Ru and Jun wares made in the Tang, Song and Jin dynasties (7-13<sup>th</sup> century, Shi et al. 2017, Ding et al. 2013, 2014) are significantly higher (1.9-3.0%) than those of the early northern celadons measured here, as are those of the Yue wares produced in South China (c. 1.7%, Xiong et al. 2010), which usually produced grey or dark grey bodies (Fig. 7).

The very low contents of iron oxide determined in the present study shows that high quality kaolinite clay was exploited in Hebei and Henan provinces in the sixth century, and this is likely to have paved a crucial foundation for the development of white porcelain. The eastern and southern borders of the Taihang mountains, which embrace the western parts of Hebei and Henan provinces, is one of the major mining areas of kaolin in modern China (Li 1998). This mining region roughly coincides with the distribution of kiln sites 2-5 in Fig.1. Meanwhile, a more complex clay which contains more impurities was used in the Zhaili kiln,

(site 1 in Fig. 1) which is likely to reflect the geological situation of Shandong province, located away from the Taihang kaolin deposits.

It is also noticed that the TiO<sub>2</sub> concentrations of Xing celadon are below 1.1%, while all the other northern groups scatter in the range of 1.1-1.5%. Low TiO<sub>2</sub> has been reported in later white porcelain bodies such as Xing, Ding and Gongyi by Li (1998) and Cui et al. (2012). Higher TiO<sub>2</sub> leads to a yellowish tone in the body, which would not be desirable in the production of white porcelain. Xing, Luoyang and Anyang kilns are all known to have produced white porcelain at a relatively early stage, but Xing porcelain is generally considered to have been most white, and this might be explained by the low TiO<sub>2</sub> content of the kaolin exploited by the Xing potters.

FeO (0.7-3.8%) and TiO<sub>2</sub> (0.3-1.2%) are the dominant colorants of the early northern celadon glazes. Under reducing conditions the yellowing effect of titania can combine with the bluish colour of iron to produce a celadon of green appearance (Wood 2011). However, our samples have yellow Munsell hues of 5Y (Table 2), implying firing in an oxidising atmosphere where both iron and titanium oxides contribute the yellow hue. Most have Munsell colours value of 7, implying a bright yellow, but the colour value of Zhaili celadon is much darker at 4. This difference is likely to reflect the high iron oxide in the Zhaili glazes, averaging 3% FeO, relative to 0.7-1.8% FeO in the products of other kilns (Table 4).

In contrast to the northern celadon bodies, which have particularly low FeO contents (Fig. 7a), the iron oxide contents of the early northern celadon glazes are not much lower than that those of the later northern celadon such as Jun (av. = 1.7%) and Ru ware (av. = 1.8%), or southern celadon such as Yue ware (av.=2.1%). This is likely to reflect the addition of loess, which is relatively high in iron, to the glazes as discussed above. The particularly high FeO in Zhaili celadon glazes reflects the addition of high Fe clay-rich loess to the glaze; Xing celadon glaze has the lowest FeO of all the glazes at around 1%, because it appears that it was formed as a mixture of body clay, ash and a soda component, with no added loess.

#### 4.4 Relative firing conditions

The attainment of firing temperatures high enough vitrify the pale kaolinitic bodies and mature their lime-rich glazes would have been a significant technical problem for the northern potters to overcome, but unfortunately, due to limited archaeological evidence, little is known about the kiln construction and firing technology in the North. What is understood is that the high temperatures used in firing southern wares were achieved using "dragon kilns" built along hilly slopes, and that this approach was not adopted in the North as suitable locations for building dragon kilns in the North China Plain are limited. However, it is clear that temperatures comparable to those in the South were attained from the macro and micromorphologies of the samples and their compositions, which if anything are more refractory than those of southern ceramics due to the higher alumina contents of the northern clays (Fig. 4).

The continuous vitrification, well generated mullite in the body and the anorthite in the interface between the body and glaze all suggest that the celadon was fired at high temperatures, in excess of 1200°C over a relatively long duration. Furthermore, all of the celadons analysed show broadly similar micromorphological characteristics (Table 2) implying a relatively constant firing regime between kilns. Based upon the shape, abundance and size of the relict quartz grains, which would have dissolved with increasing firing

temperature and duration, the Zhaili and Anyang wares are likely to have been fired at lower temperature or shorter duration than the Luoyang vessels, while Xing celadon was probably highest fired. The apparent frequency of mullite also supports a firing sequence of Zhaili, Anyang<Luoyang<Xing. It should be understood that this is a relative understanding based upon qualitative observations and an underpinning assumption is that the (admittedly minor) compositional differences between the northern porcelain bodies had no significant effect. This remains to be tested by more direct measurements of firing temperature, such as dilatometry and replication. The southern celadons of Caocun Group 1 have much higher SiO<sub>2</sub> contents than the northern wares, and their higher relict quartz and lower mullite is likely to reflect this compositional difference rather than a difference in firing temperature.

