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1 **On-site pXRF analysis of glaze composition and colouring agents of “Iznik” tiles at Edirne**  
2 **mosques (15th and 16th-centuries)**

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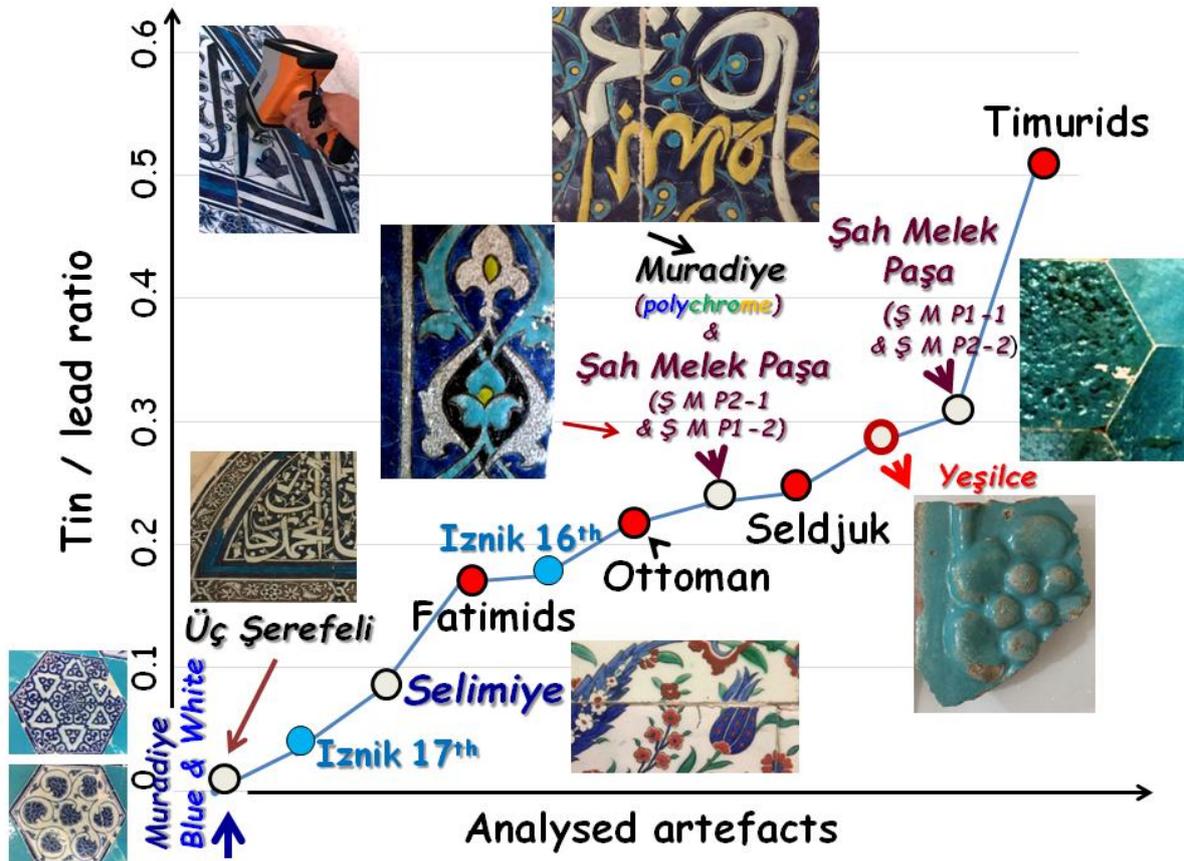
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1 **On-site pXRF analysis of glaze composition and colouring agents of “Iznik” tiles at Edirne**  
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11 **Abstract**

12 The production of the tiles in Ottoman Empire had begun as a continued workshop of Seldjuk ceramic art, and  
13 after this initial phase, its own technology was introduced into Ottoman art by local craftsmen. Iznik tiles are  
14 among the most appreciated pottery masterpieces, and wall decoration in tiles is a significant asset of Unesco  
15 World Heritage Edirne mosques. Rare glaze composition studies have been made, which justify the need for  
16 more comprehensive studies. We present here the first on-site elemental analyses performed with portable  
17 XRF instrument in four of the most representative mosques in Edirne (Şah Melek Paşa (1429), Muradiye (1435-  
18 1436), Üç Şerefeli (1410-1447) and Selimiye (1569-1575)). A handheld 785 nm Raman spectrometer was used  
19 as a complementary technique to identify some pigments. About forty tiles have been analysed in this  
20 research. Additionally, two tiles from Yeşilce Mosque (1442, Edirne) have been analysed at the Selimiye  
21 Foundation Museum. The weight percent of the elements measured with pXRF are normalised by Si amount in  
22 order to eliminate the variation due to the positioning shifts. Different glazing technologies are evidenced  
23 (Master of Tabriz Sn-free glazed tiles, Hünkar Mahfili Sn-poor glazed tiles, and Sn-(Bi) rich ones). At least three  
24 different cobalt ores have been used (with characteristic Cu, Ni, Mn and Bi content) in blue decors. Use of a  
25 chromium-based ore is demonstrated for some Selimiye Hünkar Mahfili tiles. The link between Şah Melek Paşa  
26 and polychrome (*mihrab*) Muradiye tiles and Seldjuk production is established. On the contrary, the  
27 technology of Üç Şerefeli and blue-and-white Muradiye tiles is unique and appears a precursor of 16th-century  
28 Iznik production. Bismuth, Sn/Pb, Co/Mn, and Co/Ni ratio appears very useful to compare the different glazes  
29 and to identify mining sources of cobalt.

30

31

32 **Keywords:**

33 B: impurities; B: Spectroscopies; C: Colour; D: Glaze; Iznik tiles; portable XRF instrument  
34 (pXRF); Ottoman ceramics; Edirne mosques; cobalt; tin;

35

## 1. Introduction

The production of the tiles in Ottoman Empire had begun as a continued workshop of Seldjuk ceramic art, and after this initial phase, an earlier ceramic technique had been introduced into Ottoman Art by local craftsmen. The Seldjuk period created important wall decoration in tiles. The colours employed are turquoise, blue, white, purple, black and gilt ornaments on the surface [1]. Detailed information about the beginning of the Ottoman ceramic art is given in the supplementary material.

Mainly, tiles of Ottoman period are separated into four groups depending on the production place: 1) Early 14th and 15<sup>th</sup>-century tiles with coloured glaze (undefined location), 2) Iznik production (14th -early 18th century), 3) Kütahya production (? – 20th-century) and 4) Tekfur Palace (Edirne Kapi, Istanbul) production between 1718 and 1750 [2-4]. Iznik tiles are among the most appreciated pottery masterpieces in the world, and the tile revetment is a significant asset of Unesco World Heritage - mosques of Edirne (formerly Adrianople). Edirne was established as the first Ottoman capital in 1365. Inspired from Chinese blue-and-white Ming porcelain decor, and from Persian style and technique, Iznik ceramics are characterized with a high gloss and high quality of drawing (sharp limitation between coloured areas without interdiffusion of colours), competing with China for the purity and quality of the white substrate [5-10]. Iznik city is located in the Marmara region, 140 km far from Istanbul and close to the ancient city of Nicomedia which was famous for the production of polychrome tiles during the Byzantine period [8].

Iznik production is now better known since the study of excavated shards [11-14]. The powerful colours are magnified because of the light reflexion by a slip made of crushed quartz grains. The palette of Iznik art is diversified by most of the colours, like blue, turquoise, and green, except the yellow colour which is unique. In the excavations of 1984, archaeologists found only one shard of yellow glazed tile which has a siliceous paste, dated to the first half of 15th-century [12] although, this colour was common in Seldjuk and Byzantine pottery. Because the tile was not completed, it can be assumed that it was only a tentative attempt of the technology of coloured glaze. Belgin Demirsar Arlı, third director of Iznik Tile Kilns Excavation, mentions that the production centre for the tiles having coloured glazes are unknown [14]. However, as stated above, the time of origin of the production of tiles and ceramics in Kütahya is not precise for the Seljukian and earlier period of the Ottoman Empire. There exist some indications that until 1428 when Kütahya came under the rule of the Ottoman Empire, there was using the Seldjuk ceramic tradition [15].

Most of the analytical studies have been focused on the body and slip examination. Comprehensive analyses of the glazed decoration are still little known, and the literature stays confused about it, a limited number of shards have been studied, and their date of production are not always well established. The object of this work is to compare characteristics related to the glaze production of tiles during the 15th and first half of the 16th-century. One of the big advantages of on-site measurement of building decor is that

1 the production date of the materials is much more documented than for the tableware  
2 ceramics.

3 We report here the first on-site elemental analysis of Ottoman tiles. About forty glazed tiles  
4 belonging to five different Edirne mosques have been analysed using a handheld XRF pistol.  
5 Complementary information was searched using a 785nm handheld Raman Spectrometer.  
6 Both techniques have demonstrated their potential for the non-invasive on-site study of  
7 pottery and glaze [16-21], with the support of previously made comprehensive studies at  
8 the laboratory on shards or objects [11,22-29]. The data will be compared to the previous  
9 study of samples collected in the framework of the project called “Digital Data Base of  
10 Topkapı Palace Tiles” from the collection of the museum reserved in the vaults [2,3,30]. The  
11 shards selected for determining the production technologies are of unknown origin. But, the  
12 production of these tiles is between early 16th and mid-17th-centuries. Additionally, the  
13 comparison will be made with the data collected on shards excavated from Iznik in the next  
14 paper. The Iznik production received the intensive attention of potters and scholars for a  
15 long time and for example, the famous French potter Theodore Deck made replicas to  
16 understand Iznik’s potter know-how [30] and information about the used technology  
17 [31,32]. The objective of this preliminary work is the comparison of the glaze technology  
18 used for the Edirne mosque tiles, their link and the search of the origin of the know-how and  
19 raw materials used.

## 20 **2. Experimental**

### 21 2.1. Technique

22 X-ray Fluorescence Spectroscopy analysis was performed using a Hitachi X-MET 8000 Expert  
23 Geo (Oxford Instruments) portable system equipped with rhodium (Rh) target X-ray tube of  
24 4W, 50kV max and a silicon drift detector (SDD). It was operated with the Mining LE method  
25 which uses low energy at 10 keV for determining low Z elements and high energy at 40 keV  
26 to identify the network modifiers and colouring/gilding agents found on the glaze. The beam  
27 size at the surface is 10.7mm x 9.4 mm, and a camera is used for controlling the measured  
28 area. Due to the heterogeneity in composition and variation in the glaze thickness, the  
29 measurements were carried out on at least two different areas of the same decor or glaze  
30 with 30s of radiation time (four days campaign of measurements). The results are reported  
31 as the mean values of all the individual point analyses done on each colour of the decor  
32 including the transparent glaze layer (see Table 1, Supplementary Materials). The results are  
33 reported by elementary weight percent calculated from the calibration method installed in  
34 the instrument. For semi-quantitative evaluation purposes, the  $K_{\alpha}$  lines of Mg, Al, Si, K, Ca,  
35 Sn, the transition metal elements (Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn) and trace elements (Rb,  
36 Sr, Zr, Ba, Bi), as well as the L spectral lines of lead were taken into consideration.

37 The calibration of the instrument was made by using the mineral standards which allows to  
38 measure ceramic materials with a high detection capability. As silicon is the major element

1 of the glaze, it is measured between 18 wt% and 31 wt%. To compare measurements,  
2 where the instrument tip-surface artefact distance varies, counts are normalized by those of  
3 Si wt%. For some easier comparison with the literature, some data were also transformed in  
4 wt % oxide.

5 Ancient ceramic glazes have a heterogeneous structure due to the firing conditions and  
6 constraints due to the thinness of the coating; contrarily to glass used to make objects and  
7 window, there is no long refining-annealing step of glass production. Indeed, the  
8 heterogeneity promotes the quality of the colour (opalescence or opacification, shade, etc.).  
9 Glaze precursor reacts with the substrate that led to precipitate of new phases controlling  
10 the gloss and the shade. Furthermore, the variable concentration of colouring agent,  
11 variation required for the decor design and the variable glaze thickness from place to place  
12 make the interaction volume with coloured matter (hence colouring elements) by X-ray  
13 beam is also highly variable. The determination and the comparison of 'a mean' composition  
14 does not make sense as for thick, optically clear, homogeneous glass artefacts. In this way,  
15 the glaze is more comparable to glass-ceramic of opaque mosaic tessera [33]. On the  
16 contrary, the analysis of specific elemental ratio, related to the raw material could be very  
17 informative [18]. Elements issued from single raw material in variable proportion in the  
18 glaze, for instance the colour precursor, will be located on a linear curve originating from  
19 the abacus origin. On the contrary, if two elements are independent coming from different  
20 raw materials used, the data will be distributed on a vertical or horizontal line, with some  
21 distribution due to measurement uncertainty and intrinsic heterogeneity.

