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# An integrated study of glazed ceramics from tortkul sites in the Talas River Valley, Kazakhstan (10-12th c. CE)

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#### ABSTRACT

This paper presents composition and microstructural analysis of seventy-four glazed ceramics from the recently excavated tortkul sites of Arna, Talapty, and Zhargul in the lower Talas River Valley, Kazakhstan. A tortkul is a fortified medieval settlement, traditionally identified as nodes to link Silk Road trade routes through Kazakhstan. Radiocarbon dating of the sites place the occupation of these tortkuls during the early Islamic Period (10-12th c. CE). Analysis of the ceramic pastes by neutron activation analysis (NAA) identifies six distinct compositional groups, one of which is formed by the quartz bodied ceramics, and five of which have clay bodies. Most of these groups were found at multiple sites, suggesting there was an active circulation of glazed ceramics in the area, and a diversity of production centers in the larger region. The results suggest that these tortkul sites were integrated into the larger regional circulation of goods, including local craft products, and were not isolated centers. Two of the compositional groups fall within previously identified compositional groups from southern Kazakhstan. One of those groups was produced at the site of Aktobe in the Chu River Valley, and the fact that ceramics belonging to this group were recovered from the Zhargul tortkuls supports the conclusion that there was regional trade of glazed ceramics between the Chu and Talas River Valleys. Several distinct glaze types were identified, including high-lead glaze, two distinct low-lead alkali glazes, and alkali-lime glaze. The characterization of tin- and antimony-opacified glazes on four samples further strengthens our understanding of the distinct technological traditions that were employed in the production of the early Islamic style wares, and indicates that the local craftspeople in piedmont region north of the Tianshan were producing a unique opaque glazed ceramic using local raw materials.

## 1. Introduction

The Talas River Valley, which in the upper courses lies in modern day Kyrgyzstan and in the lower courses in modern day Kazakhstan, has one of the richest concentrations of ancient and medieval monuments in Northern Central Asia. Ancient settlements are mostly concentrated along the paleochannels of the Talas River and ancient canals which drew water from the river. A common site form throughout the piedmont region on the northern edge of the Tianshan, and prominently in the Talas River Valley, was the tortkul. A tortkul is a fortified medieval settlement, traditionally identified as nodes to link Silk Road trade routes through Kazakhstan. These sites vary significantly in size but have a similar structure, including: 1) defensive walls; 2) a square or rectangular site plan; and 3) towers flanking both sides of the entrance and all corners of the enclosure (Sala and Deom, 2010). Tortkuls can be found individually, in groups, or located in the proximity of larger cities (Akymbek et al., 2022). To date, only a few tortkuls have been excavated and there is practically no published information about their material culture, making them the most enigmatic category of sites in Kazakhstan. This research specifically aims to understand the role these fortified sites played in the larger economic, cultural, and political landscape by identifying if and how glazed ceramics were produced and circulated among these sites.

While the first tortkuls appear in the 6<sup>th</sup>-7<sup>th</sup> c CE in the Talas delta, prior research has suggested that the tortkuls were mainly constructed during the 9<sup>th</sup> to early 13<sup>th</sup> c. CE (Sala and Deom, 2010). This was also a period of political change in the region as well as an economic and cultural golden age for Northern Central Asia (Deom, 2009). The late 9<sup>th</sup>-10<sup>th</sup> c. CE saw the presence of the Samanid dynasty in the Talas Valley. In 893 CE, the Samanids conquered the city of Taraz, and converted the Christian church into a mosque. The Talas Valley, and the markets of Taraz in particular, served as an important trading center for exchanges between the Samanids and the nomadic Turkish tribes (Davidovich, 1998). The occupations of portions of southern Kazakhstan by the Samanids in the 9<sup>th</sup> and 10<sup>th</sup> c. is documented by Ibn Khurdadhbih (846 CE), Qudama (889 CE) and al Makdisi (985 CE) (Deom,

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2009). In the middle of the 9<sup>th</sup> c., the Karakhanids (840–1212 CE) rose to power in Semirechye region. This group, which stemmed from a number of Turkic-speaking tribes which had belonged to the Karluks, initially held territory extending from the Semirechye region south to include the western part of the Tarim Basin and the cities of Aksu and Kashgar. As they rose to power, the Karakhanids adopted Islam in 940 CE and made it the official religion (Davidovich, 1998). The Karakhanid state established its capital at Balasagun in the Chu River Valley, and their control was centered in the Chu and adjacent river valleys (Barisitz, 2017).

While the 10<sup>th</sup>-12<sup>th</sup> c. CE is usually seen as a period of decline across portions of the overland Silk Road, trade continued to a significant extent through the northern corridor of the Silk Road which passed through the Talas River Valley (Biran, 2012). During this period, there was a peak of urbanism in the river valleys on the northern edge of the Tianshan, including in the Talas River Valley (Sala and Deom, 2010). Almost every town in the region under the control of the Karakhanids had a caravanserai and bazaar, and there was extensive craft production. It was noted that Tashkent (Chach) was famous in the period for is pottery production, sandals, fabrics, and carpets (Barisitz, 2017). There were extensive quantities of glass and ceramics being produced in urban centers, and attractive pottery became more easily affordable in the period.

While it has been hypothesized that tortkuls acted as nodes to link Silk Road trade routes, there has been little systematic study of these sites to verify this hypothesis. By studying the origin of one of the most archaeological visible trade goods along the Silk Road, glazed ceramics, we hope to identify social processes and the economic basis that supported the development of trade networks, larger urban centers, and these fortified tortkuls. By focusing on the understudied tortkul sites, we hope to determine how they fit into a broader social system.

#### 1.1. Archaeological background

In 2021, the Talas team of the South Kazakhstan Integrated Archaeological Expedition of the Institute of Archaeology was named after A. Kh. Margulan (led by Y. Sh. Akymbek) conducted a widespread survey of tortkul sites in the Talas River Valley, and systematic archaeological excavation of three sites: Talapty, Arna, and Zhargul (Fig. 1) dated to the 10-12<sup>th</sup> c. CE (Table 1).

## 1.1.1. Talapty

The archaeological site of Talapty is located 4.5 km southeast of the village of the same name in the Talas district. While today the site lies 1.5 km from the left bank of the Talas River, it is located only approximately 200 m from one of the oldest channels of the Talas River – the Karabakyr passes. The site forms a tortkul, which is oriented parallel to the left bank of the river, southeast to northwest. Surrounded by an insignificant wall (width of 8–10 m and 0.2–0.3 m high). The entire site measures  $300 \times 250$  m in size, and has a preserved height of 0.1–1 m. The southeastern half is raised following the slope of the landscape. In the middle of the tortkul there is a main square-shaped structure. The surrounding wall/shaft is  $100 \times 100$  m in size and 1.5–2 m high.

Traces of towers can be identified at the corners and in the middle of the exterior tortkul walls. These towers have diameters that range from 14 to 18 m, and a preserved height of 0.3–0.5 m. The tower in the middle of the south-eastern wall is larger than the others, its diameter is 22 m, and its height is 1 m. With the exception of the south-eastern part of the structure, the exterior walls are surrounded by a moat or ditch. The width of the moat/ditch is 5–20 m, with a depth ranging from 0.2 to 2 m. Traces of ruins of building structures up to several tens of meters in size are visible around the site.

Excavations at this site began in 1980 by the expedition of the Zhambyl Regional History Museum under the leadership of K.

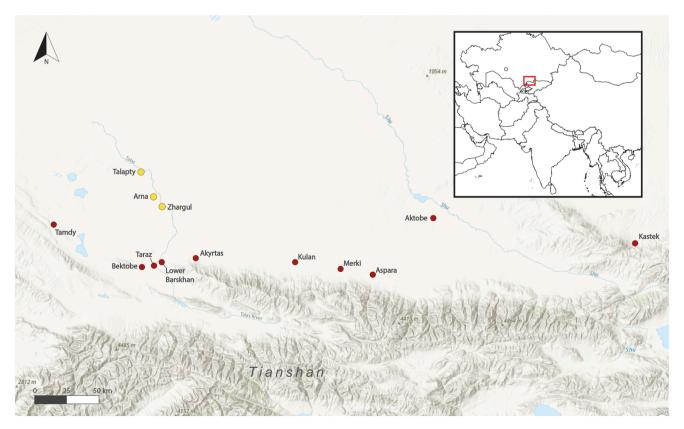


Fig. 1. Location of the archaeological sites Talapty, Arna, and Zhargul (indicated by a yellow circle) in the Talas River Valley in relation to sites where early Islamic glazed ceramics have previously been analyzed (indicated by a red circle). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

**Table 1**Radiocarbon date from the three tortkul sites under study (calibrated with OxCal 4.4 and the IntCal 20 curve).

Lab code	Site Name	Sample	<sup>14</sup> C age (BP)	σ	calibrated age (cal AD) 1σ – 68.2 % prob.	calibrated age (cal AD) 2σ – 95.4 % prob	Reference
FTMC-GR14-12	Zhargul-2	charcoal	895	26	1053-1085	1045-1220	unpublished
FTMC-GR14-13	Arna	charcoal	918	26	1046-1168	1037-1207	Akymbek et al., 2022
FTMC-GR14-14	Talapty	charcoal	1025	27	995-1028	905-1122	Akymbek and Shagirbayev, 2022

Baybosynov (Alipcheev and Baibosynov, 1982, pp. 187-188). The site is called "Tortkul - the medieval city" and it was assigned an occupational date from the 8th -12th centuries. In 2009, the expedition of the Code of Sites of the Archaeological Expertise LLP (led by Akymbek Y. and Kudabaev Sh.) carried out aerial photography and topographic plans (Akymbek, 2010, pp. 110-111). In 2021, the Talas team of the South Kazakhstan Complex Archaeological Expedition determined the current state of the site and carried out limited excavation work. Excavation of the interior structure revealed cultural levels beginning 0.3 m below ground level and extending to a depth of 1.1 m. Three small rooms were revealed, one completely, while two additional rooms were partially open. The structure was built of with pasha blocks (dimensions approximately  $60 \times 36 \times 20$ –22 cm) and mud bricks (dimensions approximately  $34-35 \times 18 \times 9$  and  $27 \times 14 \times 6$  cm), for an overall wall thickness ranging from 0.5 to 0.7 m. The floors were heavily trodden and plastered in places. Other archaeological features include a sufa, a bench made of rammed earth of bricks along a wall (Usami et al., 2017), which is preserved on the west side of one room. The results of radiocarbon analysis of burnt charcoal from the lowest cultural layer preserved in the excavation indicated that the structure dates from the  $10\text{-}11^{\text{th}}$  centuries (Akymbek and Shagirbayev, 2022). Archaeological material recovered from the excavation included both glazed and unglazed ceramics. Twenty-one glazed ceramic samples from two stratigraphic layers revealed in the excavation were selected for archaeometric analysis, including monochrome wares, sgraffito wares, and slip-painted wares (Fig. 2). The upper stratigraphic layer.