As is well-established, Chinese stoneware producers typically formulated their glazes empirically so that they unknowingly took advantage of the minimum melting temperature (eutectic) lime-alumina-silica system and matured at temperatures attainable with the available kiln technology (Wood 2009, Yin et al. 2011). Additional constraints on glaze composition would have been the need to achieve a particular colour and to ensure a good fit of expansion coefficients between the body and the glaze to avoid crazing or peeling. The glaze compositions measured in the present study are shown in the ternary phase diagram CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> in Fig. 8. Na<sub>2</sub>O, K<sub>2</sub>O and MgO were recast as equivalent weights of CaO. It should be noted that the temperatures shown are unlikely to accurately reflect the melting temperatures of the glazes, as they are lowered in the multi-component system. Even so, optimal glaze compositions should cluster around the low-melting cotectic lines and eutectics points of the phase diagram, as this would favour firing at lower temperatures. The celadon glazes from Anyang, Caocun, Luoyang and Zhaili cluster around the pseudowollastoniteanorthite-tridymite eutectic, as would be expected. This was achieved firstly by ensuring an appropriate ratio of calcium-rich ash to clay, but also by adding an appropriate amount of loess or similar siliceous material to the glaze mixture. As pointed out by Wood (2009) the glazes of southern China typically had a silica/alumina ratio of around 4.5:1 by weight, corresponding to the siliceous stoneware clays which were used in the South, and corresponding to the eutectic composition. A simple body clay plus ash mixture could therefore be used to produce a glaze without additions of other components. However, this was not possible for the northern potters. Mean silica/alumina ratios for the bodies of the northern celadons are 2.8 (Caocun 2), 2.7 (Anyang), 3.3 (Zhaili) and 2.5 (Luoyang) and binary mixtures of these clays with lime-rich ash would have produced glaze compositions requiring maturing temperatures which were undesirably high, possibly unattainable. The potters therefore modified their glazes by the addition of loess with high silica/alumina ratio, producing glazes which were remarkably consistent between Anyang (4.0), Zhaili (4.1) and Luoyang (4.1), diverging slightly for Caocun 2 (3.6). It appears that although the bodies of the celadons have varying alumina/silica ratios, and the compositions of loess added were relatively variable, the outcome of the manipulation of the glaze recipes at the different kilns was to result in a convergence of compositions which cluster around the optimum value. A departure from the foregoing pattern is seen for the Xing celadon where the glaze shows a similar SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratio (3.3) to the body (3.2) and no loess was added. This is reflected in the position of the Xing glazes in Fig. 8, where they are seen to plot away from the eutectic stretching out along the pseudowollastonite-anorthite cotectic. At face value, this might be taken to suggest that the Xing glazes matured at higher temperatures than those of the other celadon wares. This would be consistent with our tentative interpretation of the microstructures of the bodies which suggests that the Xing celadons were fired to higher temperatures than the other wares. However, the Xing glazes have very high added soda, averaging 2.9% Na<sub>2</sub>O as opposed to less than one percent for the other kilns. Soda is an

extremely effective flux, much more so than lime and it seems likely that the effect of the soda was to significantly lower the maturing range of the Xing glazes, bringing them close to those of the wares from the other kilns.

## 5. Conclusions

Following a lag of more than two thousand years after its introduction in the South, celadon manufacturing technology appeared in a mature form quite abruptly in North China, spreading rapidly across Shandong, Hebei and Henan provinces, probably within a few decades. The new northern celadons have high-fired vitrified bodies and glazes fluxed with vegetal ash which are similar to those of their southern counterparts, but differ in certain crucial respects. The porcelain stone raw materials used in southern celadons were not accessible in North China, and sedimentary kaolinitic clays richer in alumina and poor in iron oxide were exploited. It seems likely that this practice started in Shandong, in the corridor linking the South and the North, where the kaolin was richer in iron oxide and silica. Zhaili is likely to represent the celadons produced in this region. Higher quality kaolin (c. 22-29% Al<sub>2</sub>O<sub>3</sub>) with very little iron (c. 1%) was then explored in the east border of the Taihang mountains range in Henan and Hebei, yielding dense and pale celadon bodies such as those of Anyang, Luoyang and Xing. Differences in the compositions of the clays from the various kilns indicate the diversity of the northern production and reflect the ability of the northern producers to adapt to their new materials and firing conditions.

Previously, northern potters had glazed their ceramics using lead as a flux, which required a firing 2-300°C lower than the ash-glazed celadons. However, a small number of southern celadons discovered at Caocun, a kiln specialising in lead-glazed wares, provides evidence that the lead-glaze producers were familiar with southern celadon, and this may provide a perspective into the interaction between different ceramic productions in the end of the Northern Dynasties. Furthermore, a widespread familiarity with southern wares at the level of the traditional ceramic producer, as well as at the level of the elite in whose tombs these wares were placed, may help to explain the relatively rapid adoption of celadon technology across the region. However, we do not yet understand the kiln technology used at this stage in the North to attain the high temperatures equivalent to those reached by the famous "dragon kilns" of the South.

The adoption of the local high-alumina kaolin required further adjustments to the southern technological package. This was particularly the case with respect to the glazes, as the southern formula of body clay plus wood ash would not produce glaze compositions corresponding to the low temperature melting region of the lime-magnesia-alumina-silica system, due to the high alumina/silica ratios of the northern clays. To produce glazes with suitable firing properties, the northern artisans added a third component to the recipe. Typically this involved adding a material with a higher silica/alumina ratio. The Shandong (Zhaili) potters added the local loessic clay, which lowered the melting temperature of the glaze but which added a large amount of iron and titanium oxides, making the glazes darker. The potters of Anyang and Luoyang, on the other hand, exploited a silica-rich loess or similar quartz-rich material, which had higher SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> ratios so that less material needed to be added for the same effect, and which therefore resulted in pale glazes with lower iron oxide. Exceptionally, the Xing potters do not appear to have added an additional siliceous material

to their glaze but a sodium-rich component, which lowered the glaze maturing range. The identity of this component remains to be proven; irrespective of its origin, however, this sodic additive allowed the Xing kilns to produce a celadon glaze with less iron oxide than other northern kilns.

The use of vegetal ash as a flux was a new technology to the North as previously glaze had been fluxed with lead oxide. However, the northern ash appears to have been different in composition from that used in the South, imparting lower P<sub>2</sub>O<sub>5</sub>, MgO and MnO than in the southern glazes and might have been less thoroughly washed, as the soluble sodium and potassium oxides are higher than in southern celadon glazes. Even so, a similar type of wood ash, which contributed little silica, alumina and iron oxide appears to have been used to flux the glaze in the North.