22 A handheld model of B&W Tek, NanoRAM was used as a complementary technique to  
23 identify the pigments. The instrument is equipped of 785 nm NIR (Near Infrared) laser  
24 having a maximum 300 mW of power. The spectral range varies between 176 and 2900  $\text{cm}^{-1}$   
25 <sup>1</sup>. The measurement time was about one minute. The deficiencies of handheld Raman  
26 spectrometers are the lowest resolution (9  $\text{cm}^{-1}$  at 912 nm), the lack of advanced  
27 interchangeable optics than the laboratory type or of bigger mobile set-up spectrometers  
28 [19-21, 31,34]. Generally, handheld type Raman spectrometers are equipped with a single  
29 (785nm) or dual laser (785-1064 nm) system. As the laser power decreases when the  
30 wavelength increases, the Raman spectra of the glaze have lower intensities. Therefore, a  
31 baseline of the spectrum should be done very carefully in order not to lose the data of the  
32 glaze structure. The bending massif area of  $\text{SiO}_4$  is seen more widely between 400-600  $\text{cm}^{-1}$   
33 and the stretching massif area, which is the characteristic of the glassy silicate network, are  
34 observed between 900-1150  $\text{cm}^{-1}$  [19,31,34]; the narrow peaks which are characteristics of  
35 crystalline pigments, superimpose to the glassy silicate signature. However, the poor  
36 efficiency of a handheld instrument requires subtraction of the intense background that  
37 eliminates most of the broad bands.

## 38 2.2 Tiles

1 Figs 1 and 2 show the images of the analysed tiles. Details about the provenance of the  
2 samples and analysed spots are given in Table 1. As a supplementary material, the history of  
3 the mosques (Şah Melek Paşa, Muradiye, Üç Şerefeli, Selimiye tiles studied on-site, and  
4 Yeşilce tiles at the Selimiye Foundation Museum) where we analyzed the tiles is given in  
5 addition to some general view of the tiles of the mosques and the measurement set-up are  
6 given in Fig. S1.

7 In Şah Melek Paşa Mosque, blue, white, turquoise, yellow, black contour, as well as gilded  
8 decor (residues of gold), have been measured (Table 1). Designs are different for right and  
9 left contour tiles (Fig. S1d). In Muradiye Mosque, five hexagonal and one triangle tiles were  
10 measured (white, blue and turquoise areas, Table 1). Four polychrome tiles belonging to the  
11 *mihrab* were also analysed (blue, yellow, white, turquoise, green, purple areas and black  
12 line, Table 1). In Üç Şerefeli Mosque, two pointed-arch panels of tile which have been  
13 studied in this work, have been preserved (Fig. 1 and Fig. S1c, see full description in ref [1]).  
14 Although, the design of the two panels is similar, the background colours are inversed. The  
15 black-line is painted under the glaze in blue panel. Nine tiles located on the left of the two  
16 panels and the right of the blue panel have been analysed in different coloured spots, in  
17 total 36 (blue, white, turquoise, purple areas and black contour, see Table 1). On-site  
18 measurements in Yeşilce Mosque were not possible because the tiles were placed at an  
19 unreachable height. Therefore, two pieces of turquoise glazed moulded tiles fallen from the  
20 minaret of the mosque were measured at the Selimiye Foundation Museum (Table 1).  
21 Lastly, the measurements on Selimiye Mosque have been made on the tiles decorating the  
22 Sultan balcony (Fig. S1b).

### 23 **3. State of the knowledge on the Iznik ceramic technology**

24 The lime-rich alkali frit technology was introduced into Iznik ceramic art at the beginning of  
25 the 15th-century, and the production continued later with the lead-rich alkali fritware [4].  
26 Typically, the mean composition of an Iznik glaze is 20-40 wt% PbO, 10-15 wt% alkali (1.5  
27 wt% CaO+K<sub>2</sub>O), 1 wt% Al<sub>2</sub>O<sub>3</sub>. The presence of SnO<sub>2</sub> in the Iznik glaze, in which it varies  
28 between 2.5 and 8.5 wt%, plays a distinctive role in the identification of Ottoman ceramics  
29 [2]. However, in some tile revetments of the unique standing monuments of 13th-century  
30 Seldjuk period, Gök madrasah in Sivas and Tokat, were also found 3.7-5.8 wt% SnO<sub>2</sub> [35].

31 As summarized by L. Martinet et al. [36], two types of coloured glaze decoration can be seen  
32 in the beginning of the 15th-century. One of them is the legacy of the Timurid tradition  
33 (type 1) when the other one seems to be an Ottoman innovation (type 2). The second  
34 technique is well characterized on the tiles of Bursa and Konya. Tiles are made of different  
35 layers: a red body made of fine quartz grain (~1.5 mm) covered first with a quartz-rich ~200  
36 µm thick slip (~90% SiO<sub>2</sub>, Type 2) or with a cassiterite opacified lead-alkaline glaze (SnO<sub>2</sub> ~6-  
37 7%, i.e. over the solubility limit, Type 1); coloured overglazes are then deposited and fired.  
38 Then the red body was substituted with a mixture of white clay and fine quartz grains (<200  
39 µm) cemented with a soda-lead frit (also called stonepaste, fritware or even faience). For

1 16th-century productions, tin content ( $\text{SnO}_2 \sim 2\%$ ) of lead-alkaline glazes (blue, turquoise,  
2 and transparent glaze) is much lower; rare particles of cassiterite are detected: Tin remains  
3 dissolved in the glaze network. According to Martinet et al. [36], the black outline of Timurid  
4 (type 1) tiles is composed of Fe-Mn material, where iron is predominant, and aluminium  
5 quantities are important (4-8%). The Raman analysis shows a mixture of Braunitz ( $\text{Mn}_7\text{SiO}_{12}$ )  
6 and Hematite ( $\text{Fe}_2\text{O}_3$ ). The black outline is made of chromium-rich particles. On the other  
7 hand, Raman analyses of black lines of 16<sup>th</sup> century Iznik productions show the use of  
8 Chromite ( $\text{FeCr}_2\text{O}_4$ ) and/or Magnesiochromite ( $\text{MgCr}_2\text{O}_4$ ) spinels. In Ghazanid glazed  
9 potteries both 11th and 12th-centuries have been evidenced iron and magnesium chromites  
10 and manganese oxides [37]. The interest of using chromite and spinel in the borders is to  
11 capture the colorant ions (especially cobalt and copper) and prevent the diffusion of these  
12 colorants when the fusion of the glaze occurs at the high temperature of firing [31,34,36].

13 The main colorants of 15th-century productions are cobalt with traces of arsenic, iron, and  
14 nickel for blue, copper for turquoise, lead-tin for yellow (identified as type II) and  
15 manganese for brown. For 16<sup>th</sup> century productions, traces of arsenic and bismuth are  
16 measured in blue, copper in turquoise, lead-tin for yellow (identified as type II), and copper  
17 for green [36].

## 18 **4. Results**

### 19 4.1 Glaze

20 Fig. 3 compares normalized tin versus lead content measured in the transparent glaze or the  
21 glaze having a white decor beneath. Complete list of elemental composition is given in Table  
22 S1, Supplementary Materials. Data measured are comprised % wt Sn between  $\sim 0.05$  ( $0.06\%$   
23  $\text{SnO}_2$ ) and  $\sim 17$  ( $21.5\%$   $\text{SnO}_2$ ) and % wt Pb  $\sim 20$ -59 ( $21.5$ -64 %  $\text{PbO}$ ). Lines coming from the  
24 origin as expected for artefacts made with the same raw material as mixed Sn and Pb  
25 element source (Sn-containing ore or residues from bronze production?!) have been drawn  
26 as eyes guide to help the interpretation of the results.

27 Data collected on Selimiye tiles are well grouped and can be considered as a constant value  
28 or distributed along a line, considering that the measurement error is much smaller than the  
29 data distribution. The following issues are observed:

- 30 i) Glaze of (M2-M4, M9 and M10 blue-and-white tiles, monochrome (dark blue)  
31 border M8 from Muradiye *mihrab*, blue-and-white tiles from Üç Şerefeli (Fig. 1)  
32 and two polychrome tiles from Selimiye (S7 and S23, Fig. 2) are free of tin; PbO  
33 content varies between 20-30 wt% for Muradiye tiles, 31-36 wt% for Üç Şerefeli  
34 Mosque and 43-45 wt% for two polychrome tiles of Selimiye (S7 and S23).
- 35 ii) Pb and Sn contents measured on turquoise tiles of Yeşilce mosque are much  
36 higher: tile 82 with 43 wt%  $\text{PbO}$ , 14 wt%  $\text{SnO}$  and tile 83 with 64 wt%  $\text{PbO}$ , 22  
37 wt%  $\text{SnO}$ ).

- 1       iii)     Glaze with low Sn content ( $\sim 3$  wt%  $\text{SnO}_2$ ), significant lead content (30-38 wt%  
2       PbO) and traces of bismuth (0.1-0.2 wt%  $\text{Bi}_2\text{O}_3$ ) belong to Selimiye mosque.  
3       Bismuth was already observed by Martinet et al. in some similar shards [36].  
4       iv)     Glaze with high Sn content (5-9 wt%  $\text{SnO}_2$ ) belong to M1 monochrome  
5       (turquoise) and M5, M6 and M7 polychrome tiles of Muradiye and Şah Melek  
6       Paşa (polychromes and monochromes); Bi content is lower (around 0.06  $\text{Bi}_2\text{O}_3$   
7       wt%) than the glaze of type (iii).

8

9     When compared to the tiles of 16th-century collected from the collection of Istanbul  
10    Topkapı Palace Museum, it is seen that the use of high amount of tin oxide in the glaze of  
11    16th-century Topkapı shards is rather similar to that measured in Muradiye and Şah Melek  
12    Paşa panels. [2,3] Sn level measured on 17th-century shards is similar to that of Selimiye  
13    tiles. It is clear that the main criterion is not the time of production. This indicates that  
14    specific workshops (and the raw materials and recipes) have produced the tiles for each  
15    building and that different productions can be found in the same building.

16    The tiles of Şah Melek Paşa (Figs 1 & S1d, Table 1) and Muradiye (Figs 1 & S1a, Table 1) are  
17    accepted as the preliminary productions of Iznik as the mosques were constructed in the  
18    15th-century, and the quality of the quartz slip is the only thing that gives rise to the perfect  
19    white colour.

20    The two tiles from Selimiye (S7 and S23, Fig. 2), in which data are always found out of the  
21    SMP group can be considered as not genuine ones because of the inconsistency of the  
22    motifs and colours of the adjacent tiles (S8 and S22). The difference between the data  
23    measured for these two tiles (see for instance Fig. 3) can be considered as an illustration of  
24    the 'intrinsic' measurement error and variability for a same type of glaze. In addition, from a  
25    personal communication with the art historian of Selimiye Foundation Museum, we know  
26    that Üç Şerefeli mosque was restored after the earthquake of 1752, but the original tiles of  
27    the mosque which were technically similar to the blue-and-white tiles of Muradiye, were  
28    also preserved.

29    The consideration of Pb/Si vs. Mg/Si measurements (Fig. 3) gives complementary  
30    information and confirms the three groups evidenced. The glaze is expected to be prepared  
31    from a mixture of quartz (75%) and clay (25%) – present-day composition of Kütahya glaze  
32    [38,39] – plus eventually the frit made of silica and fluxing oxides: PbO,  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ , CaO,  
33     $\text{B}_2\text{O}_3$  and  $\text{Bi}_2\text{O}_3$ . Due to the low Mg content and hence intrinsic measurement error, the data  
34    are more scattered, however relatively similar, consistent with the use of (a) same/similar  
35    source(s) of silicon, magnesium being an impurity of silicates. Actually, no correlation is  
36    observed between Pb and Mg, as expected and horizontal lines should be considered: the  
37    peculiar character of S7 and S23 restoration is obvious. Muradiye tiles show the largest  
38    distribution and two groups appears (M5, M6 and M7, i.e., the polychrome tiles) and the  
39    blue-and-white ones. This difference can be related to different clays, which selected for

1 artefact made with a different shape (and techniques: moulded and flat tiles, respectively).  
2 The similarity between Selimiye and Şah Melek Paşa groups is also clear.

3 Examination of the Al vs. Si and Al/Si versus Ca/Si scatter plots (Fig. 4) confirms the  
4 differences evidenced in Fig. 3. Discrepancy between polychrome and black-and-white  
5 Muradiye tiles is obvious. Focus on the fluxing elements measured (Al/Si vs Pb/Si and K/Si  
6 versus Ca/Si can be seen in Fig. S2, Na being not detected with our portable XRF) shows  
7 small distribution of the data; a higher mean value of aluminium content is measured for  
8 Şah Melek Paşa (1.35 wt% Al<sub>2</sub>O<sub>3</sub>), as visible on Al/Si vs Ca/Si as well as Al vs. Si scatter plots  
9 (Fig. 4). Lowest Al content is measured for Selimiye and Üç Şerefeli mosque tiles. This  
10 indicates that for each mosque different clays have been used to prepare the glaze. The tiles  
11 of Muradiye and Şah Melek Paşa, which were dated to the same period, show differences in  
12 the clays used. However, similar or same Pb-Sn source was used for Şah Melek Paşa and  
13 some Muradiye tiles. This is confirmed by the similar level of Bismuth. Presence of bismuth  
14 is related to the cobalt ore used and occasionally Bi is encountered in turquoise decor  
15 associated with copper. The very low content of vanadium appears characteristically of S7  
16 and S23 replica and of S8 and S9 tiles.

#### 17 4.2. Colouring agents

18 Particular attention was paid to Co signals. Fig. 5 compares the scattered plots Co/Si vs.  
19 Cu/Si, Ni/Si, Mn/Si and Bi/Si vs. Co/Si. There is no evidence of the presence of arsenic in the  
20 tiles; measurements with the same instrument and procedure was able to detect As traces  
21 in blue decor at level lower than a level 0.01% in excavated polychrome Iznik shards from  
22 16th and 17th-centuries [40]. Re-examination of previous on-site recorded Raman spectra  
23 [34] on a circa 1580 dish by recent works [2,3] also confirm that some cobalt contains  
24 significant levels of arsenic. A good relationship between cobalt, copper and nickel is  
25 obvious. The distribution of data indicates the use of at least 4 different cobalt-based  
26 colouring agents. Different types of cobalt may have been used at the same time.