## 1.1.2. Arna

The site of Arna is located 7 km west of the village of Sarybarak. The tortkul lies 100 m east of the Karabakyr channel of the Talas River. The site is square in dimension, with the corners of the exterior walls oriented along with four directions (Fig. 3). The dimensions of the walls are  $96\times96$  m, the preserved height ranges from 1.5 to 2 m, the surrounding rampart is hardly visible, but there are traces of towers in the corners and in the middle of the walls. The wall towers are circular in shape and 10--15 m in diameter, and they retained a height above the wall of 0.2–0.4 m. The ruins of the external gate are located in the northeast wall, evidenced by a noticeable decrease in relief along this wall, suggesting the path was frequently trodden in antiquity by pedestrians and pack animals. The exterior walls are surrounded by ditches which have a preserved width of 10--12 m and a depth of 0.2–0.4 m.

The site was discovered in 2021 by the Talas team of the South Kazakhstan complex archaeological expedition. Excavations targeted the exterior wall rampart, which is preserved at a low height around the site. A narrow trench transecting the wall with a length of 15 m and a width of 2 m was laid out, and excavated to a depth of 1.5 m. Two construction horizons were identified, suggesting two distinct phases of building for the exterior defensive walls, both of which were made of mud bricks ( $40-42 \times 19-20 \times 10-12$  cm in dimension). Additionally, evidence of structures was recovered from the interior of the wall, as well as a hearth. Radiocarbon dating of material found in burnt charcoal recovered from the excavation dates the site to the  $11-12^{th}$  c. CE (Akymbek et al., 2022).

174 ceramic fragments were recovered from the 2021 excavations, the majority of which were from unglazed vessels (65 %), while the remaining 35 % came from glazed vessels. Detailed description of the

ceramic assemblages can be found in Akymbek et al. (2022). Twenty glazed ceramic samples from the two identified layers were selected for archaeometric analysis (Fig. 4), including monochrome wares, sgraffito wares, and slip-painted wares. The sherds were chosen to be representative of the larger assemblages as well as including samples that are unique and warrant further investigation.

#### 1.1.3. Zhargul

The site of Zhargul, located 4 km south of the village of Tugistik, lies between the confluence of the Saryozek and Kosheney channels of the Talas River, 100 m from the left bank of the Talas River. The complex consists of 3 distinct tortkuls (Fig. 5). The largest tortkul is Zhargul-1 which lies to the west, and immediately to the east are situated Zhargul-2 (in the North) and Zhargul-3 (in the South) along the same line.

The site was first discovered by the Kazakh branch of the Academy of Sciences of the Union of Soviet Socialist Republics and the expedition of the Institute for the History of Material Culture (led by A.N. Bernstam) in 1936 and described in detail (Bernshtam, 1949, pp. 108-110). In the register "Archaeological Map of Kazakhstan," published in 1960, under the number 3790, it is marked as "Fortified Settlements of Tegistik" and assigned an occupation period of the 8th-12th centuries (Arheologicheskaya karta Kazahstana (Archaeological map of Kazakhstan), 1960, p. 264). In 1978, the expedition of the historical and local history museum of the Zhambyl region (led by K. Baibosynov) described only two tortkuls and assigned them both a narrower occupational period ranging from the 11th-13th centuries (Alipcheev and Baibosynov, 1982, p. 172). This range of occupation was also listed for the site when it was resurveyed and described by The South Kazakhstan Complex Archaeological Expedition of the Institute of Archeology was named after A.Kh. Margulan of the Academy of Sciences of the Republic of Kazakhstan in 2000 (Baibosynov et al., 2002, pp. 120-121). The site was again survey in 2010, by the expedition of the Code of Sites of Archaeological Expertise LLP, the sites were described under the name "Tortkuls of Zhargul" and again identified with the 11<sup>th</sup>-13<sup>th</sup> centuries occupation date (Baibosynov et al., 2010, pp. 147-148).

In 2021, the survey team conducted site assessment and archaeological excavation of the three tortkuls. The walls of the tortkul Zhargul-1 are oriented to the four cardinal points. This tortkul was heavily damaged in Soviet and post-Soviet times due to cutting of channels from the Talas River and the agricultural and animal husbandry activities. The tortkul is almost square in shape (280  $\times$  270 m in dimension) and surrounded by a low rampart which is preserved to a height 2-3 m and width of 8-10 m. Traces of the tower are visible in the corners and along the walls, and placed at a distance of 20 m from each other. The towers have a diameter of 10 m and a height of 0.1-0.2 m. Evidence suggests that there were two main thoroughfares transecting the tortkul, with a gate in the middle of the four walls. The outer walls are surrounded by a ditch 8-14 m wide and 0.4-2 m deep. A trench that was 6.2 m long and 1 m wide was laid across the middle of the Zhargul-1. A cultural layer 2.5 m that was thick was identified, which consisted of several distinct construction horizons and indicated repeated use of the tortkul.

Tortkul Zhargul-2 It is located 140 m east of the tortkul Zhargul-1 (Fig. 5). The plan of the tortkul is square with the corners oriented towards the four cardinal points. The remains of towers are visible in the corners and in the middle of the walls. Dimensions of the wall  $90 \times 90$  m,



Fig. 2. Glazed ceramics from Talapty analyzed by NAA (K234-K254) and SEM-EDS (K234, K238, K240, K242, K247, K249, K250, K254). Samples K234-K244 were recovered from cultural layer 1, while samples K245-254 were identified from cultural layer 2.

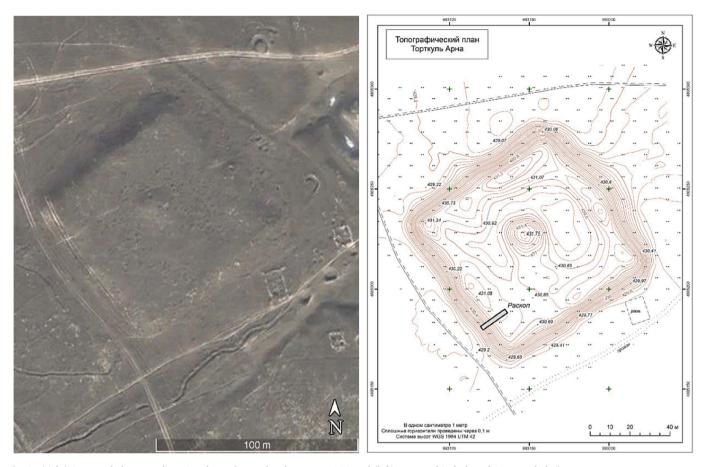


Fig. 3. (right) Arna tortkul as seen from Google Earth Pro, dated January 2016, and (left) topographical plan of Arna Tortkul. (). Adapted from Fig. 3Akymbek et al., 2022

height 0.5–1.2 m, shaft width 6–8 m, height 0.1–0.2 m. Ruins of towers have a diameter of 10–12 m and a height of 0.2–0.5 m. Excavations were carried out in the middle of Zhargul-2, close to the northern wall of the tortkul. The trench measured  $5\times4$  m (20 m²) and ran parallel to the tortkul walls, elongated in the east–west direction. Excavations at a depth of 1 m revealed the remains of the floor and structures made of mud bricks. From a depth of 0.35 m above the ground, the thick walls of the building, oriented from northeast to southwest, became clearly visible. A wall was found on the south side of the excavation, which has a thickness of 0.9 m and a preserved height of 0.4 m.

Tortkul Zhargul-3 is located 50 m south of the tortkul Zhargul-2 (Fig. 5). The north wall is parallel to the south wall of the Zhargul-2 tortkul. It is square and slightly larger than Zhargul-2, measuring 100  $\times$  100 m. The width of the surrounding wall is 10–12 m and has a preserved height of up to 2.5 m. Towers are preserved protruding from the walls at the corners, and have a diameter of 15–20 m, and are preserved at a height 0.5 m above the modern height of the exterior walls. In the middle of the eastern wall, there are traces of an  $\Gamma$ -shaped gate protruding outward. The exterior walls are surrounded by a ditch or moat with a width of 10–12 m, which reaches 1.5 m in depth in places.

Near the outer northwestern wall, in the middle of the Zhargul-3 tortkul, excavations of  $5 \times 4$  m ( $20 \text{ m}^2$ ) were carried out. Excavations were carried out in a direction from northeast to southwest, parallel to the walls of the site. The remains of two construction horizons made of pahsa and adobe brick were discovered. The upper construction horizon was recorded only from the northern corner of the excavation. Building remains were found at a depth of 0.2 m above the ground, and the floor was determined at a depth of 0.6–0.7 m. The thickness of the walls is 0.55–0.7 m, the preserved height is 0.5 m. The remains of the lower construction horizon were found at a depth of 0.7 m above the ground,

and the floor was found at a depth of 1– $1.1\,$ m. The remains of a wide wall were found, separating several rooms. Wall thickness ranges from 0.5 to 1 m, preserved heights: 0.2–0.5 m.