The overall picture is of a rapid adoption of celadon technology in the North, with localised strategies to adapt the southern celadon package. The transfer of celadon production from the South to the North required the potters of each kiln to become familiar with the local resources and adjust recipes and firing conditions accordingly. Given that ceramics which closely resembled celadon visually were already produced in the North using lead glazes at significantly lower firing temperatures, the impetus to adopt the new technology must have been considerable, presumably due to the improved handling properties (fracture strength, hardness) of the celadon wares.

Of the northern producers there is little doubt that the Xing potters were the most creative, producing a glaze which was similar to the calcium-alkali dual fluxed glaze popular in the later ceramic production of the Song Dynasty. Furthermore, our results indicate that the Xing body was more mature, suggesting that higher temperatures may have been attained in the Xing kilns, with a better control of firing technology. The pure lower-titanium clays of the Xing celadon body, and the low iron contents of its glaze are likely to have reduced the ivory tones characteristic of the Luoyang, Anyang and Caocun Group 2 wares. While white porcelain was soon produced in all of these areas, on the basis of our investigation of the early celadon production, the pathway to porcelain adopted at the Xing kiln is likely to have been different, and it is therefore not surprising that it was to emerge as the most well-known of the early porcelain producers in China.

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| Site     | Туре | Number | Dating                               | Location         |
|----------|------|--------|--------------------------------------|------------------|
| Zhaili   | Kiln | 8      | Northern Qi-Sui (550 - 618 CE)       | Central Shandong |
| Xing     | Kiln | 8      | Late Northern Qi-Sui (570s - 618 CE) | South Hebei      |
| Caocun   | Kiln | 4      | Northern Qi(550-580 CE)              | South Hebei      |
| <b>A</b> | Kiln | 7      | Sui ( 581 - 618 CE )                 | Newskie Lieuweuw |
| Anyang   | Tomb | 2      | Sui-Early Tang (581 - early 600s CE) | North Henan      |
| Luoyang  | City | 6      | Sui-Early Tang (581 - early 600s CE) | West Henan       |

# Table 1Sources of celadon samples

| Site   | No. | Vitri<br>Stage | Glaze<br>Thkns<br>(µm) | Crystal<br>at<br>Interface | Quartz Size in<br>Body(µm) | Quartz<br>Abun<br>in Body | Mullite in<br>body | Glaze Colour                     | Body<br>Colour |
|--------|-----|----------------|------------------------|----------------------------|----------------------------|---------------------------|--------------------|----------------------------------|----------------|
|        | T1  | 2              | 165                    | extensive                  | coarse, 30-50              | 40%                       | absent             | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | T2  | 2              | 150                    | extensive                  | coarse+,30-80              | 40%                       | present            | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | Т3  | 3              | 150-<br>240            | sparse                     | medium, 20-30              | 40%                       | abundant           | Moderate olive brown<br>(5Y 4/4) | Grey           |
| Zhaili | T4  | 3              | 180                    | extensive                  | coarse, 30-50              | 50%                       | present            | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | T7  | 2              | no                     | none                       | coarse, 30-50              | 50%                       | absent             | Trivet, unglazed                 | Pale grey      |
|        | Т8  | 3              | 200-<br>300            | sparse                     | coarse, 30-50              | 40%                       | absent             | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | Т9  | 3              | 380                    | none                       | coarse, 30-50              | 40%                       | absent             | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | T10 | 3              | 380                    | none                       | coarse, 30-50              | 40%                       | absent             | Moderate olive brown<br>(5Y 4/4) | Grey           |
|        | T7  | 3              | 350                    | sparse                     | coarse, 30-50              | 20%                       | present            | Light olive grey (5Y 6/1)        | Grey           |
|        | Т8  | 3              | 190                    | extensive                  | coarse, 30-50              | 20%                       | present            | Yellowish grey (5Y 7/2)          | Grey           |
|        | Т9  | 4              | 300                    | none                       | medium, 20-30              | 10%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
|        | T10 | 4              | 350                    | extensive                  | coarse, 30-50              | 20%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
| Xing   | T11 | 4              | 220                    | sparse                     | coarse, 30-50              | 10%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
|        | T12 | 3              | 250                    | extensive                  | medium, 20-30              | 20%                       | present            | Yellowish grey (5Y 7/2)          | Grey           |
|        | T13 | 3              | 400                    | sparse                     | coarse, 30-50              | 20%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
|        | T14 | 4              | 250                    | none                       | medium, 20-30              | 10%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
|        | Т32 | 2              | 360                    | sparse                     | coarse, 30-50              | 40%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
| Cassur | T40 | 2              | 155                    | none                       | coarse, 30-50              | 40%                       | present            | Yellowish grey (5Y 7/2)          | Grey           |
| Caocun | T41 | 3              | 280                    | sparse                     | fine, 10-30                | 20%                       | abundant           | Yellowish grey (5Y 7/2)          | Grey           |
|        | T42 | 3              | 250                    | extensive                  | medium, 20-30              | 20%                       | abundant           | Yellowish grey (5Y 7/2)          | Pale grey      |
|        | KT1 | 3              | 200                    | extensive                  | medium, 20-30              | 20%                       | present            | Yellowish grey (5Y 7/2)          | Pale grey      |
|        | KT2 | 3              | 200                    | none                       | fine,10-30                 | 20%                       | present            | Light olive grey (5B 7/1)        | Pale grey      |
|        | КТЗ | 2              | 250                    | sparse                     | medium, 20-30              | 40%                       | present            | Yellowish grey (5Y 7/2)          | Pale grey      |
| Anyang | KT4 | 3              | 200                    | sparse                     | medium, 20-30              | 40%                       | abundant           | Light olive grey (5B 7/1)        | Pale grey      |
|        | KT5 | 3              | 200                    | sparse                     | medium, 20-30              | 40%                       | present            | Yellowish grey (5Y 7/2)          | Pale grey      |
|        | КТ6 | 6              | 550                    | none                       | fine, few, 20              | 5%                        | abundant++         | Yellowish grey (5Y 7/2)          | Pale grey      |
|        | KT7 | 4              | no                     | none                       | fine, 10-20                | 10%                       | abundant           | Unglazed                         | Pale grey      |
|        | Т3  | 2              | >300                   | extensive                  | medium, 20-30              | 40%                       | present            | Yellowish grey (5Y 7/2)          | Pale grey      |