27 Co vs Cu and vs Ni data obtained from the tiles of Şah Melek Paşa are well distributed on a  
28 line issued from the origin. Cu content remains low. For Muradiye tiles, the two groups  
29 identified, correspond to those previously identified from Sn content, the blue-and-white  
30 and the polychrome tiles (Figs 1 and S1a). The low cobalt content measured for the first  
31 group is explained by the fact that the cobalt underglaze blue drawing is covered with a  
32 rather thick glaze limiting the Co signal excited/collected. Consequently, considering the  
33 Co/metal ratio is more informative (see Tables S2 and S3, Supplementary Materials).  
34 Selimiye results are well distributed on a single line except for S22 and S18 tiles that show  
35 the lowest Cu content. This discrepancy disappears on Co/Si vs. Ni/Si, where only three  
36 groups are observed: Selimiye, Şah Melek Paşa and Muradiye + Üç Şerefeli. Only the latter  
37 group shows that used cobalt was an association of Co, Ni and Mn. The constant (high) value  
38 (0.34-0.36 wt% CoO) of dark blue M5-M7 Muradiye and Şah Melek Paşa tiles indicates  
39 voluntary addition of manganese or more likely a mixing with Asian cobalt, very rich in

1 manganese [41], according the hypothesis of cobalt ore oriental sourcing of Porter [42].  
2 However, blue colour in M5 contains 0.5 wt% MnO but in M7 MnO is very low, 0.070 wt%.  
3 This confirms that different cobalt ores have been thus used, voluntary or not  
4 (heterogeneity of the ore?).

5 A correlation between Cu/Si and Fe/Si signal is measured except for Muradiye and Şah  
6 Melek Paşa tiles (Fig. 6). CuO varies between 1.6-3 wt% for green/turquoise glaze of  
7 polychrome and turquoise monochrome of Muradiye tiles. Chromium is detected only in  
8 some green Şah Melek Paşa (Panel 2, painted on blue) and Selimiye tiles (S7, S8 & S9), as  
9 well in M7 polychrome tile (Fig. 7). If Cr can be an impurity of copper used for turquoise Şah  
10 Melek Paşa tiles, the constant Cu content measured for S7, S8 and S9 tile glazes is consistent  
11 with the voluntary addition of a chromium-rich material. Chromite was detected by Raman  
12 scattering in some green glazed Iznik dishes [34] (as well in, of course, the black-lines).

13 The blue area of three tiles examined in this study (M9, Ş M P1-1 and 3Ş1-L) have high level  
14 of sulphur and calcium (3-4.6 wt% S, 4-8.8 Ca) and small amount of Ti (0.1-0.2 wt%)  
15 associated with cobalt: 0.007 % M9, 0.060 % Ş M P1-1, 0.020 % 3Ş1-L (Figs. 4 and S2). The  
16 high level of sulphur is surprising and can reflect the use/addition of lapis lazuli in the blue  
17 decor. Use of lapis lazuli powder, alone or in association with cobalt, is well-documented  
18 since Ptolemaic Dynasty and common in Persian and Mamluk production during 13th to  
19 15th-centuries [43,44].  $S_n^+$  ions are the chromophores of Lapis Lazuli. Raman measurements  
20 excited with green laser or invasive study are needed to have a definitive conclusion.

21 Yellow decor of the tiles of Muradiye (M5, M7, Fig. 1) and the panels of Şah Melek Paşa (see  
22 Figs 1 and S1d) is measured and found quasi-constant (0.38-0.63 wt%  $SnO_2$ , Fig. S3); the tin  
23 content was similar to that of the transparent glaze, but with a higher amount of Fe (1.4-1.7  
24 wt% FeO), which indicates incorporation of iron in lead-tin pyrochlore solid solution, as  
25 commonly observed [45]. Raman spectra collected on M5 and M7 yellow areas (Fig. 8)  
26 confirms the use of Pb-Sn(-Fe) pyrochlore as yellow pigment (reference peaks at 325 and  
27  $450\text{ cm}^{-1}$  [45,46]). The amount of Sb is very low in these tiles ( $\sim 0.08\text{ wt}\% Sb_2O_3$ ). The strong  
28 peak at  $\sim 1365\text{ cm}^{-1}$  observed on all Muradiye polychrome glaze is assigned to fluorescence,  
29 perhaps with additional contribution of hematite minor phase [34,46]. CaO content in the  
30 yellow decor of Şah Melek Paşa is around 3wt% and  $Al_2O_3$  1.3 wt%.

31 Among the tiles examined, only two tiles of Muradiye (M6 and M7) and the panel of Üç  
32 Şerefeli Mosque contain purple colour in the decor (Figs 1, S1a & S1c). The analysis showed  
33 that the mixture of manganese and iron was used in the tiles of Muradiye (M6 & M7). But  
34 the amount of Mn and Fe is different for these two tiles with a ratio of Mn to Fe 0.8 for M6  
35 and 4 for M7 (Table S3). The lighter purple colour of M7 tile contains less Mn and Fe having  
36 the relative ratio as 2.1. The purple colour is more brownish in the panel of Üç Şerefeli  
37 mosque and it does not contain any manganese in the composition. Iron was also found in  
38 lower amounts than the Muradiye tiles, as  $\sim 0.27\text{ wt}\%$ .

1 Black contours have also been analysed in this study. However, their small size makes that  
2 the measured data are always contaminated by the adjacent area that is the surface  
3 analysed being much larger than the black lines. As reported by Martinet [36] two types of  
4 black lines are identified (Fig. 7): Selimiye and Üç Şerefeli mosque tiles are free of  
5 manganese. On the contrary some Muradiye mosque polychrome tiles (M7 & M5) show  
6 significant Mn content (and no chromium) although chromium is clearly measured in  
7 Selimiye tile lines, as in many 16th-century Iznik dishes [34].

8 Raman spectroscopy was used as a complementary technique to identify the pigments. In  
9 this study, we used a 785 nm handheld model. The quality of the recorded signal is not so  
10 efficient compared to the signal, which collected under blue or green excitation at the  
11 laboratory or with heavy mobile set-up [21] but, some phases are identified. Hematite (peak  
12 at  $1340\text{ cm}^{-1}$ ) is identified in the brown colour of Selimiye tiles in agreement with XRF data  
13 of the same colour. On the white and blue area of M5, a weak doublet at  $630$  and  $775\text{ cm}^{-1}$   
14 as expected for  $\text{SnO}_2$  cassiterite precipitate, not only Sn dissolved in the glaze is observed  
15 for the tile exhibiting the highest measured Sn content (Fig. 3). Whatever the high noise, a  
16 feature observed at  $\sim 450$  and  $990\text{-}1055\text{ cm}^{-1}$  fit well with previous Raman analysis of Iznik-  
17 type glaze. [34,47] The narrow stronger peak of quartz is also measured at  $460\text{ cm}^{-1}$ , the  $5$   
18  $\text{cm}^{-1}$  downshifts arising from the compressive stress of the glaze, as usual.

## 19 5. Discussion

20 Hexagonal blue and white tiles of Muradiye Mosque are visually very similar to those  
21 decorated in the tomb of Halil el Tebrizi and Umayyad mosque in Damascus [5]; therefore, it  
22 has been assumed that the tiles of Muradiye were produced by the artists coming from  
23 Damascus in relation with “Masters of Tabriz” (or better “Master of Samarkand”, [48,49]).  
24 This blue-and-white décor representing the “Chinoiserie” style was an innovation for Edirne  
25 [48]. On the contrary of Lane [6] who states that hexagonal blue and white tiles and  
26 polychrome tiles of the *mihrab* could be produced by the same potter, we identified that  
27 different raw materials and technology were used for these two groups of tiles. As the  
28 coloured glazed tiles of the *mihrab* are stylistically similar to those of Green Complex in  
29 Bursa where the Masters of Tabriz worked, further on-site research must be carried out to  
30 identify the origin of this group of tiles. Damascus ceramic art in Syria had started in 1250  
31 and existed until 1350 when Tabriz ceramics started to be produced in Persia until 1500  
32 [24]. Application of decor under the transparent alkaline glaze is likely to have been a long-  
33 standing practice in Damascus in the contrary of Iranian technique. The body of Damas  
34 ceramic, called stonepaste, is very rich in quartz (90 %) and low in clay (3 %) with higher CaO  
35 (4 %) which makes it much whiter like Chinese porcelains while Tabriz ceramic paste  
36 contains around 75 %  $\text{SiO}_2$ , 12 %  $\text{Al}_2\text{O}_3$ , 4 % CaO, 5.5 %  $\text{Na}_2\text{O}$ , 1.5 % MgO according ref [24].  
37 Tin opacified alkaline glazes become very popular in that period with the use of soda flux. A  
38 Persian pottery treatise mentions that the flux soda was extracted from the burnt plant  
39 salsola soda [50]. A. Lane also states that blue and white technology was introduced into

1 Syria by the Iranian artists [6]. Last but not least, the polychromic tile panel of the *mihrab* of  
2 Muradiye also has similarity on composition of the glaze, and black contour (Fe-Mn spinel)  
3 with the polychromic shard (TOP F18a1 reserved in the vaults of Topkapı Palace Museum)  
4 examined during the project of “Digital Data Base of Topkapı Palace Tiles [3, 30].” This shard  
5 was different with the use of Mn and Fe instead of the use of chrome-based pigment in the  
6 black lines of tiles assigned as “Iznik”. Sn ve Pb content in the glaze of TOP F18a1 is closer to  
7 the polychromic tiles of the *mihrab* of Muradiye. And also, the stonepaste of TOPF18A1 was  
8 found rich in clay on the contrary of rich in quartz, so it was assumed that the tile was  
9 produced in Kütahya, in the earlier periods than the traditional Kütahya production during  
10 the Ottoman Empire. Fig. 9 compares the Sn/Pb ratio of Edirne tiles with characteristic  
11 ratios extracted from elemental analyses of the literature regarding Timurid [24], Seldjuk  
12 [24], Ottoman [2,3] and Fatimids [24] productions. The Sn/Pb ratio of polychrome *mihrab*  
13 tiles are close to that of Seldjuk production (Fig. 9) [2,3,24]

14 The two tiles of Yeşilce Mosque exhibit much more amounts of lead and tin oxide than the  
15 other tiles, reflecting a completely different technology. Their Sn/Pb ratio is the closer one  
16 of that of Timurid production (Fig. 9). Tite and his colleagues studied some Egyptian and  
17 Syrian yellow and green glazes [51] and they found that Syrian glazed family was containing  
18 lead stannate ( $\text{Pb}(\text{Sn},\text{Si})\text{O}_3$ ) particles as an opacifier. For Syrian glazes, they found around 65  
19 wt% PbO, 25 wt%SnO<sub>2</sub> and 10 wt% SiO<sub>2</sub>. However, the content of SnO<sub>2</sub> in the opacifier agent  
20 was higher in Egyptian glazes. From our analyses done on Yeşilce tiles (especially tile 83), we  
21 defined exactly the same particle composition of lead stannate as Syrian glazes contain.  
22 Because lead stannate particles were dispersed broadly on the glaze layer, we were able to  
23 detect with pXRF instrument without seeing the microstructure of the tile. Thus, it can be  
24 expected that the tiles of Yeşilce Mosque could be produced in Syria and imported to Edirne  
25 during the construction of the mosque (1440-1441).

26 When compared Sn/Pb ratio of the tiles studied with the data of the literature, it is seen  
27 that Sn/Pb decreases from Timurids period to the 17th-century of Iznik production [2,3,24].  
28 Tin is a high cost ingredient and it is understandable that its amount will be reduced with  
29 the increasing production and efficiency of the production. The earlier productions, namely,  
30 panels of Şah Melek Paşa and *mihrab* of the Muradiye and also Yeşilce mosque, are grouped  
31 around Seldjuk mean data and before the beginning of the master period of Iznik. The tiles  
32 of Selimiye mosque are exactly in between 16th and 17th-centuries of Iznik production. As  
33 the tiles of Üç Şerefeli and Muradiye blue-and-white do not contain any tin oxide in the  
34 glaze, they are placed at Sn/Pb: 0.

35 Masters of Tabriz was using the technology of lead-lime frit technology, and very high  
36 content of calcium in the glaze of Şah Melek Paşa tiles shows the first technology introduced  
37 by the Masters of Tabriz before starting the use of lead-alkali frit.

1 The variation of Al vs. Pb clarify the use of different raw materials mainly in three groups,  
2 Şah Melek Paşa, Muradiye and Selimiye with the tiles of Üç Şerefeli Mosque. The tiles out of  
3 their own groups can reflect a restoration.

4 The presence of bismuth in cobalt minerals is seen for the tiles of Selimiye (Fig. 3, Table S1).  
5 This is consistent with Porter's hypothesis for the use of Erzgebirge (European) cobalt ores  
6 and the remark of Constantinescu which tells that a change in the composition of the blue  
7 glazes was observed in 1520 by the presence of As and Bi by reduced amounts of Fe and Ni  
8 [29]. In the tiles of Edirne, arsenic was not detected, but bismuth is relatively higher in  
9 Selimiye Mosque than the older mosques tiles. For some Muradiye tiles (M5, M7, M11), and  
10 tiles of Şah Melek Paşa panels, bismuth is independent of cobalt source and nearly  
11 inexistent. Detection of bismuth can be an evidence of the period of Iznik tiles. It must be  
12 taken into account primarily in blue pigments and also in turquoise colour associated with  
13 copper.