A total of thirty-three ceramics were analyzed from the Zhargul complex, eleven recovered from Zhargul-1, twelve recovered from Zhargul-2, and ten from Zhargul-3. The ceramics include a range of wares, included monochrome wares, including blue alkali-glazed ceramics, underglaze painted wares, and sgraffiato wares, including monochrome, splashed, and painted ceramics wares with sgraffiato decoration (Fig. 6).

## 2. Methods

## 2.1. Electron microscopy

Scanning electron microscopy—energy dispersive spectrometry (SEM-EDS) was performed on carbon coated embedded and polished ceramic cross sections using an FE-SEM Zeiss  $\Sigma$ igma HD in the Scientific Research Department at the Metropolitan Museum of Art. The SEM is equipped with an Oxford Instrument X-Max 80 SDD (Oxford, Tubney Woods, Abingdon, Oxon, UK). Back-scattered electron (BSE) imaging, as well as EDS elemental spot analysis and elemental mapping, were carried out with an accelerating voltage of 20 kV under high vacuum.

## 2.2. NAA

NAA was carried out at the Archaeometry Laboratory and the University of Missouri Research Reactor (MURR). Samples were prepared for NAA using procedures standard at MURR, which have been described in detail elsewhere (Glascock, 1992; Glascock and Neff, 2003).

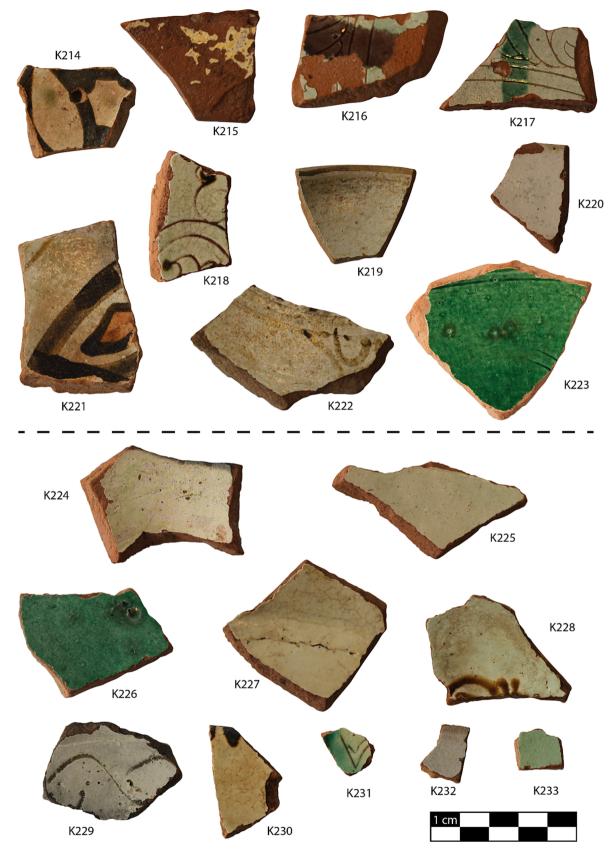


Fig. 4. Glazed ceramics from Arna analyzed by NAA (K214-K233) and SEM-EDS (K215, K216, K218, K221, K224, K231). Samples K214-K223 were recovered from the first cultural layer, while samples K224-233 were recovered from the second identified cultural layer.



Fig. 5. Location of the three tortkuls at the site of Zhargul. Google Earth©, 2021.

A small portion of the sherd ( $\sim 1 \text{ cm}^2$ ) was removed and a siliconcarbide burring tool was used to physically abrade the surface of the sherd to remove decorative features and other contaminants on the sample. The sample was then homogenized and ground to a fine powder using an agate mortar and pestle. Along with the unknown samples, standards from NIST certified SRM-1633b (coal fly ash) and SRM-688 (basalt rock) were similarly prepared, as were quality control samples (e.g., standards treated as unknowns) of SRM-278 (obsidian rock) and New Ohio Red Clay (a standard developed for in-house applications). Procedures used for the irradiation and gamma ray spectroscopy, consisting of two irradiations and a total of three gamma counts, were followed using established MURR Archaeometry Laboratory protocols (Glascock, 1992; Neff, 2000). A total of 33 element concentrations (Na, Al, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Zn, As, Rb, Sr, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Tb, Dy, Yb, Lu, Hf, Ta, Th, U) were determined. The data were analyzed using the statistical software Gauss 8.0.

#### 3. Results

## 3.1. NAA

74 samples were analyzed by NAA. Given the lack of Ni in most of the samples, it was excluded from analysis although it is reported in the summary statistics for the compositional groups. Five samples (K240, K247, K278, K284, and K286) behave as one group (TV-QTZ) which is distinct from the other 69 samples (Fig. 7). These ceramics have and have a distinct quartz body with an alkali-lime (n=4) or low lead-alkali glaze (n=1).

The remaining 69 samples have a clay based ceramic body which ranges in color from reddish yellow to brown. The composition of the ceramic fabrics is fairly consistent with 15.11  $\pm$  0.71 wt%  $Al_2O_3,\,6.29$   $\pm$  0.52 wt%  $Fe_2O_3,\,$  and 3.95  $\pm$  0.29 wt%  $K_2O.$  The most variation in the major elements comes from the amount of calcium present in the ceramic fabrics, which ranges from 2.54 to 10.14 wt% CaO, with an average of 6.42  $\pm$  2.04 wt% CaO. These ceramics have a high lead glaze, with the exception of K254 which has a unique type of low lead-alkali

glaze discussed below. Five compositional groups were distinguished for the clay bodied ceramics (Fig. 7, Table 2). Summary of the compositional groups the full compositional data is available in Supplementary Material 1.

A principal component analysis was performed on all the clay based ceramic fabrics (n = 69) for 32 elements. The first eight principal components describe 92 % of the total variance. Most defining elements for these ceramic fabrics are Sb, As, Ca, Sr, and Mn (Fig. 8). The distribution of the five lead glazed compositional groups in relation to the first two principal components (60.5 % of total variance) can be seen in Fig. 8, where principal component 1 enriched in Sr, Mn, Ca, Sb, and As and depleted in the rare earth elements, while principal component 2 is depleted in Sr, Ca, and U, and enriched in Sb, As.

Group TV1 is of six ceramics (K248, K269, K274, K277, K283, and K285) that were recovered from the site of Talapty (n = 1) and Zhargul (n = 5). It has elevated concentrations of Na and Hf, and relatively depleted concentrations of Ba, Sb, Fe, Ca. TV2 is a large group composed of 34 ceramics, which were recovered from all three sites, and all cultural layers within those sites. It is defined by high concentrations of Mn and Sb, the latter of which is a characteristic previously identified as being elevated in ceramics recovered from the Talas Valley. TV3 is a small group of three samples (K262, K273, and K275) which were recovered from the site of Zhargul and has relatively high concentrations of Ca in the ceramic fabric, as well as elevated levels of Rare Earth Elements (REEs). TV4 is composed of 11 ceramic samples from the sites of Arna and Talapty which is defined by low concentrations of Ca. TV5 is a small group of three samples (K222, K228, and K230), which were recovered from the site of Arna, and have a similar decoration with a white slip covered in a very faint brown/yellow decoration covered in a thin transparent glaze. They have relatively elevated levels of As compared with the other samples. Twelve ceramics, or 16 % of the assemblage analyzed by NAA, were either clear outliers or could not be confidently assigned to a compositional group.

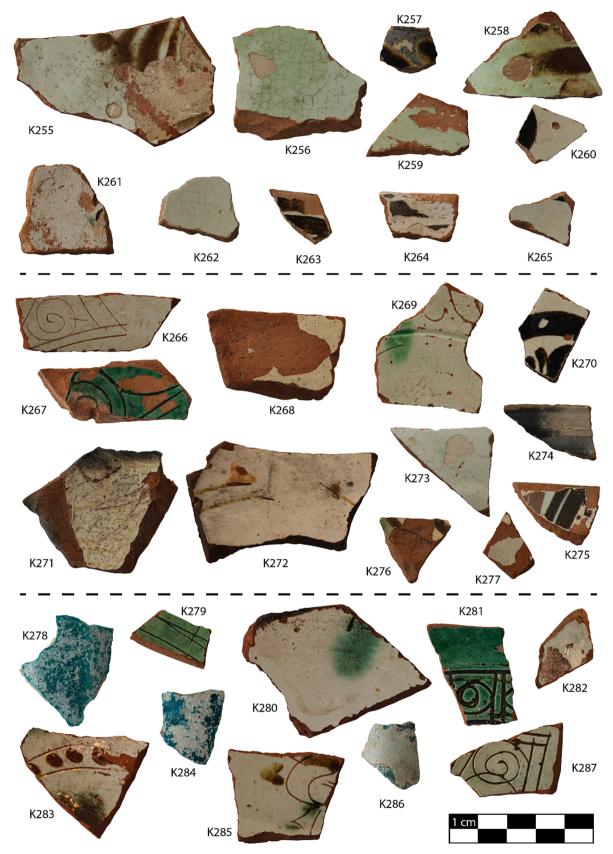


Fig. 6. Glazed ceramics from Zhargul analyzed by NAA (K255-K287) and SEM-EDS (K255, K257, K258, K267, K269, K272, K273, K278, K280, K281, K283, K284, and K286). Samples K255-K265 were recovered from Zhargul-1, samples K266-K277 were recovered from Zhargul-2, and Samples K278-K287 were recovered from Zhargul-3.