# Table 2 Micromorphologies and colours of individual sherds analysed

|         | T4        | 2 | 125 | extensive      | medium, 20-30 | 40% | present  | Light olive grey (5Y 6/1) | Pale grey |
|---------|-----------|---|-----|----------------|---------------|-----|----------|---------------------------|-----------|
|         | 06<br>T5  | 5 | 200 | extensive<br>+ | coarse, 30-50 | 20% | abundant | Yellowish grey (5Y 7/2)   | Pale grey |
|         | 89T<br>16 | 3 | 250 | sparse         | fine, 10-20   | 20% | present  | Yellowish grey (5Y 7/2)   | Pale grey |
|         | 89T<br>17 | 2 | 180 | sparse         | coarse, 30-50 | 20% | present  | Yellowish grey (5Y 7/2)   | Pale grey |
| Luoyang | 89T<br>19 | 2 | 300 | sparse         | coarse, 30-50 | 20% | present  | Yellowish grey (5Y 7/2)   | Pale grey |
|         | 89T<br>20 | 3 | 350 | sparse         | coarse, 30-50 | 20% | present  | Yellowish grey (5Y 7/2)   | White     |
|         | 89T<br>21 | 3 | 800 | sparse         | coarse, 30-50 | 20% | present  | Yellowish grey (5Y 7/2)   | White     |

| Site    | Group   | No.     | Na <sub>2</sub> O | MgO  | Al <sub>2</sub> O <sub>3</sub> | SiO2  | K <sub>2</sub> O | CaO  | TiO <sub>2</sub> | FeO  | Total  |
|---------|---------|---------|-------------------|------|--------------------------------|-------|------------------|------|------------------|------|--------|
|         |         | T1      | 0.15              | 0.29 | 20.49                          | 74.09 | 1.50             | 0.56 | 1.10             | 1.81 | 100.00 |
|         |         | T2      | 0.17              | 0.35 | 21.26                          | 73.56 | 1.50             | 0.38 | 1.29             | 1.49 | 100.00 |
|         |         | Т3      | 0.28              | 0.46 | 22.43                          | 71.16 | 1.92             | 0.78 | 1.33             | 1.63 | 100.00 |
|         |         | T4      | 0.15              | 0.41 | 23.51                          | 70.89 | 1.77             | 0.43 | 1.26             | 1.57 | 100.00 |
|         |         | T7      | 0.20              | 0.35 | 22.75                          | 70.95 | 2.00             | 0.64 | 1.38             | 1.73 | 100.00 |
| Zhaili  |         | Т8      | 0.20              | 0.33 | 21.97                          | 72.49 | 1.57             | 0.55 | 1.20             | 1.70 | 100.00 |
|         |         | Т9      | 0.18              | 0.32 | 22.03                          | 72.26 | 1.56             | 0.57 | 1.45             | 1.64 | 100.00 |
|         |         | T10     | 0.17              | 0.48 | 21.64                          | 73.36 | 1.72             | 0.38 | 1.22             | 1.05 | 100.00 |
|         |         | Average | 0.19              | 0.37 | 22.01                          | 72.35 | 1.69             | 0.54 | 1.28             | 1.58 | 100.00 |
|         |         | Stdev   | 0.04              | 0.07 | 0.93                           | 1.25  | 0.19             | 0.14 | 0.11             | 0.23 |        |
|         |         | T7      | 0.24              | 0.50 | 20.45                          | 73.60 | 2.22             | 0.67 | 0.98             | 1.33 | 100.00 |
|         |         | Т8      | 0.42              | 0.61 | 22.23                          | 71.13 | 2.49             | 0.64 | 1.02             | 1.46 | 100.00 |
|         |         | Т9      | 0.34              | 0.57 | 21.99                          | 71.89 | 2.36             | 0.56 | 0.96             | 1.34 | 100.00 |
|         |         | T10     | 0.31              | 0.54 | 22.80                          | 71.14 | 2.48             | 0.56 | 0.95             | 1.22 | 100.00 |
| Nin a   |         | T11     | 0.26              | 0.57 | 22.45                          | 71.58 | 2.41             | 0.49 | 0.91             | 1.34 | 100.00 |
| Xing    |         | T12     | 0.30              | 0.46 | 23.85                          | 69.80 | 1.92             | 0.97 | 1.14             | 1.55 | 100.00 |
|         |         | T13     | 0.29              | 0.57 | 22.83                          | 71.04 | 2.42             | 0.59 | 1.07             | 1.19 | 100.00 |
|         |         | T14     | 0.47              | 0.55 | 22.78                          | 71.36 | 2.23             | 0.38 | 0.95             | 1.27 | 100.00 |
|         |         | Average | 0.33              | 0.54 | 22.42                          | 71.44 | 2.32             | 0.61 | 1.00             | 1.34 | 100.00 |
|         |         | Stdev   | 0.08              | 0.04 | 0.97                           | 1.06  | 0.19             | 0.17 | 0.08             | 0.12 |        |
|         | Crown 1 | T32     | 0.16              | 0.60 | 16.33                          | 76.93 | 3.41             | 0.23 | 0.92             | 1.42 | 100.00 |
|         | Group 1 | T40     | 0.23              | 0.42 | 13.42                          | 81.02 | 1.94             | 0.44 | 0.97             | 1.55 | 100.00 |
| Coordin |         | Average | 0.20              | 0.51 | 14.87                          | 78.98 | 2.68             | 0.34 | 0.95             | 1.48 | 100.00 |
| Caocun  | Crown 2 | T41     | 0.20              | 0.43 | 24.81                          | 69.34 | 2.27             | 0.51 | 1.54             | 0.90 | 100.00 |
|         | Group 2 | T42     | 0.23              | 0.38 | 24.87                          | 68.94 | 2.44             | 0.48 | 1.36             | 1.30 | 100.00 |
|         |         | Average | 0.22              | 0.40 | 24.84                          | 69.14 | 2.36             | 0.50 | 1.45             | 1.10 | 100.00 |
|         |         | T1      | 0.28              | 0.42 | 25.91                          | 68.31 | 2.40             | 0.29 | 1.33             | 1.07 | 100.00 |
|         |         | T2      | 0.30              | 0.46 | 25.71                          | 68.17 | 2.48             | 0.32 | 1.32             | 1.24 | 100.00 |
|         |         | Т3      | 0.37              | 0.37 | 26.01                          | 68.00 | 2.45             | 0.31 | 1.28             | 1.20 | 100.00 |
|         | Kiln    | T4      | 0.26              | 0.44 | 25.63                          | 68.18 | 2.59             | 0.35 | 1.34             | 1.22 | 100.00 |
|         |         | T5      | 0.28              | 0.45 | 25.85                          | 68.55 | 2.60             | 0.25 | 1.24             | 0.77 | 100.00 |
| Anyang  |         | Т6      | 0.37              | 0.49 | 25.10                          | 68.72 | 2.66             | 0.28 | 1.33             | 1.04 | 100.00 |
|         |         | T7      | 0.44              | 0.45 | 25.46                          | 68.41 | 2.37             | 0.40 | 1.33             | 1.13 | 100.00 |
|         | T       | Т3      | 0.23              | 0.34 | 24.08                          | 70.76 | 2.34             | 0.29 | 1.22             | 0.75 | 100.00 |
|         | Tomb    | T4      | 0.33              | 0.39 | 25.68                          | 68.42 | 2.48             | 0.20 | 1.28             | 1.22 | 100.00 |
|         |         | Average | 0.32              | 0.42 | 25.49                          | 68.61 | 2.49             | 0.30 | 1.30             | 1.07 | 100.00 |
|         |         | Stdev   | 0.07              | 0.05 | 0.59                           | 0.83  | 0.11             | 0.06 | 0.04             | 0.19 |        |
| 1       |         | 06 T5   | 0.46              | 0.40 | 28.28                          | 65.11 | 2.53             | 1.06 | 1.25             | 0.92 | 100.00 |
| Luoyang |         | 89T16   | 0.23              | 0.31 | 26.05                          | 68.68 | 2.08             | 0.29 | 1.44             | 0.91 | 100.00 |