14 Fig. 10 compares the Co/Mn and Co/Ni ratio measured on Edirne tiles with characteristic  
15 values of the literature: blue-and-white Chinese porcelain decorated using Asian cobalt  
16 (Ming, Xuande and Qing Dynasty [52-54]) or with cobalt imported from the West (Iran [55]  
17 and/or Europe, Yuan Dynasty [56], Della Robbia [57,58], Renaissance Europe [59]); cobalt  
18 from Egypt was also considered [55]. When compared with the reference data [52,59], the  
19 blue colour of restored tiles of Selimiye (S7 & S23) is closer to the Asian sources whereas  
20 the tiles of the *mihrab* of Muradiye is around the Egyptian source. The central panel of the  
21 *mihrab* of Muradiye is different than the adjacent tiles of the *mihrab*. The main panel (M7) is  
22 close to the Yuan (low Mn) blue colour which reflects the use of European or Persian  
23 sources. On the contrary, M5 to M8 tiles are made with Asian cobalt. Selimiye is also close  
24 to the Egyptian sources, not far from Asian cobalt.

25 The ratio of CoO vs. NiO (Fig. 10 and Table S3) increases depending on different periods of  
26 production, which reflect the change of the raw materials used by centuries. For Muradiye  
27 and Üç Şerefeli Mosques, the ratio is around 1, for Şah Melek Paşa 1-2, and for Selimiye it is  
28 increased up to 3-4. On the other hand, MnO/CoO ratio is less for Selimiye (2-3) and higher  
29 for Muradiye and Şah Melek Paşa tiles (7-8). The results report that Selimiye production at  
30 16th-century is separated from the mosques constructed at 15th-century.

31 From the Co/Mn and Co/Ni ratios, it is definitively stated that blue-and-white tiles of  
32 Muradiye is different from the tiles of the *mihrab*. And one tile of Üç Şerefeli on the right  
33 side of the Panel 1 has a different blue than the other tiles of the same (3Ş P1) and adjacent  
34 panel (3Ş P2).

## 35 **6. Conclusions**

36 This preliminary study presented the first on-site elemental analyses performed with  
37 portable XRF instrument complemented by a handheld Raman spectrometer, in four of the

1 most representative mosques in Edirne (Şah Melek Paşa (1429), Muradiye (1435-1436), Üç  
2 Şerefeli (1410-1447) and Selimiye (1569-1575)). The techniques which are configured as  
3 portable systems are very efficient and time-saving for better understanding of the nature  
4 of the objects and enlightening scientifically the art history. In this study, the objective was  
5 to understand the technology of Ottoman tile revetments used in architectural buildings in  
6 Edirne, depending on the origin and production period. Table 2 summarizes the main  
7 conclusions. The consideration of Sn/Pb and Co/Mn ratio gives a good view of the  
8 technological links between the different productions, for instance between Timurids,  
9 Seldjukid and Ottoman productions. The tiles of Selimiye represent a very controlled  
10 production of Iznik technology with the colours used in the decor assigned definitively to  
11 Iznik origin, in the 16th-century. The very good homogeneity of glaze production related to  
12 the efficiency of the Nakkashane office was already observed for Iznik tableware [34,60].  
13 However, Şah Melek Paşa and Muradiye tiles which represent the earlier production of  
14 Ottoman ceramic art, differ stylistically (different colours used in the decors) and technically  
15 (different raw materials used in the glaze). Şah Melek Paşa tiles appear to be an assemblage  
16 of tiles from different sources. And also, blue-and-white tiles of Muradiye differentiate from  
17 the polychrome tiles of the *mihrab*. Blue-and-white decor of these tiles could be assigned as  
18 the “Chinoiserie” style which was an innovation for Edirne. It is assumed that the craftsmen  
19 who worked in the production of hexagonal and polychromic tiles of the *mihrab* are  
20 different. The evidence of the use of different cobalt ores is a novel information which  
21 allows to clarify the studies of provenance carried out on Ottoman tiles. The other  
22 discriminating point of this work is that Muradiye and Üç Şerefeli tiles are unique and  
23 precursors of the 16th-century Iznik production. Further on-site investigations must be  
24 carried out in Green Complex in Bursa, as well as Kütahya’s buildings dated to the end of  
25 Seljuk and earlier period of Ottoman.

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## 37 **Appendix A. Supplementary material**

38 Supplementary data associated with this article can be found in the online version at  
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1 **Table 1.** Description of the samples analysed in this study

Code of the tile	Origin of the tile	Decors measured with pXRF	Remarks	
S4	Selimiye Mosque, Edirne (Hünkar Mahfili)	White		
S7		Blue, Green, White, Red	reproduction?	
S8		Blue, Green, White, Red		
S9		Blue, White, Turquoise line		
S10		Blue, White		
S11		Blue		
S12		Red, Black line		
S13		Red		
S14		Black line		
S15		Black line		
S16		N.A.	XRF meas. was not successful.	
S17		Black line		
S18		Blue line		
S19		Black line		
S20		Blue line		
S22		Blue, White, Red		
S23		Blue, White, Red	reproduction?	
S24		Red		
S25		Turquoise		
S-border		Turquoise		
M5	Muradiye Mosque, Edirne	Blue, Yellow, White, Turquoise, Black line	<i>mihrab</i> Polychrome tiles	
M6		Green, Turquoise, Purple		
M7		Blue, Green, Yellow, Purple, White, Turquoise, Black line		
M8		Blue		
M1		Turquoise	decorated with blue-and-white tiles	
M2-4		Blue, white	Hexagonal tiles	
M9		Blue		
M10		Blue, White		
M11		Blue		
M12		Blue		
S M P1-1		Şah Melek Paşa Mosque, Edirne	Blue, Yellow, White, Turquoise, Black line	Panel (left), polychrome
S M P1-2			Turquoise, gilded	Monochrome, hexagonal
S M P2-1	Blue, blue contour, Yellow, White, Turquoise, Black line		Panel (right), polychrome	
S M P2-2	Turquoise, gilded		Monochrome, hexagonal	
3S-P1L (3 tiles)	Üç (3) Şerefeli Mosque, Edirne	Blue, White, Turquoise	1- White base, 2-White base, 3- Blue base	
3S-P1R (3 tiles)		Blue, White, Turquoise, Black line	1- Blue base, 2- White base, 3- White base	
3S-P2L (3 tiles)		Blue, Purple, White, Turquoise, Black line	1- Purple base, 2-white and blue base on the same tile 3- purple and blue base	
82	Yeşilce Mosque, Edirne	Turquoise	Fallen tiles preserved in the Selimiye Foundation Museum	
83		Turquoise		

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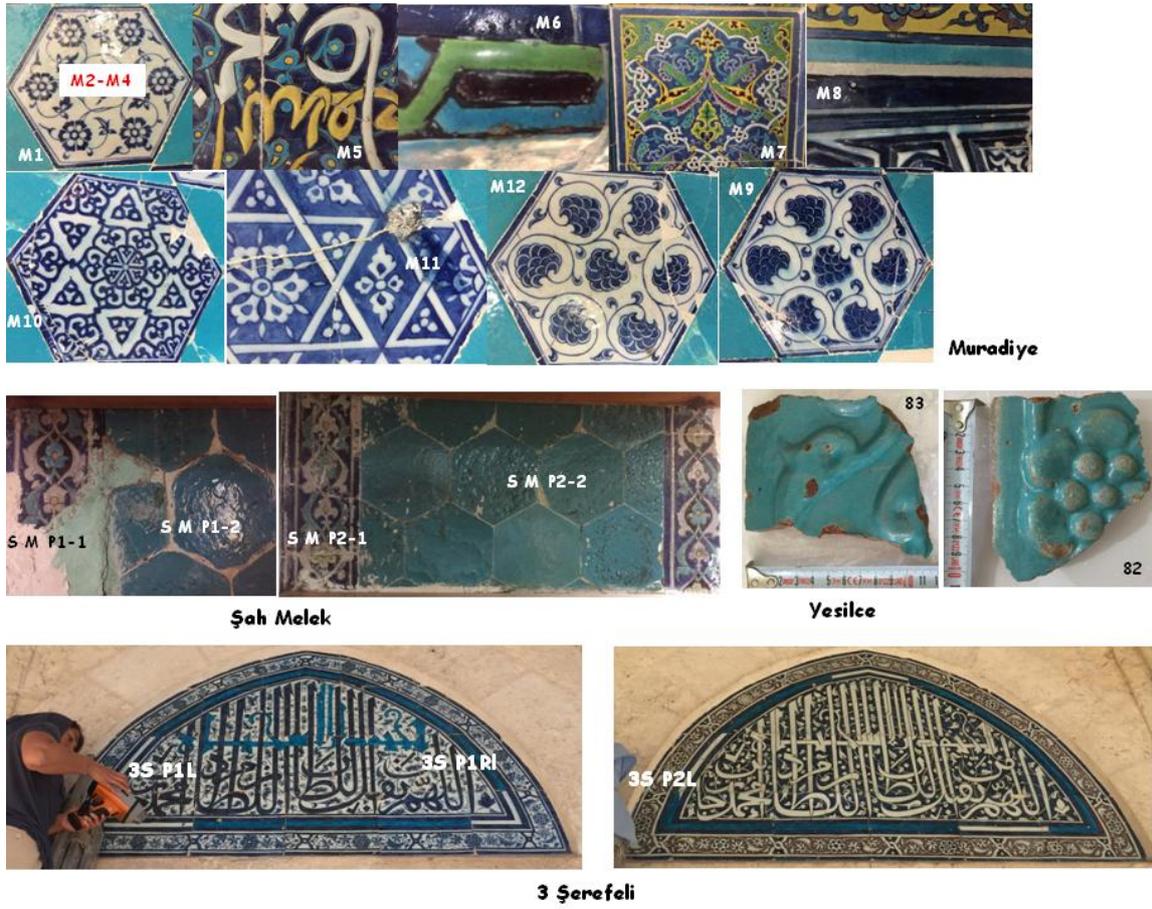
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1 **Table 2.** Final remarks of the tiles examined in this study according to pXRF measurements

Tiles examined on this study	Conclusion	
Selimiye	S7 and S23 different from other Selimiye tiles studied	Probably, modern restoration
	Other Selimiye tiles studied	Iznik production, intermediate between 16 <sup>th</sup> and 17 <sup>th</sup> Iznik tablewares [2, 41]
Muradiye	Turquoise triangle tile (M1) similar to tiles of the <i>mihrab</i> (M5-M6-M7)	Related to Timurid and Seldjuk heritage
	Blue-and-white tiles	Very specific production connected to Üç Şerefeli tiles
Şah Melek Paşa	The two panels not similar	Close to M7 polychrome Muradiye (assemblage made of different sources)
Üç Şerefeli	All the three panels similar	Close to B&W tiles of Muradiye
Yeşilce	82-83 different	Different from all the tiles, Linked to Seldjuk and Timurides production