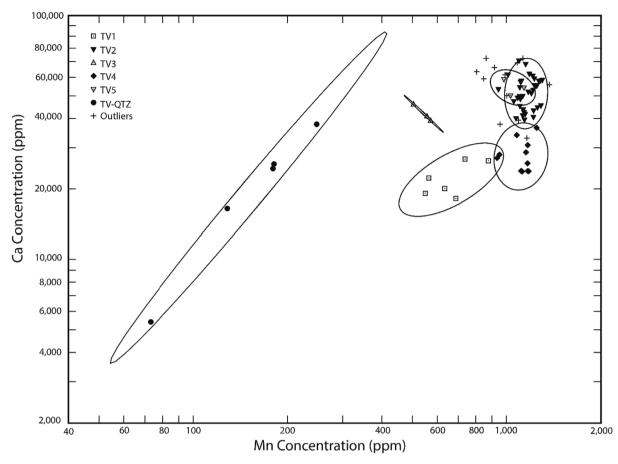


Fig. 7. A scatterplot of Mn versus Ca concentrations (in ppm − log base 10 scale) for the glazed ceramic groups TV1 ( $\square$ ), TV2 ( $\blacktriangledown$ ), TV3( $\triangle$ ), TV4 ( $\spadesuit$ ), TV5( $\nabla$ ), TV-QTZ( $\bullet$ ), and the outliers in the dataset (+). Confidence ellipses indicate 90 % confidence.

 Table 2

 Description of the ceramic compositional groups.

-				
Compositional Group	N=	Sites	Defining elements	Notes
TV-QTZ	5	Talapty, Zhargul		Quartz bodies
TV1	6	Talapty, <b>Zhargul</b>	High Na, Low Fe, Ca, Mn, As, Sb, Zn, Co	Some have high Cr
TV2	34	Arna, Talapty, Zhargul	High Mn, Sb	
TV3	3	Zhargul	High Ca, REEs, Co, Low Mn	
TV4	11	Arna, Talapty	Low Ca, High Zn, Mn, Sb	
TV5	3	Arna	High Sr, Mn, Sb, As	

## 3.2. Glaze chemistry

A subset of twenty-seven of the glazed ceramics were analyzed by SEM-EDS to determine their glaze type and chemistry. The subset was chosen to be representative of the larger assemblage and include the range of stylistic wares as well as colors present in the glaze and other decorative features such as sgraffito. Three types of glaze were observed in the data set: alkali-lime glaze (defined as < 2.0 wt% PbO), low lead-alkali glaze (defined as < 0.0 yt% PbO), and a high lead glaze (defined as < 0.0 yt% PbO) (Tite, 0.0 yt% PbO). Distribution of the glaze types can be seen in Fig. 9.

Four samples (K247, K278, K284, and K286) have an alkali-lime glaze with an average alkali (Na<sub>2</sub>O + K<sub>2</sub>O) concentration is 14.08  $\pm$  3.48 wt%, with a significant amount of CaO (5.32  $\pm$  1.21 wt%) and SiO<sub>2</sub>

(69.85  $\pm$  5.17 wt%) present. There four glazes are all blue in color and have a copper-based colorant, which has an average CuO concentration of 1.65  $\pm$  0.64 wt%. Compositionally they are fairly homogeneous, but do have air bubbles and quartz particles suspended in the glaze matrix (Fig. 10).

The four alkali-lime glazed ceramics as well as sample K240 have a quartz body style fabric, which contain >75 wt%  $SiO_2$ . The average  $SiO_2$  concentration in the fabric of these five ceramics is  $85.28\pm2.43$  wt %, with only  $3.25\pm1.32$  wt%  $Al_2O_3$  and  $3.08\pm1.49$  wt% CaO measured for these samples. This is in line with the observation by Tite (2011) who indicated that Islamic quartz bodies typically have >85 wt %  $SiO_2$ , <8 wt%  $Al_2O_3$  and <9 wt% CaO present. Prior research into Islamic glazes has concluded that the majority (>80 %) of alkali-lime glazes on Islamic ceramics are associated with a quartz body (Tite, 2011, p. 333).

Two samples (K240 and K254) have what is classified as a low lead-alkali glaze, although their compositions are quite distinct. K240 has a fairly typical composition of an Islamic low lead-alkali glaze, with a high alkali concentration (15.30 wt% Na $_2$ O + K $_2$ O), low alumina concentration (2.22  $\pm$  0.18 Al $_2$ O $_3$ ) and has a blue color imparted from 2.06  $\pm$  0.32 wt% CuO. It is opacified with tin particles (see Fig. 11) and has an average SnO $_2$  concentration of 0.72 wt% SnO $_2$ . As mentioned above, it also has a quartz body, which is common for Islamic low-lead alkali glazes. Tite (2011) reported in his synthesis of over 1200 Islamic glazed ceramics that were analyzed by the late Alexander Kaczmarczyk that 76% of the low lead–alkali glazes they studied had a quartz body.

K254, however, has a unique glaze that was not preserved across the entire surface of the ceramic fragment, but isolated in incised groves present on the surface. The composition is also unique with high concentrations of alumina (11.56 wt%  $\rm Al_2O_3$ ) and calcium (7.81 wt%  $\rm CaO$ )

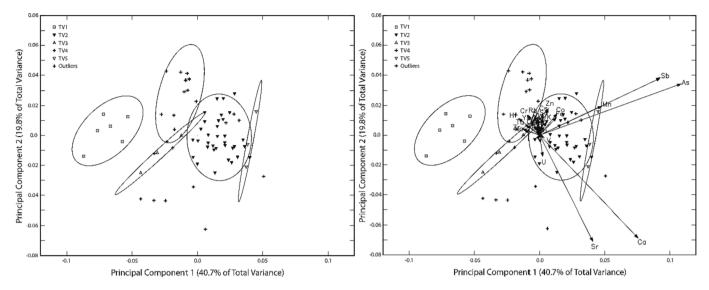


Fig. 8. Bivariate plot comparison of compositional data using principal component 1 (representing 40.7% of total variance) versus principal component 2 (19.8% of total variance) for the lead glazed ceramics measured by NAA. Confidence ellipses indicate 90% confidence interval for group centroid.

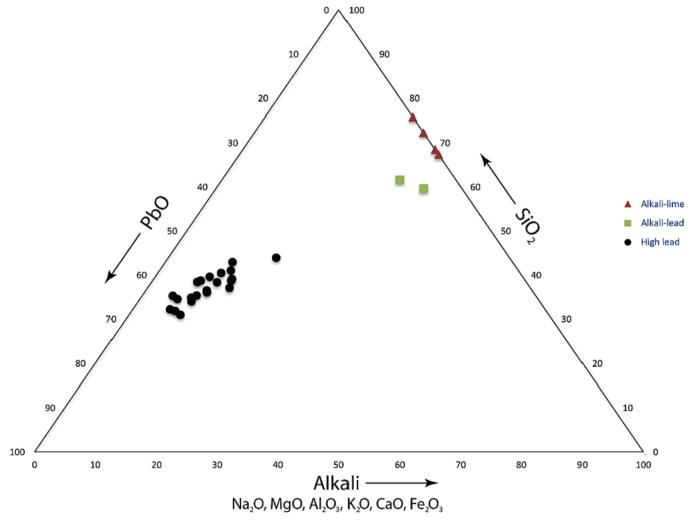


Fig. 9. Ternary diagram showing the major chemical compositions for the alkaline-lime glaze (♠), alkali-lead glaze (♠), and high-lead glazes (♠) for the 27 glazed ceramics analyzed by SEM-EDS. Concentrations are in oxide weight percent.

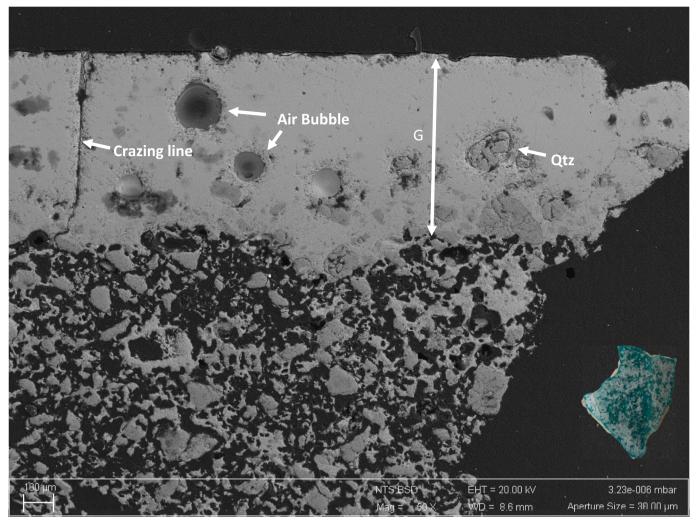


Fig. 10. Backscattered electron (BSE) image of the glaze-body cross section of the alkali lime glaze on sample K278. G = Glaze.

present with a relatively low alkali concentration (7.34 wt% Na $_2$ O + K $_2$ O). The glaze does have a lead flux (6.14 wt% PbO) and an iron and copper colorant (4.08 wt% Fe $_2$ O $_3$  and 1.76 wt% CuO). It is fairly heterogeneous in composition (see Fig. 12), with particles suspended throughout the matrix, including what appear to be prills of metallic copper. The sample has a standard clay body.

Twenty-one samples have a high lead glaze with an average PbO concentration of 52.33 wt%,  $Al_2O_3$  concentration of 4.85 wt%, silica concentration of 36.74 wt%  $SiO_2$ , and total alkali concentration ( $Na_2O$ + K<sub>2</sub>O) below 2 wt% (Table 3). This composition is in line with other early Islamic lead glazed ceramics from the eastern Islamic World as observed in southern Kazakhstan (Klesner et al., 2021a), the Fergana valley (Henshaw, 2010), and Termez (Molera et al., 2020). Similar compositions have also been recorded from early Islamic style glazed ceramics in Iraq and Syria (Shen, 2017). Most of the glazes appear to be made of a mixture of PbO and SiO<sub>2</sub> and not as a straight application of PbO. This is determined by comparing the ratio of silica in the slip and in the glaze (Fig. 13). The higher average amount of silica in the glaze compared to the underlying slip indicates that the potters were mixing SiO<sub>2</sub> with the lead oxide to create the glaze. All the transparent glazes are homogeneous with few undissolved quartz grains or other inclusions present. The high lead glazes were applied over an underlying white slip, and no appreciable particle growth was observed at the slip-glaze interface. In portions of the glaze where there was incision to the underlying ceramic fabric, extensive particle growth is present. The glaze is depleted in values of aluminum compared to the underlying ceramic slip

layers.