# Table 3 Bulk compositions of the celadon bodies, determined by SEM-EDS

| 89T17   | 0.43 | 0.39 | 29.45 | 64.92 | 2.30 | 0.60 | 1.19 | 0.72 | 10 |
|---------|------|------|-------|-------|------|------|------|------|----|
| 89T19   | 0.38 | 0.40 | 26.39 | 68.55 | 1.77 | 0.75 | 1.13 | 0.63 | 10 |
| 89T20   | 0.36 | 0.31 | 26.73 | 68.62 | 2.00 | 0.30 | 1.08 | 0.59 | 10 |
| 89T21   | 0.38 | 0.35 | 26.83 | 68.43 | 1.96 | 0.27 | 1.26 | 0.54 | 10 |
| Average | 0.37 | 0.36 | 27.29 | 67.38 | 2.11 | 0.54 | 1.23 | 0.72 | 10 |
| Stdev   | 0.08 | 0.04 | 1.30  | 1.84  | 0.27 | 0.32 | 0.13 | 0.16 |    |

| Site    | Group      | No.   | Na₂O  | MgO  | Al <sub>2</sub> O <sub>3</sub> | SiO <sub>2</sub> | P <sub>2</sub> O <sub>5</sub> | K₂O  | CaO   | TiO₂ | MnO   | FeO  | Total  |
|---------|------------|-------|-------|------|--------------------------------|------------------|-------------------------------|------|-------|------|-------|------|--------|
|         |            | T1    | 0.54  | 2.35 | 12.83                          | 60.59            | 0.93                          | 2.30 | 15.65 | 0.89 | <0.21 | 3.85 | 100.00 |
|         |            | T2    | 0.46  | 2.29 | 14.53                          | 57.79            | 0.91                          | 2.25 | 17.64 | 0.95 | <0.21 | 2.98 | 100.00 |
|         |            | Т3    | 1.01  | 2.91 | 11.59                          | 57.86            | 1.02                          | 2.80 | 18.41 | 0.75 | <0.21 | 3.49 | 100.00 |
|         |            | T4    | 0.38  | 2.38 | 14.00                          | 56.44            | 0.72                          | 2.28 | 18.95 | 0.97 | <0.21 | 3.81 | 100.00 |
| Zhaili  |            | Т8    | 0.49  | 1.66 | 16.58                          | 61.06            | 0.52                          | 2.36 | 13.48 | 1.06 | <0.21 | 2.71 | 100.00 |
|         |            | Т9    | 0.41  | 1.65 | 15.90                          | 60.71            | 0.44                          | 2.33 | 14.69 | 1.07 | <0.21 | 2.70 | 100.00 |
|         |            | T10   | 0.33  | 1.19 | 16.87                          | 63.11            | 0.44                          | 2.46 | 12.76 | 1.06 | <0.21 | 1.75 | 100.00 |
|         |            | Ave   | 0.51  | 2.06 | 14.62                          | 59.66            | 0.71                          | 2.40 | 15.93 | 0.97 | <0.21 | 3.04 | 100.00 |
|         |            | Stdev | 0.23  | 0.58 | 1.97                           | 2.34             | 0.25                          | 0.19 | 2.45  | 0.12 |       | 0.75 |        |
|         |            | Τ7    | 2.60  | 1.71 | 16.20                          | 53.00            | 0.94                          | 2.30 | 21.46 | 0.64 | <0.21 | 1.01 | 100.00 |
|         |            | Т8    | 2.21  | 1.40 | 16.78                          | 56.26            | 0.61                          | 3.82 | 16.69 | 0.73 | <0.21 | 1.39 | 100.00 |
|         |            | Т9    | 2.86  | 1.25 | 17.60                          | 56.12            | 0.62                          | 2.57 | 17.23 | 0.72 | <0.21 | 0.97 | 100.00 |
|         |            | T10   | 3.65  | 1.38 | 15.75                          | 55.70            | 0.71                          | 2.25 | 19.20 | 0.55 | <0.21 | 0.73 | 100.00 |
| Ving    |            | T11   | 2.93  | 1.26 | 17.91                          | 57.09            | 0.63                          | 2.87 | 15.61 | 0.69 | <0.21 | 0.92 | 100.00 |
| Xing    |            | T12   | 2.83  | 1.54 | 16.86                          | 55.99            | 0.95                          | 2.38 | 17.50 | 0.74 | <0.21 | 1.11 | 100.00 |
|         |            | T13   | 3.35  | 1.31 | 16.58                          | 54.65            | 0.85                          | 2.48 | 19.12 | 0.65 | <0.21 | 0.97 | 100.00 |