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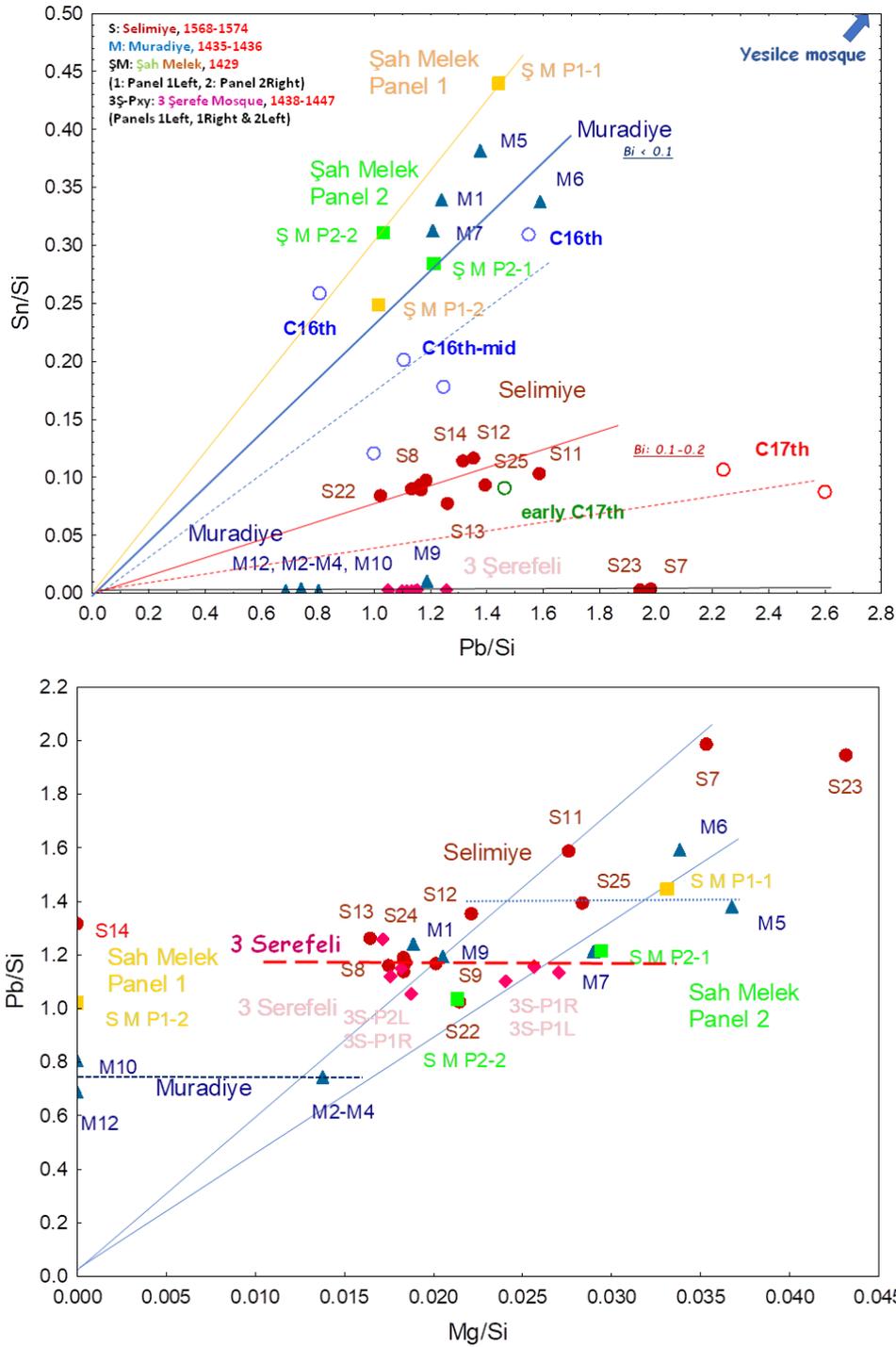
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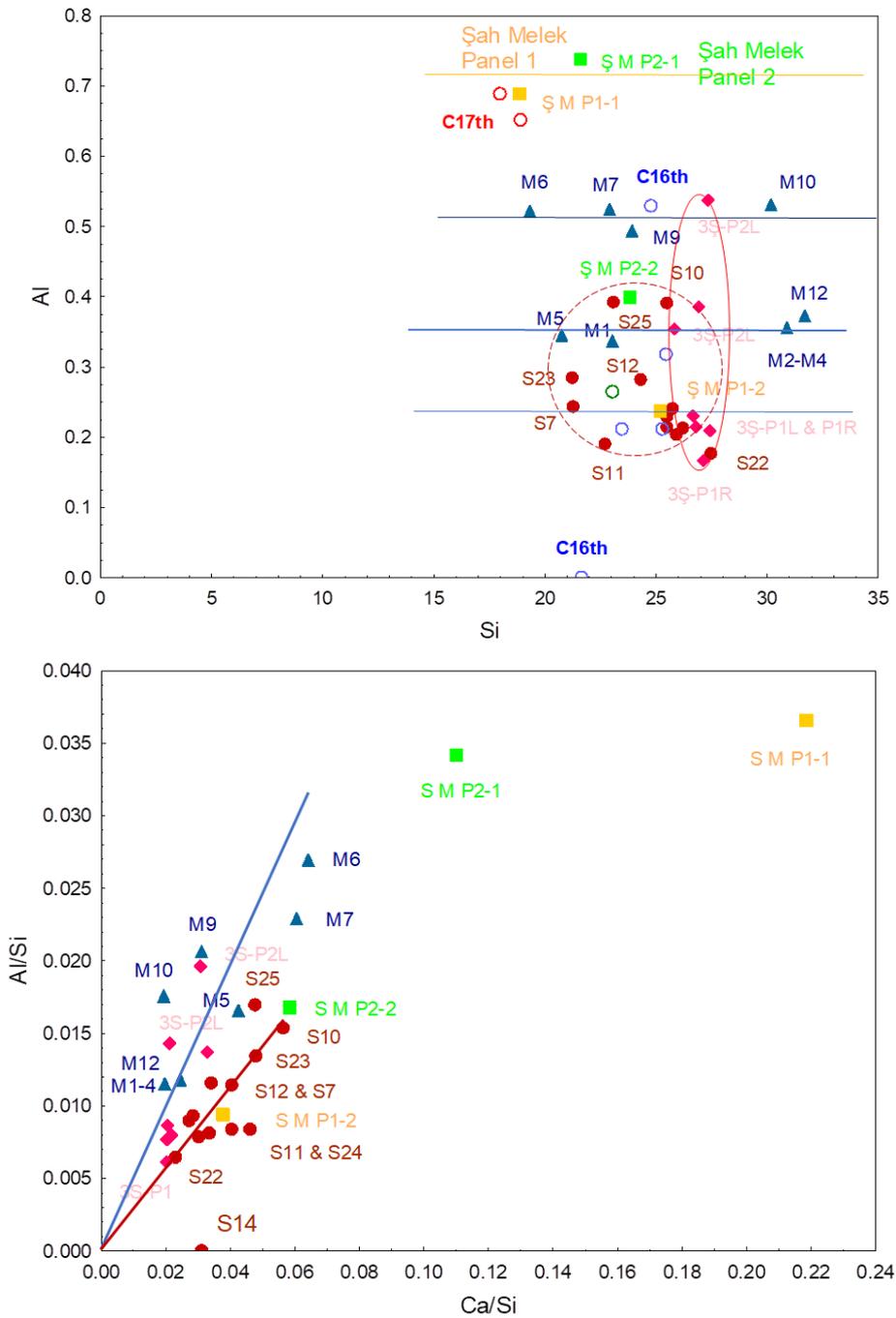
**Fig. 1:** On-site studied tiles of Muradiye (M), Şah Melek Paşa (Ş M) and Üç Şerefeli mosque in Edirne (Turkey); see Table 1 for details. Tiles fallen from Yeşilce mosque (Edirne) have been analyzed at Selimiye Foundation museum.



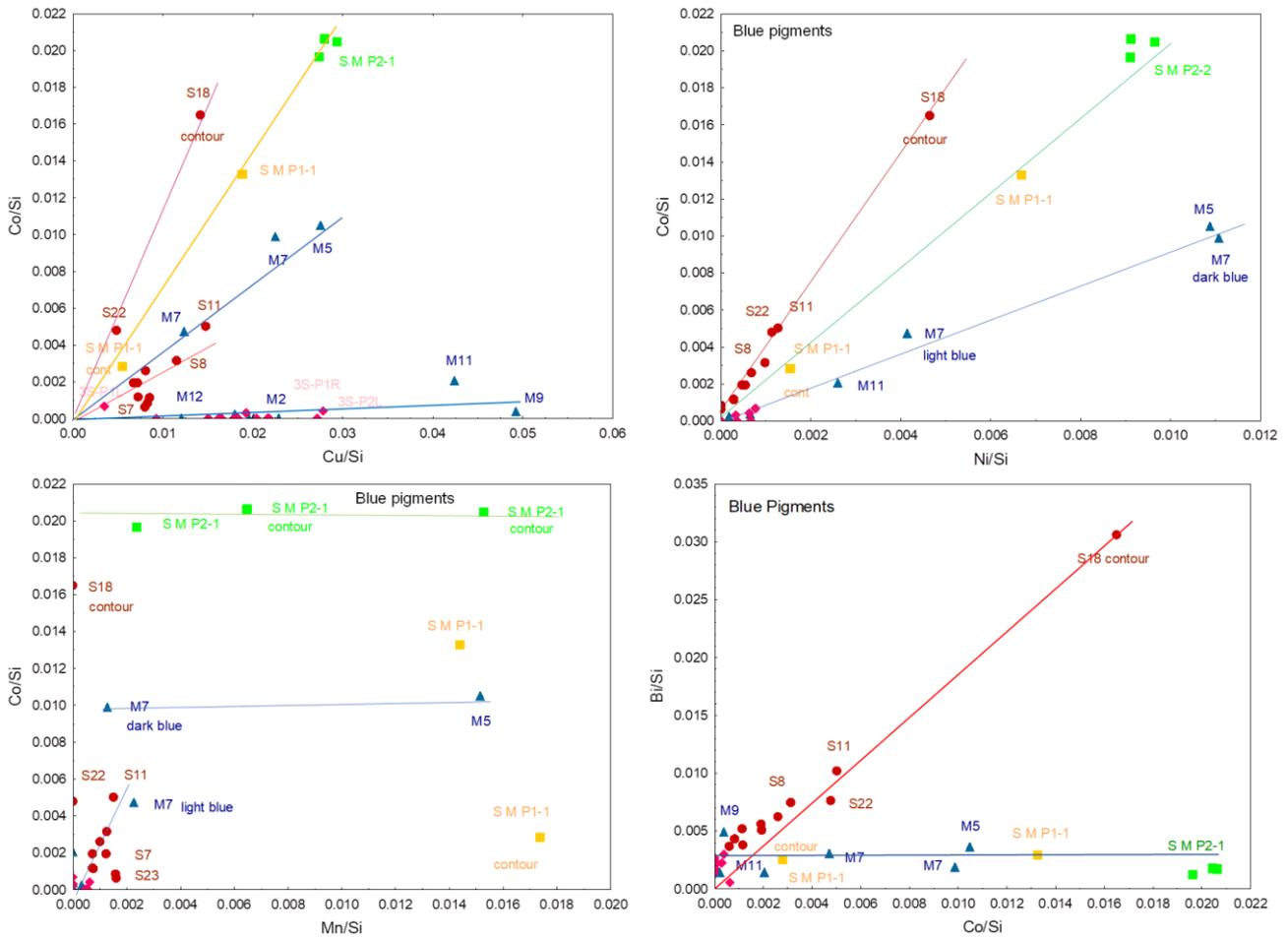
**Fig. 2:** On-site studied tiles of Selimiye mosque, Edirne (Turkey); see Table 1 for details.



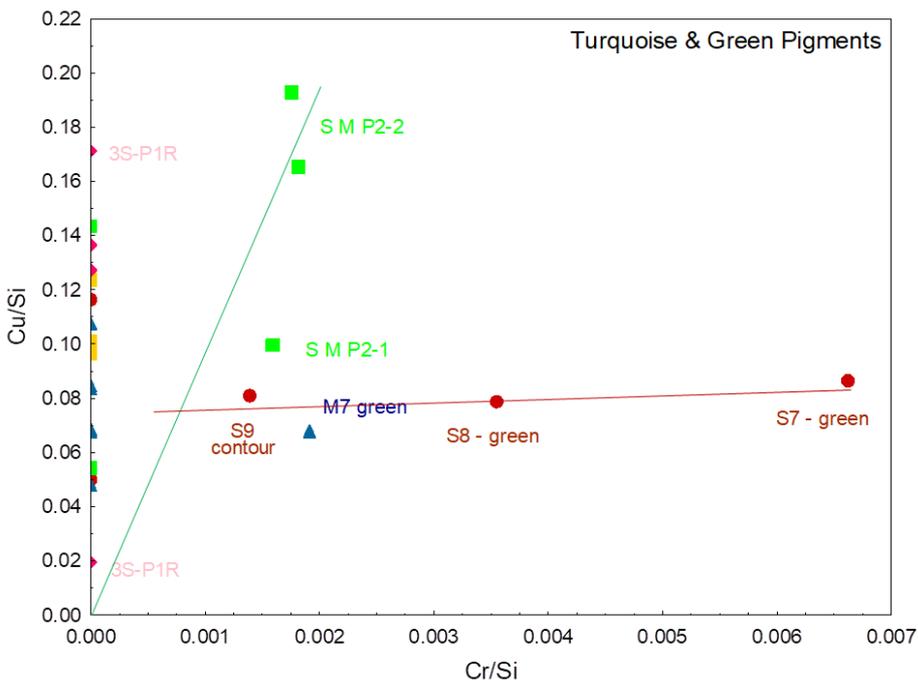
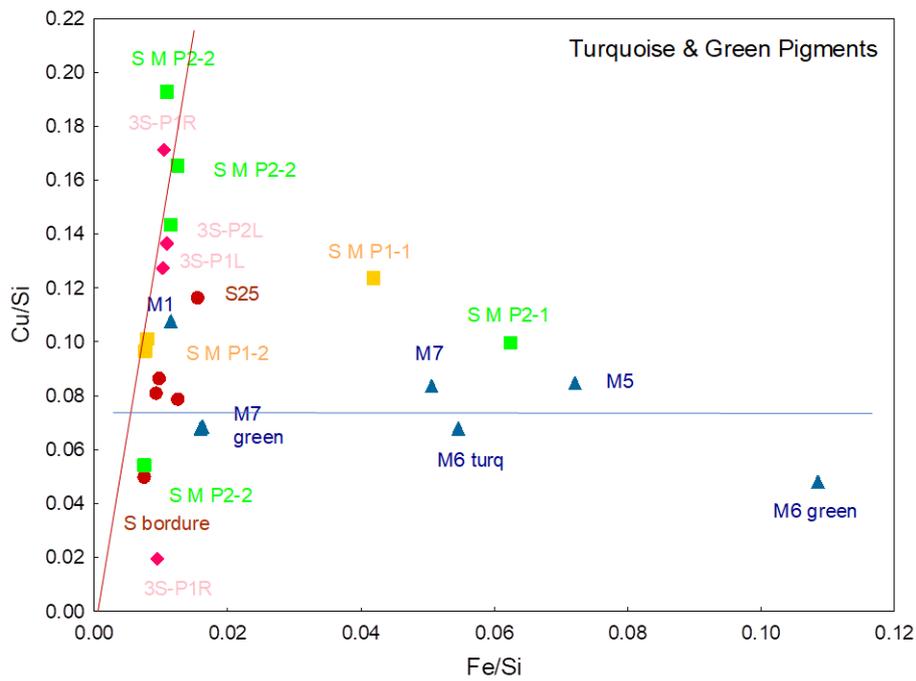
**Fig. 3 :** Scatter plots of the weight % ratios of Sn/Si versus Pb/Si (upper) and Pb/Si versus Mg/Si (bottom). Data collected at Mining LE method of pXRF instrument on the tiles of Selimiye Mosque (red circle), two panels of Şah Melek Mosque (green and orange solid square), Muradiye Mosque (blue solid triangle) and Uç Şerefeli Mosque (pink solid diamond) are plotted in the graph; see Table 1 for details of the tiles measured. Lines are guide for eyes.