Mn, Fe, Cu, Cr, Sb, and Sb colorants and opacifiers are observed in this collection (Table 5). Five samples (K216, K221, K225, K257, K273) have Mn concentrations above 0.2 wt%, which imparts a brown/black color to the glaze. One of these samples (K257) also had very elevated concentrations of Fe<sub>2</sub>O<sub>3</sub> (9.50 wt%). Four samples are only elevated in Fe<sub>2</sub>O<sub>3</sub>. These include samples that have a red painted decoration (K272 and K283) and brown painted decoration (K218, K250, and also K272). The brown colored glazes all also have minimal amount of MnO present (average = 0.15 wt%). Seven of the samples with a high lead glaze have a copper green colorant, ranging from 0.53 to 3.48 wt% CuO (average = 1.76 wt% CuO). As previously mentioned, four of the alkali-glazed samples also have copper-based colorant, however, in these samples, the glaze is blue in color. Copper is able to impart a wide range of colors from blue to green to glazes depending on coordination and nature of the neighboring atoms in the glaze (Pradell and Molera, 2020). One sample (K218) had a chromium colorant 0.59 wt% Cr<sub>2</sub>O<sub>3</sub> which imparted a greenish brown color.

The four monochrome yellow glazed samples (K215, K224, K234, and K238) and one of the monochrome green sgraffito glazes (K269) had elevated concentrations of  $\mathrm{Sb_2O_3}$  (>0.1 wt%), with an average of 0.42 wt%  $\mathrm{Sb_2O_3}$  present in the glaze matrix. The four yellow glazes all have tin and antimony particles suspended in the matrix, ranging from 1:5 to 1:2 ratios of Sn:Sb concentration, respectively. The particles are submicron, and appear to have cubic and triagonal faces (see Fig. 14).

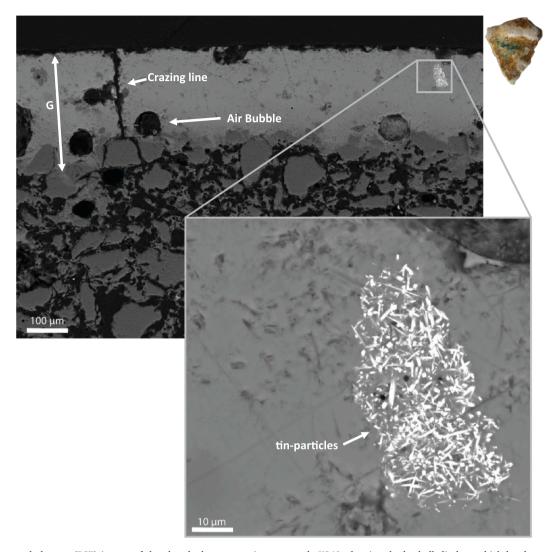


Fig. 11. Backscattered electron (BSE) images of the glaze-body cross section on sample K240, showing the lead-alkali glaze which has been opacified with tin-particles (appear white in BSE). G = Glaze.

#### 3.3. Slips

The high lead glazed ceramics have a white slip, also referred to as an engobe, which was applied over the ceramic, and under the glaze layer. Six samples (K250, K255, K257, K258, K272, and K273) additionally had a colored slip layer applied over the white slip (see Fig. 15).. The white slip is compositionally quite distinct from the underlying ceramic fabric (see Table 4), having very low concentrations of iron (average  $Fe_2O_3$  concentration = 0.72 wt%) and high concentration of  $SiO_2$  (68.28) wt%). As has been observed in other early Islamic glazed ceramics (Henshaw, 2010; Klesner et al., 2021a; Molera et al., 2020; Ting and Taxel, 2020), the white slip also has high concentrations of PbO (average  $11.56 \pm 4.30$  wt%). In these samples the PbO appears to be is distributed throughout the entire depth of the slip, with an accumulation of lead rich particles on the slip-body interface observed on many samples (see Fig. 16 for an example). The lead oxide appears to be intentionally added to the slip, and is not present in the slip solely as the result of lead diffusion during the firing process. This is demonstrated on samples K221, where the white slip which was under a high lead glaze on the interior of the vessel had similar concentrations of PbO (4.95-9.7 wt%) as the slip on the exterior of the vessel (9.0-10.23 wt%) which was never covered with a glaze. Several of the samples also demonstrated a double application of the same white slip on the interior of the vessel (see Fig. 16).

#### 4. Discussion

## 4.1. NAA groups

To determine potential production locations of the Talas Valley compositional paste groups identified above, the chemical compositions of the pastes in this study were compared to prior NAA of early Islamic glazed ceramics from the Talas and Chu River Valleys in Kazakhstan (Klesner et al., 2021a, 2019). On the whole, all of the high lead glazed ceramics from Arna, Talapty and Zhargul are similar compositionally to only two previously identified compositional groups: KAZ Groups 3 and 14 (see Fig. 17). This was determined by examining scatterplots of element compositions as well as calculating group classification using Mahalanobis distance (see Supplementary Material).

This group classification was calculated using the first seven Principal components (representing 89 % of total variance) for the prior identified groups. The best group membership for the 69 pastes was determined based on the highest membership probability greater than  $10\ \%$ .

TV2 falls within the previously identified compositional Group 3 (Fig. 18). Group 3 is a large compositional group (n = 43) which was defined by having high concentrations of Fe, Mn, Co, and Cr (Klesner, 2021). Group 3 is composed of ceramics from eight sites in the Talas and Chu Valley, including the sites of Tamdy, Lower Barskhan, Bektobe,

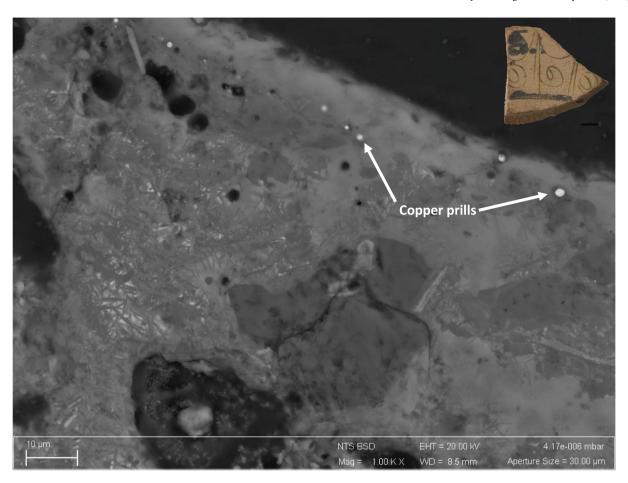


Fig. 12. Backscattered electron (BSE) image of the heterogeneous lead-alkali glaze on sample K254. The circular white inclusions visible near the surface of the glaze are metallic copper prills.

Table 3 Mean (m) and standard deviation ( $\sigma$ ) of glaze compositions in this study by glaze type as determined by EDS.

Oxide %	(n = 6)	reas on 4	Low lea glaze (n = 2)	d-alkali	High lead glaze (n = 36 distinct gi areas on 21 sherds	
	m	σ	K254	K240	m	σ
Na <sub>2</sub> O	10.19	3.60	1.53	12.41	0.29	0.23
MgO	3.38	0.51	2.43	3.12	0.65	0.16
$Al_2O_3$	3.37	0.99	11.56	2.22	4.85	1.89
$SiO_2$	69.85	5.17	58.05	59.21	36.74	4.07
$P_2O_5$	0.43	0.11	0.19	0.50	0.14	0.05
$SO_3$	0.34	0.09	0.13	b.d.		
K <sub>2</sub> O	3.88	1.01	5.81	2.90	1.11	0.84
CaO	5.32	1.21	7.81	6.73	1.41	0.53
$TiO_2$	0.14	0.03	0.47	0.11	0.19	0.06
$Cr_2O_3$				b.d.		
MnO			0.09	b.d.	0.40 (n = 12)	0.45
$Fe_2O_3$	1.06	0.29	4.08	0.66	1.69	2.13
CuO	1.65	0.64	1.76	2.06	0.64	0.88
$SnO_2$				0.72		
$Sb_2O_3$					0.42 (n = 5)	0.20
ZnO				b.d.	0.57	0.79
PbO			6.14	8.88	52.33	6.33
TOTAL	99.63	0.16	99.98	99.51	100.00	0.00
$K_2O+Na_2O$	14.08	3.48	7.35	15.30	1.40	1.06

Taraz, and Akyrtas in the Talas River Valley (80 % of the compositional group). Given the fact that almost half of the high lead glazed ceramics from Arna, Talapty, and Zhargul fall within this compositional group,

compiled with the high percentage of previous ceramics recovered from the Talas River Valley, it is tempting to assign a production location for these ceramics somewhere within the Talas River Valley. While pottery kilns have been excavated in the city of Taraz (Yeleuov et al., 2014), there has not been any glazed ceramics from a production site in the Talas River Valley characterized by compositional analysis to date. Until such work is done, any assignment to the Talas River Valley remains tentative.

TV3 falls within the previously identified compositional Group 14, which is defined by having low concentrations of Mn and Sb, and elevated concentration of Fe, Co, and Cr. Group 14 is one of two compositional groups associated with local production at the site of Aktobe in the Chu River Valley (Klesner, 2021). Group 14 was identified as produced at Aktobe as it includes sherds that were recovered from a kiln approximately 500 m from the citadel at the site. However, unlike the other compositional group associated with Aktobe production, ceramics in Group 14 were recovered beyond the confines of the Aktobe site, and included samples which were recovered from the sites of Bektobe and Lower Barskhan in the Talas River Valley, and Kulan, which lies in the western portion of the Chu River Valley (Klesner, 2021). Thus, the identification of the TV3 group ceramics from Zhargul in the Talas Valley belonging to this compositional group further supports the conclusion that there was regional trade of glazed ceramics produced at the site of Aktobe in the Chu and Talas River valleys.