|         |            | T14   | 2.81  | 1.25 | 17.97                          | 56.84            | 0.72                          | 3.18 | 15.29 | 0.78 | <0.21 | 1.09 | 100.00 |
|         |            | Ave   | 2.90  | 1.39 | 16.96                          | 55.71            | 0.75                          | 2.73 | 17.76 | 0.69 | <0.21 | 1.02 | 100.00 |
|         |            | Stdev | 0.44  | 0.16 | 0.81                           | 1.32             | 0.14                          | 0.54 | 2.06  | 0.07 |       | 0.19 |        |
|         |            | T32   | <0.24 | 3.09 | 12.82                          | 55.76            | 1.17                          | 2.05 | 22.30 | 0.85 | 0.68  | 1.28 | 100.00 |
|         | Group<br>1 | T40   | <0.24 | 2.94 | 12.00                          | 60.13            | 1.57                          | 1.34 | 18.88 | 0.70 | 0.77  | 1.50 | 100.00 |
| Cassura | _          | Ave   | <0.24 | 3.01 | 12.41                          | 57.95            | 1.37                          | 1.70 | 20.59 | 0.77 | 0.73  | 1.39 | 100.00 |
| Caocun  |            | T41   | 0.30  | 1.90 | 20.24                          | 61.25            | 0.26                          | 2.67 | 10.82 | 1.15 | <0.21 | 1.37 | 100.00 |
|         | Group<br>2 | T42   | 0.38  | 2.55 | 13.97                          | 62.82            | 0.43                          | 2.57 | 14.80 | 0.73 | <0.21 | 1.73 | 100.00 |
|         |            | Ave   | 0.34  | 2.23 | 17.10                          | 62.04            | 0.35                          | 2.62 | 12.81 | 0.94 | <0.21 | 1.55 | 100.00 |
|         |            | T1    | 0.49  | 1.58 | 15.35                          | 59.82            | 0.72                          | 2.72 | 16.80 | 0.90 | <0.21 | 1.57 | 100.00 |
|         |            | T2    | 0.41  | 1.15 | 16.19                          | 58.28            | 0.52                          | 2.35 | 18.51 | 0.98 | <0.21 | 1.59 | 100.00 |
|         | Kilo       | Т3    | 0.47  | 1.70 | 12.96                          | 59.63            | 0.79                          | 2.05 | 19.97 | 0.75 | <0.21 | 1.59 | 100.00 |
|         | Kiln       | T4    | 0.40  | 1.07 | 16.09                          | 58.62            | 0.33                          | 2.63 | 18.08 | 1.05 | <0.21 | 1.68 | 100.00 |
| Anyang  |            | Т5    | 0.27  | 3.30 | 12.84                          | 62.91            | 0.57                          | 1.88 | 16.49 | 0.40 | <0.21 | 1.24 | 100.00 |
| Anyang  |            | Т6    | 0.46  | 1.03 | 18.70                          | 61.73            | 0.31                          | 2.16 | 13.13 | 1.00 | <0.21 | 1.42 | 100.00 |
|         | Tomb       | Т3    | 0.31  | 3.67 | 12.85                          | 58.79            | 0.84                          | 2.09 | 19.95 | 0.34 | <0.21 | 0.99 | 100.00 |
|         |            | T4    | 0.31  | 1.84 | 13.94                          | 59.34            | 0.75                          | 2.16 | 18.74 | 1.15 | <0.21 | 1.70 | 100.00 |
|         |            | Ave   | 0.39  | 1.92 | 14.86                          | 59.89            | 0.60                          | 2.26 | 17.71 | 0.82 | <0.21 | 1.47 | 100.00 |
|         |            | Stdev | 0.08  | 1.02 | 2.10                           | 1.62             | 0.20                          | 0.29 | 2.24  | 0.30 |       | 0.25 |        |
| Luovere |            | 06 T5 | 0.94  | 1.61 | 16.08                          | 56.67            | 0.44                          | 3.41 | 17.88 | 0.89 | <0.21 | 2.03 | 100.00 |
| Luoyang |            | 89T16 | 0.40  | 3.21 | 17.05                          | 59.74            | 0.36                          | 2.60 | 14.49 | 0.96 | <0.21 | 1.12 | 100.00 |