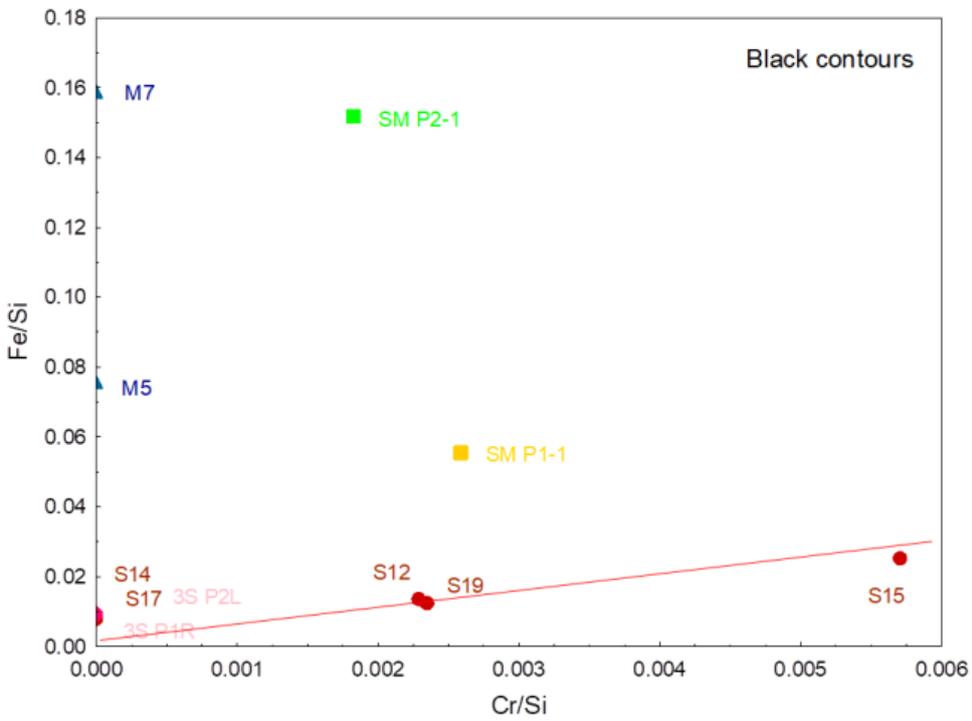
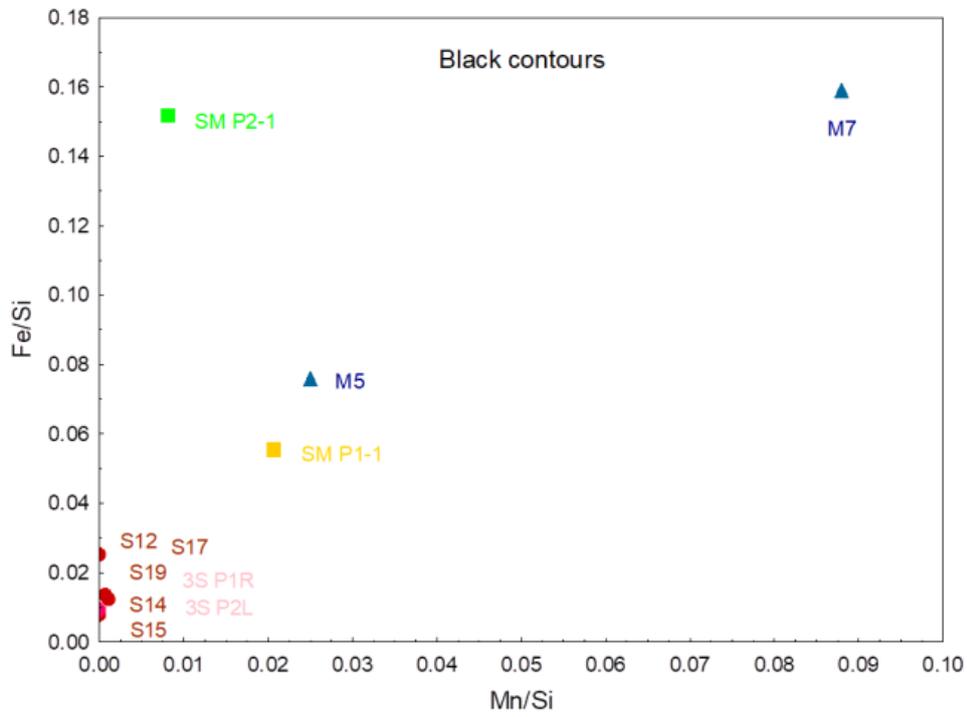
**Fig. 4:** Scatter plots of the weight % ratios of Al versus Si (upper) and Al/Si vs Ca/Si (bottom); see previous figures for symbols. Lines are guide for eyes.



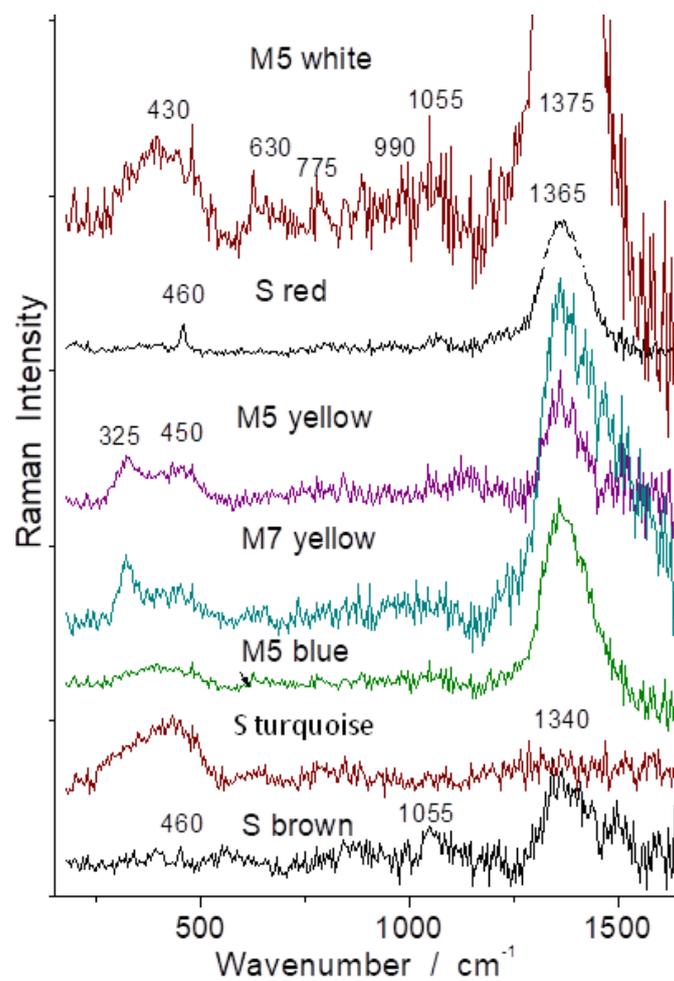
**Fig. 5:** Scatter plots of the weight % ratios of Co/Si versus Cu/Si (upper left), Co/Si versus Ni/Si (upper right), Co/Si versus Mn/Si (bottom left) and Bi/Si versus Co/Si (bottom right) measured on blue areas; see previous figures for symbols. Lines are guide for eyes.



**Fig. 6:** Scatter plots of the weight % ratios of Cu/Si versus Fe/Si (upper) and versus Cr/Si (bottom) measured on turquoise and green areas; see previous figures for symbols. Lines are guide for eyes.



**Fig. 7:** Scatter plots of the weight % ratios of Fe/Si versus Mn/Si (upper) and versus Cr/Si (bottom) measured on black contours; see previous figures for symbols. Lines are guide for eyes.



**Fig. 8:** Representative Raman spectra recorded on Seliminye (brown, turquoise and red area) and Muradiye (M5 yellow, white and blue and M7 yellow area) tiles

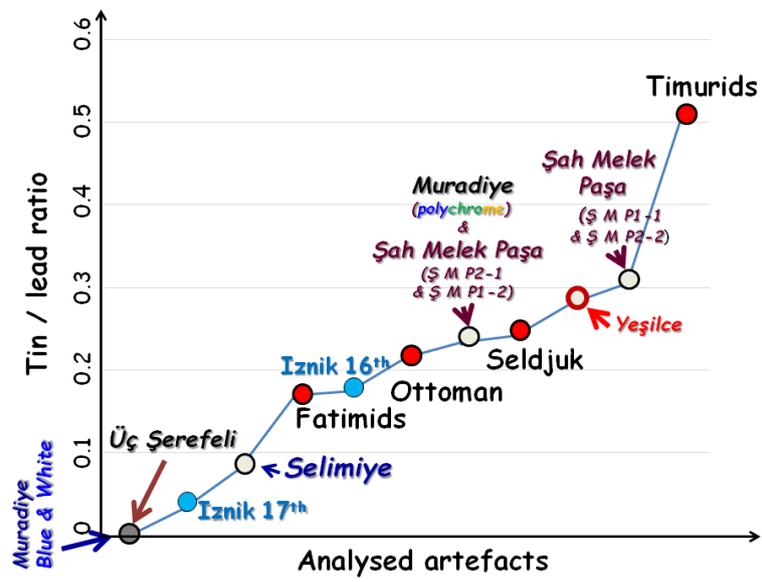


Fig. 9: Evolution of the Sn/Pb [reference data obtained from 2,3,41]



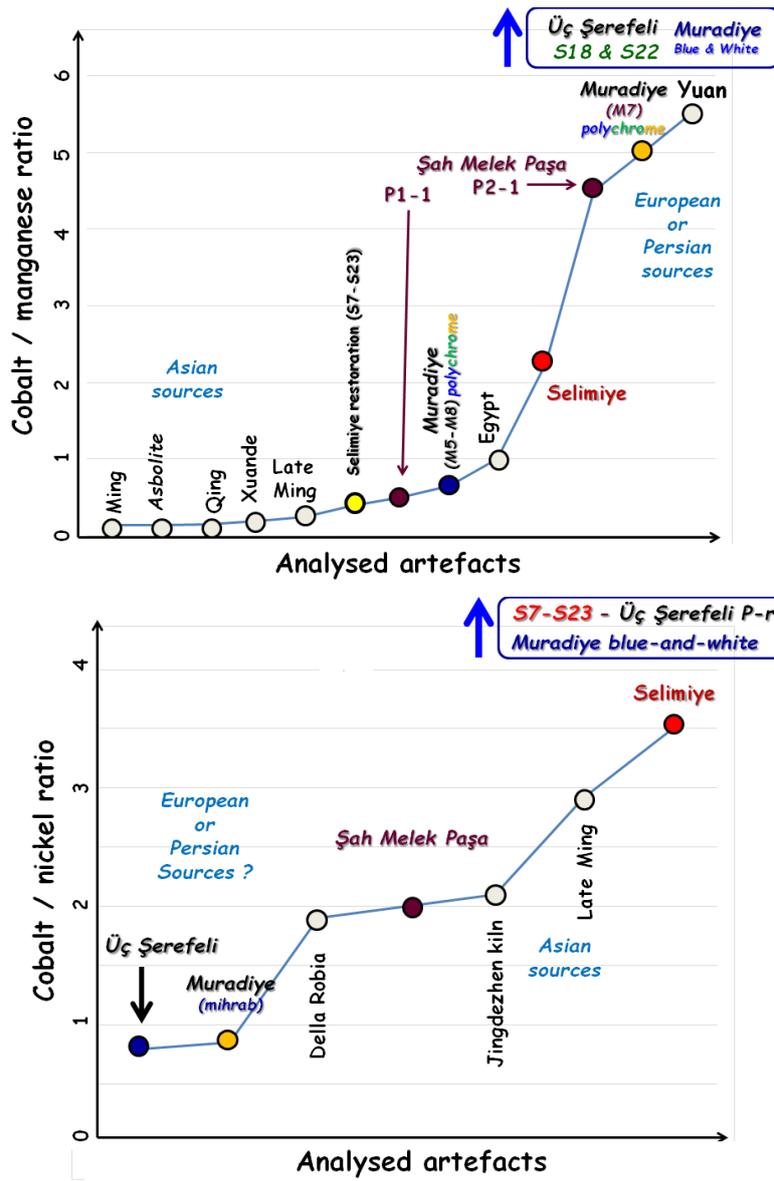


Fig. 10: Evolution of the Co/Mn and Co/Ni ratio [reference data obtained from 76-83]

## **Supplementary Materials**

### **On-site pXRF analysis of glaze composition and colouring agents of “Iznik” tiles at Edirne mosques (15<sup>th</sup> and 16<sup>th</sup> centuries)**

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## 1. Introduction to the Ottoman Ceramic Art discussed with the tiles studied