Group TV1, TV4, and TV5 appear to be unique groups unrelated to any previously analyzed materials. While TV1 does have some compositional similarity to TV3/Group 14, it has markedly higher concentrations of Na and lower concentrations of Ca, Ba, Co, Sb than TV3/Group 14 and is considered compositionally distinct. Three of the four

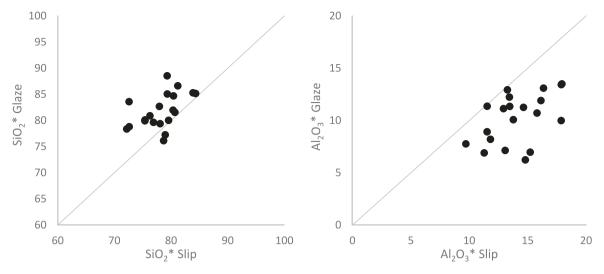


Fig. 13. Comparison between the normalized concentration (reported in ox. wt%) of  $SiO_2^*$  (left) and  $Al_2O_3^*$  (right) in the glaze and the slip of the high lead glazed samples. The total compositional values are normalized to 100% after subtraction PbO and colorants from the composition.

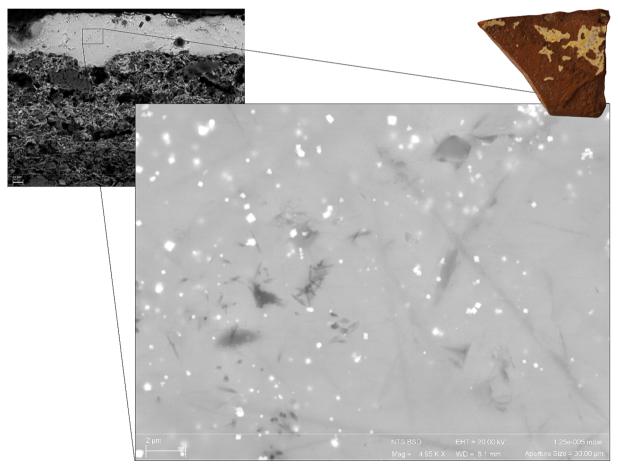


Fig. 14. SEM backscatter image of a cross section of the yellow glaze on K215 showing body, underlying slip, and glaze layer, and at higher magnification the distribution of the sub-micron particles containing Sb and Sn oxides (appear white in BSE).

monochrome yellow glazed samples with the antinomy and tin particles (K215, K234, and K238) fall within TV4, and that group has a great deal of compositional and technological similarities to TV2/Group 3. It could have been produced in the same workshop using slightly different raw materials, or by the same people in a different location, but more research is needed to confidently determine the production location of this compositional group.

Future research will attempt to identify potential raw materials in the Talas and Chu River Valleys to help assign production locations for the defined compositional groups TV1, TV2, TV4, and TV5. All of the provenance research to date has relied only on creation of compositional reference groups, tied to specific production centers only when products from kiln sites have fallen within that group. While the process of preparing clay and aplastic materials for the production of ceramic pastes

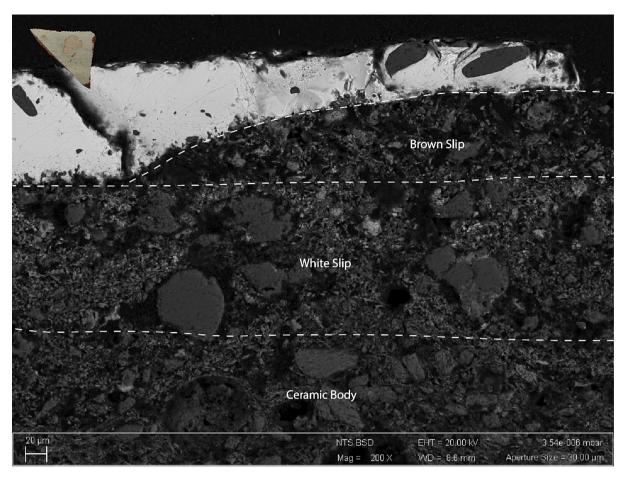


Fig. 15. SEM backscatter image of the glaze-slip-body interface for the brown underglaze painted area on sample K273.

Table 4 Average (m), standard deviation ( $\sigma$ ), and normalized average compositions of the slips (n = 19) underlying the high lead glazes as determined by EDS. The low totals are indicative of porous glazes.

Oxide %	Slips under high le	ad glaze
	m	σ
Na <sub>2</sub> O	0.74	0.26
MgO	0.77	0.31
$Al_2O_3$	12.31	2.03
$SiO_2$	68.28	4.85
$P_2O_5$	0.31	0.16
$SO_3$	0.54	0.51
K <sub>2</sub> O	3.65	0.90
CaO	0.72	0.36
TiO <sub>2</sub>	0.41	0.19
MnO		
Fe <sub>2</sub> O <sub>3</sub>	0.72	0.23
CuO	0.14	0.01
$SnO_2$		
PbO	11.56	4.30
TOTAL	99.53	0.44

often impedes the direct comparison of ceramic compositional data to raw material sources, the results of raw material studies can provide information about potential sources in the vicinity of the sites (Gliozzo, 2020; Hein and Kilikoglou, 2020). Given the relative absence of excavated ceramic workshops in the region to compare against, the systematic survey and sampling of potential clay sources poses the best avenue for further refining their potential production locations in the Talas River Valley.

## 4.2. Decoration

The transparent high lead glazed ceramics are very similar in both their composition and microstructure to previously characterized ceramics from the eastern Islamic World. This includes ceramics from the Talas and Chu River Valleys (Klesner, 2021), the Ferghana Valley (Henshaw, 2010), and Termez (Martínez Ferreras et al., 2019; Molera et al., 2020) in the heart of Central Asia, as well as ceramics from further afield including wares from Nishapur (Bouquillon et al., 2013). We see the use of a common glaze recipe, which appears to have been made by prefritting lead oxide and silica (Fig. 13), a common and systematic use of a white slip which had added lead oxide (Table 4), and common metal oxides employed in the same way in the decoration of the early Islamic style wares. There is a high level of standardization and specialization employed in the production of these ceramics, and these indicators together suggest that there was a common technological package known for the production of transparent high lead glazes, and it was introduced into the region by skilled craftspeople.

The relatively high proportion of ceramics which have either monochrome or incised decoration is typical for ceramics from the region in  $11^{th}$  and  $12^{th}$  c. CE. During that time there is an observed shift.

in the preferred ceramic's styles, with simpler monochrome glazing and incised styles being favored in Central Asia (Henshaw, 2010). In this time, there were two primary incised styles: monochrome green transparent glaze over a white slip with incision, or green, brown, and yellow "splashes" in the transparent glaze over the white slip with incision (Henshaw, 2010). The "splashed sgraffito ware" is thought to be stylistically connected to the imported Tang *sancai* ceramics (Shen, 2017), although it has also been argued that it was independently developed in the Islamic world (Watson, 2004, pp. 40–41, 47–48). Both of these styles

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Table 5a
Properties of the high lead glazed ceramics from Arna, Talapty, and Zhargul in the Talas River Valley. Composition it reported in oxide weight %.