# Table 4 Bulk compositions of the celadon glaze, determined by SEM-EDS<sup>3</sup>

 $^{3}$  The detection limit is 3 times weight % sigma, data below the detection limit is indicated by "<".

| 89T17 | 1.37 | 1.87 | 13.01 | 56.65 | 0.95 | 3.71 | 19.77 | 0.78 | <0.21 | 1.84 | 100.00 |
|-------|------|------|-------|-------|------|------|-------|------|-------|------|--------|
| 89T19 | 0.89 | 2.33 | 11.47 | 55.96 | 1.46 | 2.92 | 21.73 | 0.83 | <0.21 | 2.40 | 100.00 |
| 89T20 | 0.64 | 1.95 | 15.03 | 56.62 | 0.51 | 2.03 | 20.07 | 0.83 | <0.21 | 2.33 | 100.00 |
| 89T21 | 0.39 | 3.17 | 12.38 | 58.82 | 0.61 | 1.39 | 21.62 | 0.32 | <0.21 | 1.14 | 100.00 |
| Ave   | 0.77 | 2.36 | 14.17 | 57.41 | 0.72 | 2.67 | 19.26 | 0.77 | <0.21 | 1.81 | 100.00 |
| Stdev | 0.37 | 0.69 | 2.21  | 1.50  | 0.42 | 0.87 | 2.73  | 0.23 |       | 0.56 |        |

| Glazes   | MgO | P <sub>2</sub> O <sub>5</sub> | K₂O | CaO | MnO  |
|----------|-----|-------------------------------|-----|-----|------|
| Zhaili   | 10  | 3                             | 11  | 75  | b.d. |
| Xing     | 6   | 3                             | 12  | 78  | b.d. |
| Caocun 2 | 12  | 2                             | 15  | 71  | b.d. |
| Anyang   | 9   | 3                             | 10  | 79  | b.d. |
| Luoyang  | 9   | 3                             | 11  | 77  | b.d. |
|          |     |                               |     |     |      |
| Ashes    | MgO | P <sub>2</sub> O <sub>5</sub> | K₂O | CaO | MnO  |
| Oak      | 10  | 6                             | 14  | 59  | 11   |
| Poplar   | 3   | 14                            | 14  | 69  | 0    |
| Sorghum  | 20  | 8                             | 31  | 39  | 2    |
| Pine     | 8   | 5                             | 15  | 68  | 5    |

Table 5 Normalised ash components of northern celadon glazes (Table 4) and selectednorthern Chinese woods and plants (Zhang 1984)

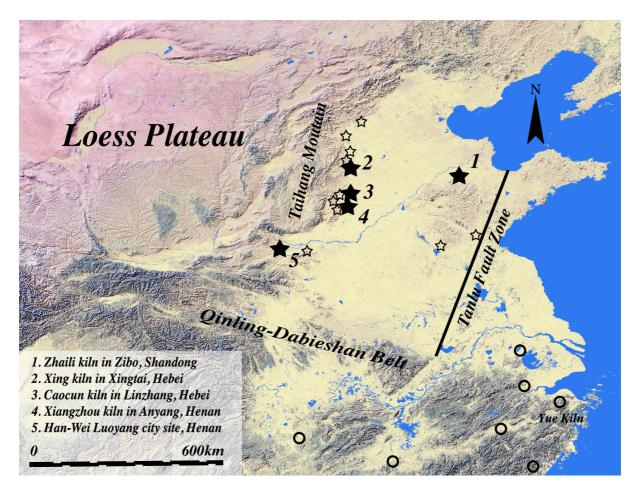


Figure 1 The distribution of celadon kiln sites by the end of the sixth century CE. The southern sites are indicated with circles and the northern sites are stars, samples in this study were selected from the solid stars with numbers 1-5.

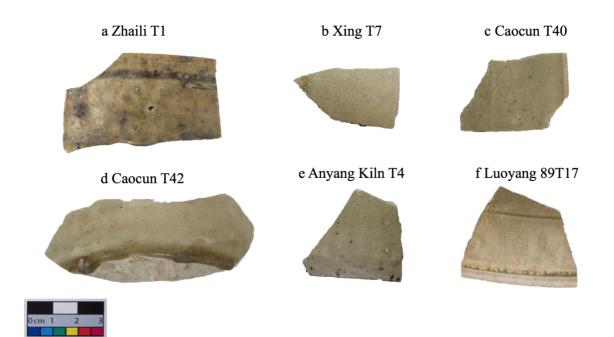


Figure 2 Examples of Analysed Celadon Sherds

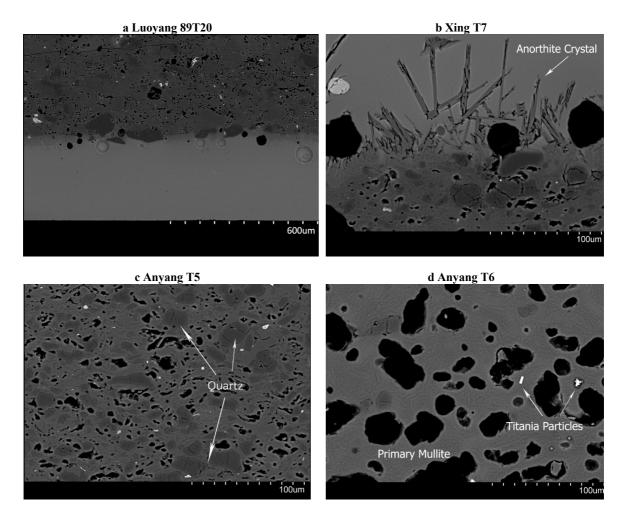


Figure 3 Back-scattered electron image showing: a. celadon glaze covering a fine body; b. anorthite crystals on the interface between the body and glaze; c. relatively lower fired celadon body; d: relatively higher fired celadon body.