Nowadays, in Kütahya, there are three buildings which have coloured glazed tile decorations, where the production can be related to the period of Beyliks (13th -early 15th-century).<sup>[1]</sup> Some hexagonal, turquoise tiles and gilded decorated ones are similar; technically and compositionally to the tiles of Green Complex in Bursa and border tiles of Şah Melek Paşa Mosque in Edirne [1]. There were also some blue and brown painted underglaze tiles seen in the *mihrab* of Hisarbey Mosque (Kütahya), which represents the practice of the underglaze technique by the craftsmen of Kütahya at the end of 15th-century. Border tiles of Muradiye were decorated in the same manner, but the colours used were blue and white, unlike Hisarbey Mosque.<sup>[1]</sup> According to Necipoglu and Golombek, tiles of Muradiye have a similar design with the porcelain tiles of Ming used in the Chini-Khaneh (porcelain room) pavilion which was built by Ulugh Beg in 1430 in Samarqand and now it has been destroyed.<sup>[2,3]</sup> Necipoglu also mentions that blue-and-white tile bodies of Muradiye contain lime-alkali frit rather than lead-alkali that Iznik technology uses.<sup>[2]</sup>

After a gap of production of 50 years, tiles with coloured glazes once again decorated the buildings of Suleiman the Magnificent. Gülru Necipoğlu<sup>[2]</sup> states that a group of immigrant craftsmen from Iran with local assistants were working on the production of 15th-century tile revetments in an unknown ceramic workshop in Istanbul. They were supervised by one of the Tabrizi craftsmen whom Selim I had brought from Tabriz to the Ottoman court following the victory of the battle of Chaldiran in 1514.<sup>[4,5]</sup>

The coloured glaze decoration, where a non-vitreous line surrounds different areas of the decor serving to keep them separated during the firing. This ornamentation has spread in Central Asia from the end of the 14th-century to the Ottoman Empire during the 15th and the 16th-century and finally in Iran and India during the Safavid reign.<sup>[2, 3, 6, 7]</sup> Henderson and Raby<sup>[8-11]</sup> identified different phases: firstly, 13th-century Seldjuk pottery linked to Turkistan Iranian fritware technology,<sup>[12-15]</sup> but analysed fragment from the Gök Madrasah in Sivas dated to 1271 reveals that it had a clay body, with no trace of frit.<sup>[8]</sup> Then Masters of Tabriz blue-and-white productions are identified, e.g., at Üç Şerefeli (Three Balconies) Mosque, Edirne; Henderson stated that *laboratory analysis has shown that there was considerable technical variation and, most significantly, that they did not use lead frit". The Masters of Tabriz artists who reintroduced the fritware technique into Turkey, but the stonepaste composition they used, was not consistent. It differed visibly in colour, ranging from a brick colour in the 1420 to a buff white for their underglaze tiles in the 1430s.* [8]

On the other hand, J. Soustiel<sup>[16]</sup> noted that Muradiye and Üç Şerefeli tiles do not follow the time sequence proposed by A. Lane<sup>[17]</sup> and underlined the high specificity of these tiles. A stylistic similarity does not imply the same place of production, due to 'à la mode' style being copied in many places. Most of the analytical studies have been focused on the body and slip examination. Comprehensive analyses of the glazed decoration are still little known, and the literature stays confused about it, a limited number of shards have been studied, and their date of production are not always well established. The object of this work is to compare characteristics related to the

glaze production of tiles during the 15th and first half of the 16th-century. One of the big advantages of on-site measurement of building decor is that the production date of the materials is much more documented than for the tableware ceramics.

## 2. History of the mosques

*Şah Melek Paşa Mosque* (1429): This mosque was built by Şah Melek on the road that goes to Edirne from Gazi Mihal Bridge in 1429. It is a single-dome type mosque. Inside of the mosque was remarkable with tiles covered walls through the windows. These tiles were identical to the tiles in Green Mosque and Green Tomb in Bursa city as well as being the Ottoman's not-well-investigated artefacts remaining from the 15th century. Most of the tiles are fallen, and some geometric decor made of plaster was painted over the tiles that are covering *mihrab* (altar). These six-cornered turquoise coloured tile plates which are preserved on the left part of the *mihrab* (see Fig. 1 in the text and Fig. S1d), remind us of the Ishak Fakih tombs of Kütahya city dating back from the Germiyanids (1300-1429).<sup>[18]</sup>

*Muradiye Mosque* (1435-1436): The mosque was constructed by an unknown architect at the behest of Murad II.<sup>[3]</sup> Two large walls covered with blue-and-white hexagonal (~25.5 cm in diameter) and turquoise triangle tiles (see Fig. 1 in the text and Fig. S1a) are located on both lateral sides of a central polychrome *mihrab* made with a technique close to *cuerda seca*. The *mihrab* of Edirne Muradiye mosque was constructed in a similar way to the Bursa Green mosque *mihrab* but with an even more sophisticated design.<sup>[19]</sup> This *mihrab* is the largest preserved in Turkey. The designs of blue-and-white tiles are varied: undulated vines with flowers, palmettes (a decor characteristic of Seldjuk art)<sup>[16]</sup> or arabesque leaves, all in the Chinese manner.<sup>[16,20]</sup> We did not measure tiles with Persian design (rosettes) as well as the tiles of modern production which were installed instead of the original tiles stolen in the 2000s. The rectangle (Fig S1d) is put in relief with an edge made of a narrow border of tiles decorated in blue underglaze on white in a pattern of undulating vines of Chinese style. Measurements have been made in the lower part, close to the columns supporting the arch (Fig. S1d). Riefstahl considered from the visual examination that both *mihrab* and hexagonal tiles were made by the same "group of workmen".<sup>[20]</sup>

*Üç Şerefeli (Three Balconies) Mosque* (1410-1447): This building is one of the most important mosques among the Ottoman culture heritages. According to the Rifat Osman's "Rehnuma" and Ahmet Badi's "Riyazi Belde-i Edirne" works, building of this mosque was started in 1410 during Musa Çelebi (2<sup>nd</sup> Sultan who ruled in Edirne Palace), continued but could not be completed during the successor Sehzade Mehmet Celebi's period, and finally completed in 1447 during the Murad II period under the Elvend Mirza Bey's responsibility.<sup>[21,22]</sup> The inscription in tall *thuluth* characters painted on blue or on white, depending of the arch-panel is a dedication to the Sultan Mehmed Han and his son Murad and that in Kufic characters a sentence on Heaven. The design is recognized as similar to "Golden Horn" style.<sup>[23]</sup> The cartouche border is decorated in the Chinese manner.

*Yeşilce Mosque* (1440-1441): Originally named as Mezid Bey found in the Mezid Bey district, Hatice Sultan Street, was built as a masjid in 1440-1441. Afterward, it was transformed into a mosque by building a *minbar* (mosque pulpit) by Hasan Efendi who was sent to Edirne as a successor by Uskudardari Aziz Mahmud Hudayi. Half of one minaret was collapsed due to a big earthquake in 1752, and only half of it was remained. <sup>[21]</sup> It has salient rare turquoise tile adornments around the cylinder of only minaret of the mosque are located towards to the basement wall of the minaret (Fig. S1e), some of them are fallen off, and are now inventoried by Selimiye Foundation Museum (see in the text, Fig. 1).

*Selimiye Mosque* (1569-1575): It was built by Sinan, the Grand Architect of Ottoman Empire, between 1569 and 1575 by the order of Sultan Selim, II. With this masterpiece, Sinan brought the central and single dome plan to the highest level, not only the Ottoman architecture but also the architecture of the world at that time. <sup>[18]</sup> Selimiye, which is the summit of the attempts to reach space integrity, outdistanced sultans, including Suleymaniye, ordered all the mosques and it left behind the Hagia Sofia, too. With this feature, the mosque made a new record in art history. <sup>[24]</sup> Measurements have been made on the tiles decorating the Sultan balcony (Fig. S1b).

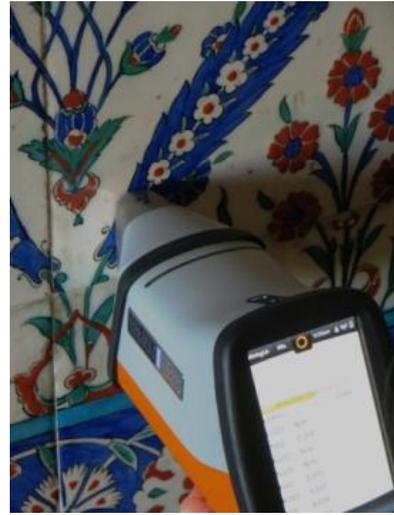
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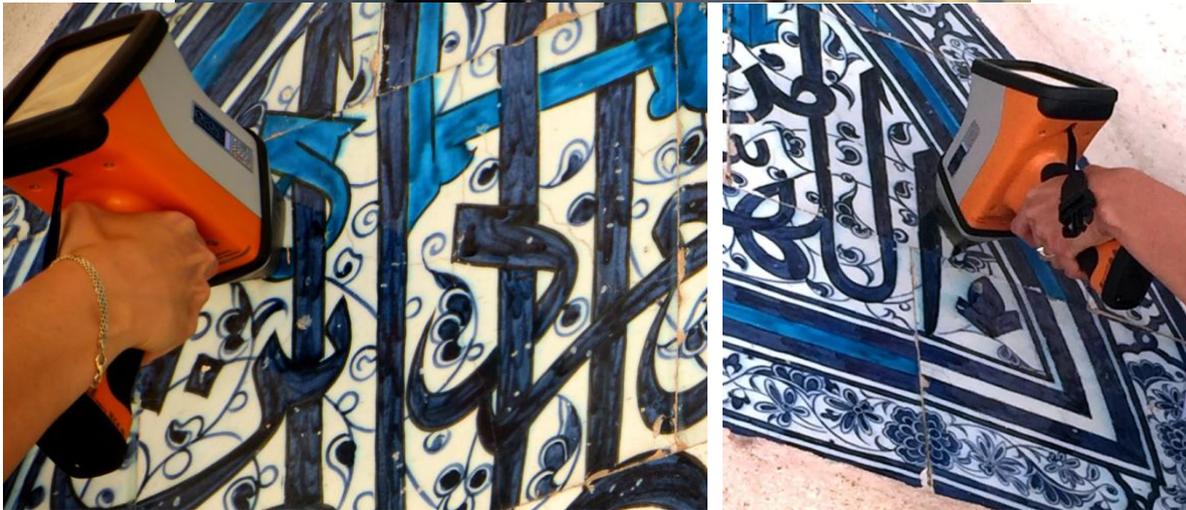
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**Fig. S1a:** On site measurement at Muradiye Mosque.



**Fig. S1b:** On site measurement at Selimiye Mosque.



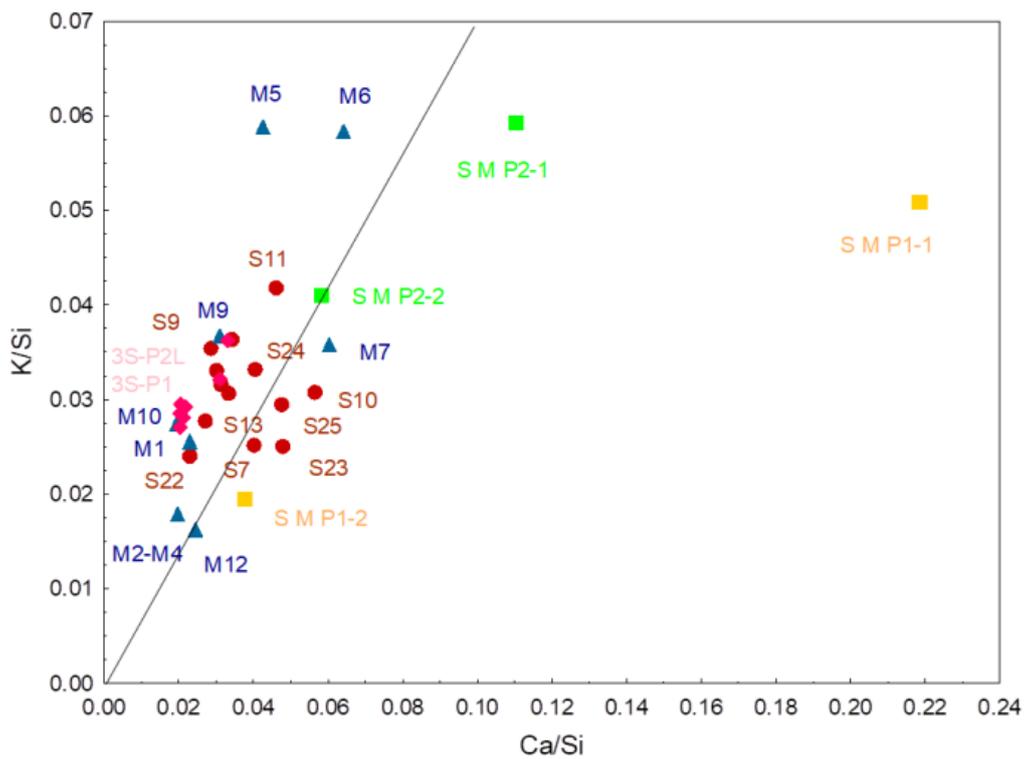
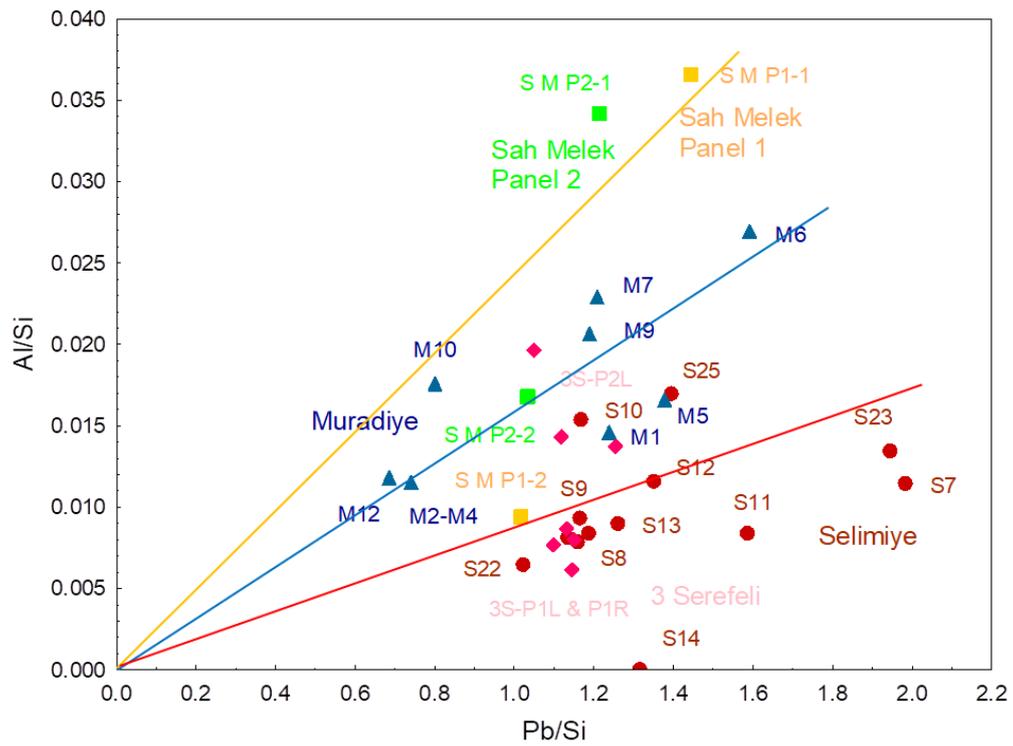
**Fig. S1c:** On site measurement at Üç Şerefeli Mosque.