ANID	Site	Layer	Ware	Glaze type	Glaze color	colorant/opacifier	n=	Na <sub>2</sub> O	MgO	$Al_2O_3$	$SiO_2$	$P_2O_5$	$SO_3$	K <sub>2</sub> O	CaO	$TiO_2$	$Cr_2O_3$	MnO	$Fe_2O_3$	CuO	$SnO_2$	$Sb_2O_3$	ZnO	PbO	TOTAL
K215	Arna	1	M	high Pb	Y	Sb, Sn	5	0.26	0.92	6.30	42.46	0.18	-	0.92	1.79	0.19	_	_	0.75	_	part.	0.62	-	45.59	100.00
K216	Arna	1	SSG	high Pb	Br	Mn	5	0.12	0.61	2.11	27.19	0.11	-	0.48	2.50	0.17	-	0.66	0.64	0.25	-	-	-	65.16	100.00
					T	-	5	0.20	0.78	4.44	34.05	0.15	-	1.03	3.20	0.15	-	_	0.66	0.55	-	-	0.11	54.78	100.00
K218	Arna	1	SSG	high Pb	Br	Fe	5	0.212	0.69	3.44	35.12	0.17	-	0.63	1.48	0.14	-	0.15	8.80	0.10	-	-	-	49.93	100.00
					G/Br	Cr	5	0.18	0.91	4.342	36.86	0.24	-	0.62	1.27	0.16	0.59	_	1.14	0.16	-	-	-	53.66	100.00
					T	-	5	0.24	0.63	3.136	39.42	0.16	-	0.63	1.20	0.11	_	-	0.64	0.08	-	-	-	53.85	100.00
K221	Arna	1	UG	high Pb	Br	Mn	5	0.31	0.56	4.72	35.92	0.11	-	1.02	1.10	0.20	_	0.22	2.60	0.09	-	-	-	53.38	100.00
					T	-	5	0.38	0.36	3.05	33.49	0.11	-	0.87	0.98	0.11	-	_	0.58	0.11	-	-	-	59.98	100.00
K224	Arna	2	M	high Pb	Y	Sb, Sn	5	0.18	0.69	4.57	38.32	0.26	-	0.79	0.84	0.20	-	0.07	0.82	0.12	0.21	0.13	-	52.85	100.00
K231	Arna	2	SSG?	high Pb	Y/G	Cu	5	0.15	0.51	3.06	31.63	0.13 (n=3)	-	0.61	1.15	0.13	-	_	0.59	1.40	-	-	-	60.63	100.00
K234	Talapty	1	M	high Pb	Y	Sb, Sn	5	0.38	0.63	6.98	40.53	0.13	-	1.76	1.04	0.28	-	_	0.81	0.34	part.	0.42	-	46.71	100.00
K238	Talapty	1	M	high Pb	Y	Sb, Sn	5	0.41	0.60	6.45	37.84	0.11	_	1.50	0.95	0.17	_	-	0.76	0.47	part	0.33	-	50.26	100.00
K242	Talapty	1	SG	high Pb	G	Cu	5	0.40	0.86	5.16	34.91	0.14	b.d.	0.99	1.45	0.21	b.d.	b.d.	1.12	1.10	-	_	1.76	51.98	100.00
K249	Talapty	2	SG-UG	high Pb	T	_	5	0.22	0.63	5.47	39.30	0.18	b.d.	1.04	1.13	0.22	b.d.	b.d.	0.40	0.25	-	-	b.d.	51.09	100.00
K250	Talapty	2	UG	high Pb	Br	Fe	5	0.17	0.59	3.03	34.42	0.11	b.d.	0.71	1.24	0.16	b.d.	0.18	5.00	b.d.	-	_	b.d.	54.87	100.00
					O	Fe?	4	0.10	0.81	4.42	36.62	0.21	b.d.	0.60	1.79	0.13	b.d.	0.09	2.15	b.d.	-	_	b.d.	53.17	100.00
					T	_	3	0.12	0.67	5.19	38.22	0.14	b.d.	0.76	1.39	0.15	b.d.	b.d.	1.28	b.d.	-	_	b.d.	52.15	100.00
K255	Zhargul	1	UG	high Pb	Br	Mn	5	0.18	0.33	3.26	31.22	b.d.	b.d.	0.59	1.04	b.d.	b.d.	0.47	1.96	0.08	-	_	b.d.	61.10	100.00
					R/Br		5	0.09	0.36	3.46	32.11	0.05	b.d.	0.55	1.20	0.15	b.d.	0.14	1.28	b.d.	-	_	b.d.	60.64	100.00
K257	Zhargul	1	UG	high Pb	Br (dark)	Mn, Fe	5	1.02	0.70	5.49	38.29	0.15	b.d.	3.36	2.42	0.32	b.d.	1.52	9.50	0.36	_	_	0.25	37.70	100.00
					Br (light)	_	5	1.20	0.52	3.79	48.55	0.11	b.d.	4.84	1.67	0.28	b.d.	b.d.	0.62	0.21	-	-	b.d.	38.08	100.00
K258	Zhargul	1	UG	high Pb	Br	Fe (low)	3	0.38	0.96	5.94	40.60	0.13	b.d.	1.15	1.94	0.27	b.d.	b.d.	2.03	0.22	_	_	b.d.	46.38	100.00
					G	Cu (low)	5	0.24	0.62	3.56	36.10	0.09 (n=3)	b.d.	0.71	1.24	0.18	b.d.	b.d.	0.61	0.53	_	_	b.d.	56.03	100.00
					T		5	0.51	0.56	7.12	43.74	0.09	b.d.	1.57	1.38	0.16	b.d.	b.d.	0.61	0.19	-	_	b.d.	44.02	100.00
K267	Zhargul	2	SG	high Pb	G	Cu	5	0.10	0.54	4.66	32.85	0.08	_	1.09	1.60	0.20	_	_	0.52	3.03	-	_	_	55.37	100.00
K269	Zhargul	2	SSG	high Pb	G	Sb, Cu	5	0.15	0.74	2.83	33.79	0.16	_	0.64	1.27	0.15	_	_	0.41	1.34	-	0.58	_	58.01	100.00
K272	Zhargul	2	UG	high Pb	Br	Fe	5	0.12	0.75	4.52	38.28	0.17 (n=4)	b.d.	0.72	1.10	0.15	b.d.	0.11 (n=3)	3.42	b.d.	_	_	b.d.	51.00	100.00
					R	Fe	5	0.23	0.74	11.93	36.34	0.16 (n=3)	b.d.	1.74	1.37	0.23	b.d.	0.08	3.11	0.32	-	_	b.d.	44.03	100.00
					T		3	0.15	0.76	5.32	41.10	0.13	b.d.	0.79	1.35	0.12	b.d.	b.d.	0.65	b.d.	_	_	b.d.	49.61	100.00
K273	Zhargul	2	UG	high Pb	T		5	0.17	0.38	2.64	33.59	b.d.	b.d.	0.50	0.66	b.d.	b.d.	b.d.	0.28	0.15	_	_	b.d.	61.51	100.00
	-			_	Br	Mn	3	0.18	0.47	2.90	36.44	b.d.	b.d.	0.62	0.75	b.d.	b.d.	0.83	0.47	b.d.	_	_	b.d.	57.17	100.00
					В	Cu	3	6.24	3.66	4.95	71.63	0.47	0.27	4.33	6.10	0.19	_	b.d.	0.81	1.12	_	_	b.d.	b.d.	99.78
K280	Zhargul	3	SSG	high Pb	G	Cu	5	0.23	0.54	4.87	32.04	0.10 (n=4)	b.d.	0.88	1.37	0.15	b.d.	b.d.	0.45	1.46	_	_	b.d.	57.89	100.00
	3.			Ü	T		5	0.25	0.62		37.78					0.22		b.d.	0.46	0.18	_	_	0.17		100.00
K281	Zhargul	3	SG	high Pb	G	Cu	5	0.44	0.80	6.13	35.63					0.29		b.d.	1.79	3.48	_	_	b.d.	47.60	100.00
K283	Zhargul			high Pb	R	Fe	5	0.28		7.82	35.91					0.36		b.d.	3.37	0.18 (n=3)	_	_	b.d.		100.00
	3.			Ü	T		5	0.35	0.54	6.79	41.97					0.20		b.d.	0.55	0.16 (n=3)	_	_	b.d.	47.36	
					В	Cu	5	12.85			68.74					0.11		_	0.99	1.83	_	_	b.d.	b.d.	99.50

M = Monochrome, SG= Sgraffito, SSG = Splashed Sgraffito. UG = underglaze painted.

Op. = opaque, Y = Yellow, G = Green, Br = Brown, O=orange, R = Red, T = transparent glaze, b.d. = below detection, part. = detected in particles in the glaze matrix. A dash (-) indicates the element was not analyzed.

Properties of the high lead glazed ceramics from Arna, Talapty, and Zhargul in the Talas River Valley. Composition it reported in oxide weight

	Site	Layer	Ware	Glaze type	Glaze color	NID Site Layer Ware Glaze type Glaze color colorant/opacifier	=u	Na <sub>2</sub> O I	MgO	$Al_2O_3$	${ m SiO}_2$	$P_2O_5$	$SO_3$	$K_2O$	CaO I	$iO_2$	$Cr_2O_3$ MnO $Fe_2O_3$	MnO		CuO	$SnO_2$	$\mathrm{Sb}_2\mathrm{O}_3$	ZnO	Pbo	TOTAL
K240	Talapty 1	1	M	low lead-alkali	В	Sn, Cu	5	12.41	3.12	2.22	59.21	0.50	b.d.	2.90	6.73	0.11	b.d.	b.d.	99.0	2.06	0.72	1	b.d.	8.88	99.51
K247	Talapty	2	M	alkali-lime	В	Cu	2	11.38	3.17	3.38	66.05	0.58	0.32	5.29	5.71	0.15			1.60	2.13	1	1	p.d.	b.d.	9.75
K254	Talapty	2	٠.	low lead-alkali	Br	Fe, Cu?	2	1.53	2.43	11.56	58.05	0.19	0.13	5.81	7.81	0.47	1	0.09	4.08	1.76	1	1	1	6.14	86.66
K278	Zhargul	3	M	alkali-lime	В	Cu	2	7.65	3.88	4.09	68.71	0.49	0.28	4.62	69.9	0.12	1	0.00	0.88	2.15	1	1		-	99.66
					В	Cu	3	6.24	3.66	4.95	71.63	0.47	0.27	4.33	6.10			b.d.	0.81		1	1	p.d.	b.d. 9	82.6
K284	Zhargul 3	3	M	alkali-lime	В		2	7.59	2.49	2.29	79.20	0.27	0.27	2.74	3.21 (	0.13	ı	ı	0.92	0.59	1	1	b.d.	b.d.	9.72
					В	Cu	2	12.85	3.33	2.89	68.74	0.34	0.41	3.12	4.88	0.11	1	ı	66.0	1.83	1	1		Ī	9.50
K286	Zhargul 3	3	М	alkali-lime	В	Cu	4	15.45	3.73	2.63	64.75	0.44	0.48		5.34 (	0.15		ı	1.17		b.d.	1		b.d. 9	9:36

= opaque, Y = Yellow, G = Green, Br = Brown, O=orange, R = Red, T = transparent glaze, b.d. = below detection, part.= detected in particles in the glaze matrix. A dash (-) indicates the element was not analyzed. M=Monochrome, SG=Sgraffito, SSG=Splashed Sgraffito. UG=underglaze painted.

are seen in a high proportion in the assemblages from Arna, Talapty, and Zhargul, and together with the alkali-glazed ceramics, suggest an 11- $12^{\rm th}$  c. CE occupational usage of the sites.