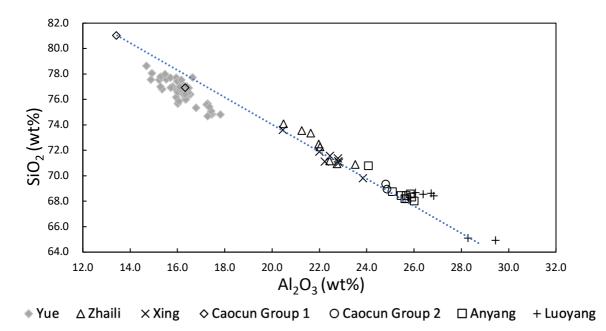
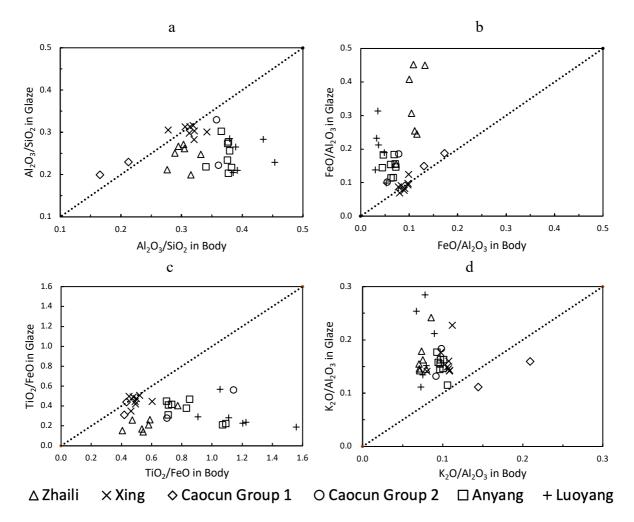


Figure 4 Concentrations of  $Al_2O_3$  versus  $SiO_2$  in the celadon bodies. Comparative data for the southern celadon Yue ware from Xiong et al. 2010. Trendline is for all the wares analysed in this paper.



*Figure 5 Comparison of oxide ratios of celadon bodies with those of glazes, the 1:1 line is indicated* 

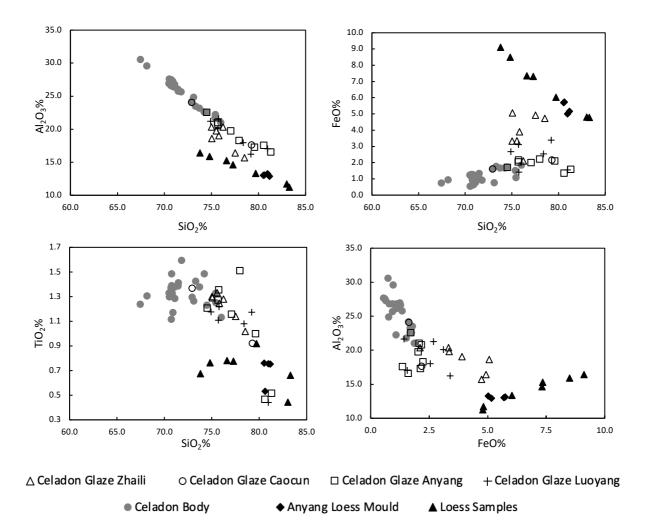
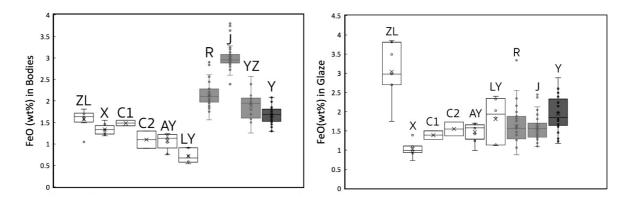


Figure 6 Comparison of flux-free compositions of loess mould and loess samples (Freestone et al. 1989) with celadon bodies and glaze analysed in the present paper



ZL=Zhaili, X=Xing, C1=Caocun Group 1, C2=Caocun Group 2, AY=Anyang, LY=Luoyang, R=Ru, J=Jun, YZ=Yaozhou, Y=Yue

Figure 7 The concentration of FeO in celadon bodies(left) and glaze(right). The open boxes are samples from this study, the shaded boxes are from Shi et al. 2017, Ding et al. 2013,2014 and Xiong et al. 2010.

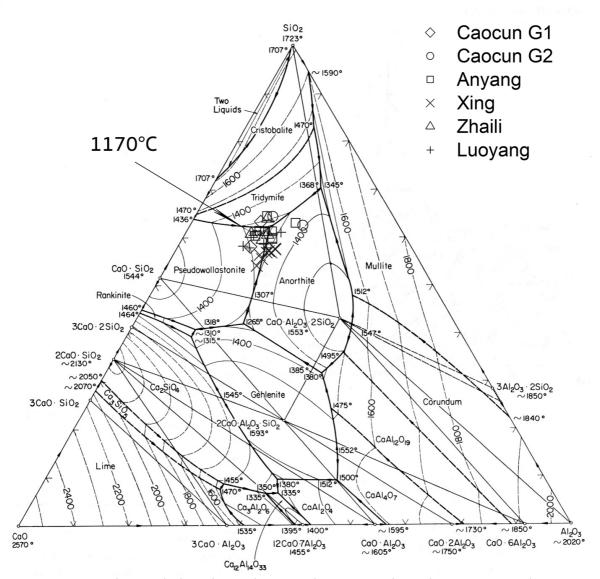


Figure 8 Northern celadon glazes shown in the ternary phase diagram CaO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> (base diagram from Levin et al. 1964:219, fig.630) . Note the clustering of most glazes around the eutectic composition, whereas the Xing glazes have lower silica and higher alumina.