## 4.3. Colorants

Glazes can be transparent and colorless, made opaque due to the presence of crystalline materials or air bubbles suspended in the matrix (Matin et al., 2018; McCarthy, 1996), or colored by the present of transition metal cations (e.g. Cu, Co, Cr, Mn, Fe) to produce a range of visual finishes (Gulmini et al., 2006; Molera et al., 2001; Pradell and Molera, 2020). The Talas Valley glazed ceramics display both of the main ways in which glazes were colored in early Islamic period. The first method of introducing color was achieved by applying the colored decoration as a paint or slip-paint on an unglazed ceramic and then coating it with a transparent lead glaze. Both the paints and the slippaints were colored using metal oxides (Co, Cr, Mn, Fe) and this was the method favored for decoration in the eastern Islamic World (Watson, 2020). In this study only black, brown, and red colors were created this way, and were achieved by using Mn, Fe, and Cr based colorants. The use of iron and manganese does not seem to be isolated to a single compositional group or production center – instead the ceramics with these colorants are widely dispersed across sites and within compositional groups.

The only sample with the chromium colorant ( $Cr_2O_3 > 0.1$  wt%) was K218, which fell within Group TV2/ KAZ Group 3. Chromium based colorants were previously identified within this group although not in the groups which were identified as being produced in the Chu River Valley, suggesting that it is a colorant that is limited in its application.

The other primary method of achieving colored decoration is by having the colorant in solution, which we can see in both the blue and green monochrome glazes in this collection. Similar to Fe and Mn, the use of copper to create a green monochrome glaze (K242, K267, K281) and green splashes on the splashed sgraffito wares (K231, K269, K280) does not seem to be isolated to one compositional group. This is expected, as copper green is one of the most common colorants used across lead glazed wares, including early Islamic glazed wares in Central Asia (Henshaw, 2010). The use of copper to create a monochrome blue/turquoise color is isolated to the ceramics in the TV-QTZ group, because copper is only able to impart a blue color in glazes with low lead concentrations.

## 4.4. Opacifiers

One sample (K240) was opacified with tin-oxide, which was the predominant opacifier used in early Islamic glazed ceramics (Matin et al., 2018; Ting and Taxel, 2020; Tite et al., 2015; Watson, 2014). The majority of the opaque ceramics in this study, however, had a yellow antimony- and tin-opacified glaze. Antimony- and tin-opacified yellow glazes have previously been identified in ceramics from the Talas and neighboring Chu River Valleys in Kazakhstan (Klesner et al., 2021a, 2021b), and the concentration of Sb<sub>2</sub>O<sub>3</sub> is in line with other early Islamic antimony-opacified glazes (Salinas et al., 2019). The opacifying particles, which have consistent ratios of Sn:Sb within single sherds, indicate that they were likely added together as a single component into the glaze mixture to serve as a colorant and opacifier. The cooccurrence of the two opacifying agents does not appear to be from application of multiple glaze layers with distinct opacifying agents, as was seen in early Islamic period Egypt (Salinas et al., 2019), instead, the potters are adding them together as a single opacifying agent. XRD of the antimony- and tinopacified yellow glaze on K212 (Klesner et al., 2021b) indicated that the particles were a form of lead-tin-antimony pyrochlore (see Fig. 19), although it is not known whether the particles were added in that form or they formed during the firing process.

The use of antimony- and tin-opacifiers in glazes in this region has been hypothesised to be the result of relative availability of antimony in

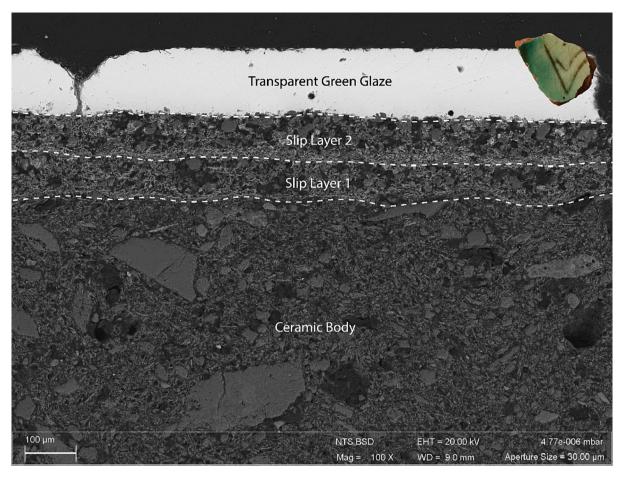


Fig. 16. SEM backscatter image of the glaze-slip-body interface for the green glazed are on sample K231. Two distinct applications of the white slip can be seen under the transparent glaze. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

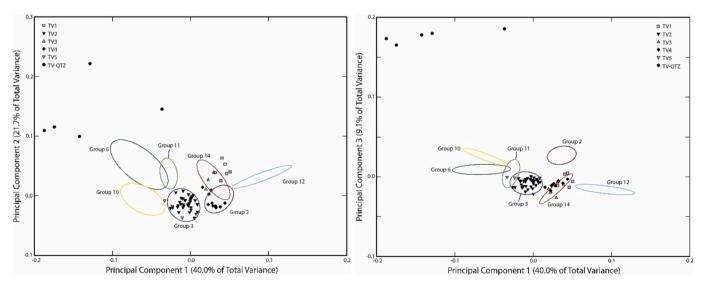


Fig. 17. Bivariate plot of Principal Component 1 (representing 40.0% of variance) versus Principal Component 2 (21.7% of variance) for seven comparative compositional groups from southern Kazakhstan (from Klesner (2021), groups represented by ellipses), compared to the NAA data for the glazed ceramics from the three tortkuls under study. Confidence ellipses indicate 90% confidence interval for group centroid.

the region. The Talas silver ore district, which was a known silver production site in antiquity (Bubnova, 1963; Pavlova and Borisenko, 2009), contains high-Sb sulfosalts, Sb sulfides, and native antimony which could have been sourced to be used as a glaze opacifier during the process of silver mining (Klesner et al., 2021a). It has also been

suggested that this was the antimony- and tin- opacifier was more directly linked to silver metallurgical activities, and potentially was obtain in the form of antimony- and tin-rich dross which this a byproduct of silver cupellation (Klesner et al., 2021b). The identification of this unique opacifier in the ceramics from Arna and Talapty further

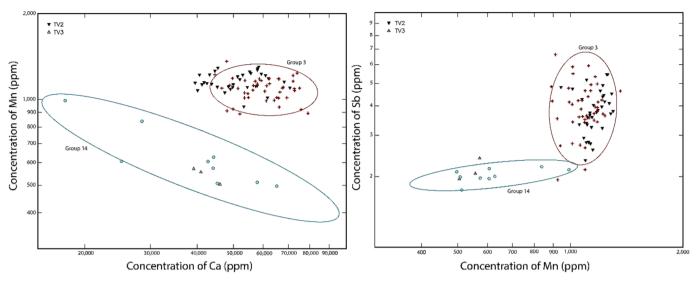


Fig. 18. A bivariate plot of (left) Ca versus Mn concentrations and (right) Mn versus Sb (in ppm – log base 10 scale) for the bodies of the glazed ceramics in: TV2 (), TV3(△), and Group 3 (+) and Group 14 (○). Ellipses represent 90 % confidence interval for group centroid for members belonging to Groups 3 and 14.

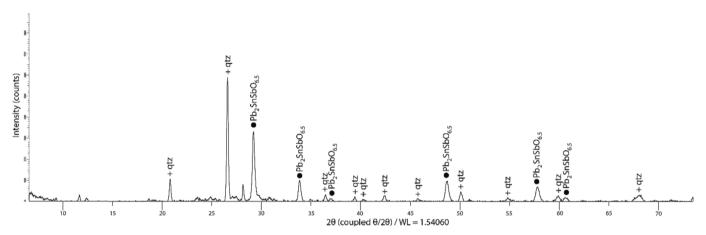


Fig. 19. XRD spectra of sample of the yellow opaque glaze particles on samples K212 from Aktobe, compositional group 14. Quartz (qtz) and a lean-tin-antimony pyrochlore (Pb<sub>2</sub>SnSb0<sub>6.5</sub>) are dominant phases in the spectra.

confirms that the local craftspeople in southern Kazakhstan were producing a unique opaque glazed ceramic using local raw materials.

## 5. Conclusions

Six distinct compositional groups, one of which is formed by the quartz bodied ceramics, and five of which have clay bodies with highlead glazes were identified in the ceramics from Arna, Talapty, and Zhargul. Most of these groups were found at multiple sites, suggesting there was an active circulation of glazed ceramics in the area, and a diversity of production centers in the larger region. The results suggest that these tortkul sites were integrated into the larger regional circulation of goods, including local craft products, and were not isolated centers.

Two of the compositional groups fall within previously identified compositional groups from southern Kazakhstan. One of those is a large compositional group that is found at many sites across the Talas and Chu River Valleys, although predominantly composed of ceramics recovered from the Talas Valley. The fact that almost half of the ceramics in this study fall within this compositional group further strengthens the hypothesis that this group was produced somewhere within the Talas River Valley, potentially in the city of Taraz. However, this identification remains tentative pending further research. The other group was produced

at the site of Aktobe in the Chu River Valley, and the fact that ceramics belonging to this group were recovered from the Zhargul tortkuls supports the conclusion that there was regional trade of glazed ceramics between the Chu and Talas River Valleys.

The microscopic and compositional analysis of the glazed wares identified several distinct glaze types including transparent high-lead glaze, opaque high-lead glaze, two distinct low-lead alkali glazes, and alkali-lime glaze. The majority of the glazed ceramics from the Talas Valley have a transparent high-lead glaze, which have a very uniform composition and similar to other Central Asia sites in the Early Islamic period. The characterization of the production techniques and use of colorants and opacifiers further strengthens are understanding of the technological traditions that were employed in the production of the early Islamic style wares in this region. The identification of the antimony and tin opacifier in the ceramics from Arna and Talapty indicates that the local craftspeople in piedmont region north of the Tianshan were producing a unique opaque glazed ceramic using local raw materials.

## CRediT authorship contribution statement

**Catherine Klesner:** Conceptualization, Methodology, Formal analysis, Writing – original draft. **Yeraly Akymbek:** Conceptualization,

Funding acquisition. **Brandi L. MacDonald:** Formal analysis, Funding acquisition.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2023.104339.

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