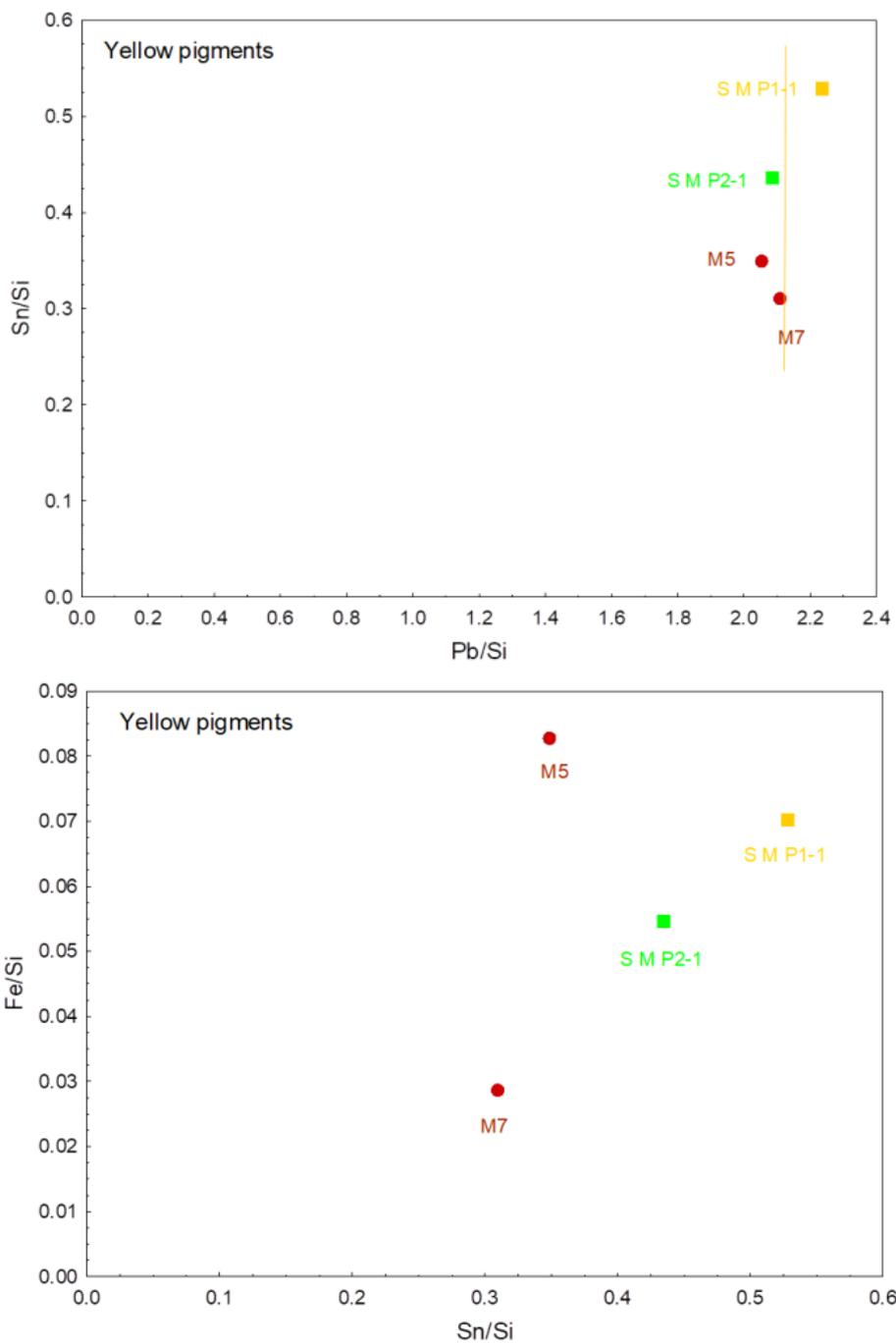
Fig. S1d: On site measurement at Şah Melek Mosque.



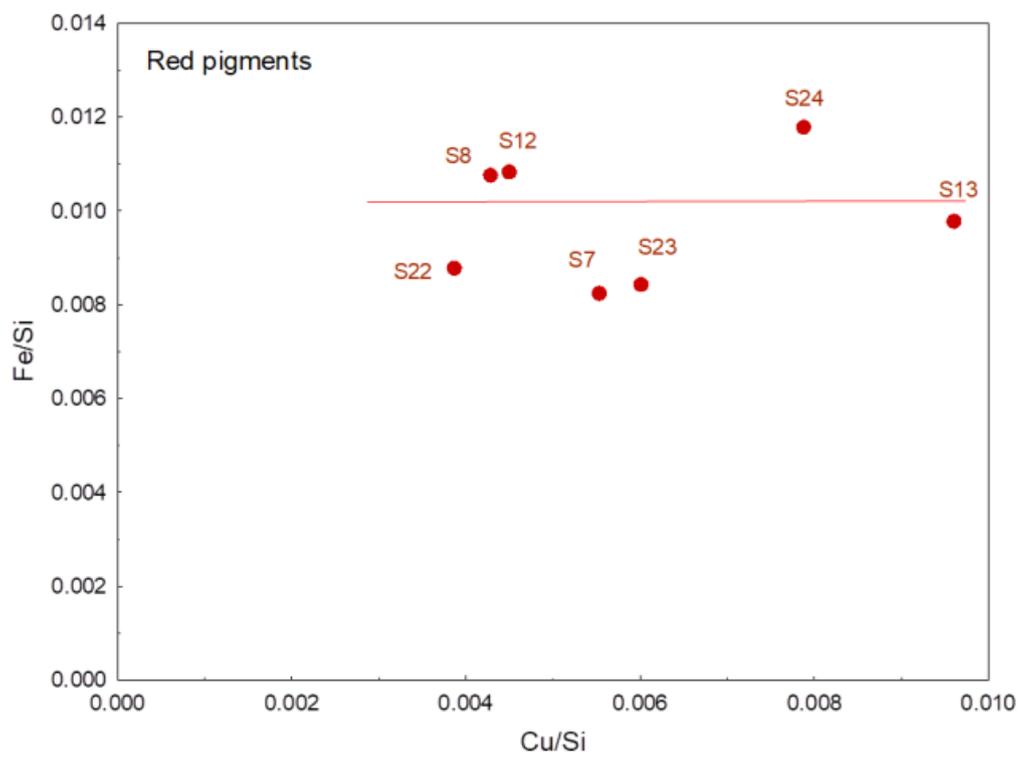
**Fig. S1e:** View of the glazed tiles on Yeşilce Mosque minaret.



**Fig. S2:** Scatter plots of the weight % ratios of Al/Si versus Pb/Si (top), and K/Si vs. Ca/Si (bottom measured on yellow areas; see text for symbols).



**Fig. S3:** Scatter plots of the weight % ratios of Sn/Si versus Pb/Si (top), and Fe/Si vs. Sn/Si (bottom) measured on yellow areas; see text for symbols.



**Fig. S4:** Scatter plots of the weight % ratios of Fe/Si versus Cu/Si measured on red areas; see text for symbols.

**Table S1.** pXRF results of the glaze & decors (wt%)

<b>Blue decor</b>	Si	Fe	Co	Ni	Cr	Cu	Zn	Mn	V	Sn	Pb	Bi	Ba	S	Ca	Ti
S7	21.226	0.186	0.013			0.170	0.005	0.034	0.345	0.066	42.600	0.078	0.041			
S8	25.637	0.306	0.050	0.014		0.173	0.009	0.032	0.242	2.495	31.093	0.130	0.097			
S8	25.270	0.332	0.079	0.025	0.047	0.292	0.010	0.032	0.264	2.526	31.858	0.188	0.081			
S9	24.821	0.285	0.028	0.007		0.213	0.008	0.019	0.255	2.487	32.207	0.128	0.064			
S10	24.880	0.216	0.029	0.007		0.182	0.005	0.018	0.240	2.246	30.249	0.094	0.099			
S11	25.057	0.277	0.065	0.017	0.034	0.204	0.012	0.025	0.248	2.264	31.245	0.156	0.040			
S11	20.350	0.318	0.102	0.026		0.301	0.009	0.031	0.221	2.418	40.849	0.207	0.146			
S18 contour	20.675	0.375	0.341	0.096	0.038	0.293	0.009			2.913	36.586	0.632	0.225			
S20 contour	25.963	0.227	0.050	0.012	0.030	0.188	0.004	0.019	0.232	2.207	29.518	0.145	0.051			
S22	27.205	0.251	0.130	0.031		0.132	0.004		0.235	2.307	28.161	0.207	0.065			
S23	21.315	0.185	0.018			0.177	0.005	0.034	0.338	0.053	42.141	0.092	0.052			
M2	28.558	0.287				0.655		0.015	0.234	0.232	27.546	0.060				
M5	25.462	1.665	0.267	0.277		0.701	0.005	0.386	0.239	4.081	26.398	0.092	0.053			
M7	28.637	1.188	0.283	0.317	0.030	0.645		0.037	0.202	4.904	20.230	0.054	0.039			
M7	24.104	0.622	0.114	0.100		0.299	0.004	0.055	0.248	6.892	27.966	0.073	0.062			
M8	32.576	0.300	0.007	0.006		0.586		0.011		0.156	19.448	0.046	0.074			
M9	18.568	0.348	0.007	0.012		0.915	0.004		0.208	0.258	31.495	0.091	0.132	3.180	8.881	0.121
M10	28.022	0.409				0.550			0.213	0.068	28.640	0.054	0.078			
M10	31.078	0.311				0.626			0.212	0.060	21.883	0.043	0.073			
M11	28.378	0.475	0.058	0.074		1.204	0.005			1.622	20.073	0.040	0.061			
M12 - intersection	31.704	0.324				0.384			0.212	0.049	21.782	0.062	0.035			
ŞM-1	19.436	0.901	0.258	0.130		0.366	0.005	0.280		7.096	24.497	0.056	0.051	4.606	4.945	0.163
ŞM-1 contour	21.360	1.008	0.060	0.033		0.119	0.004	0.371		7.182	24.264	0.054	0.035	2.803	4.199	0.222
ŞM-2 contour	27.486	1.191	0.567	0.251		0.770		0.178		3.288	16.714	0.046				
<b>Blue decor</b>	Si	Fe	Co	Ni	Cr	Cu	Zn	Mn	V	Sn	Pb	Bi	Ba	S	Ca	Ti

ŞM-2 contour	27.231	1.618	0.557	0.263		0.800	0.003	0.416		2.967	15.865	0.048	0.025			
ŞM-2	29.627	1.063	0.582	0.270		0.812		0.070		2.169	13.442	0.037				
3Ş-P1L	27.132	0.242				0.592	0.005	0.012	0.259	0.038	30.262	0.054	0.044			
3Ş-P1L	27.569	0.252				0.447		0.015	0.265	0.068	30.195	0.056	0.051			
3Ş-P1L	31.131	0.839	0.020	0.024		0.110	0.006			0.028	7.901	0.017	0.057	1.777	4.058	0.061
3Ş-P1R	26.969	0.255				0.734	0.004		0.262	0.053	30.782	0.058	0.051	0.555		
3Ş-P1R	19.608	1.109		0.013	0.041	0.183	0.007			0.026	16.311	0.027	0.078	13.546	6.775	
3Ş-P1R	26.324	0.240				0.537	0.003		0.271	0.044	32.767	0.065	0.067	0.583		
3Ş-P1R	27.232	0.237		0.010	0.033	0.590			0.256	0.057	30.564	0.046	0.048	0.541		
3Ş-P1R	27.250	0.249				0.489			0.249	0.053	29.887	0.049	0.039	0.539		
3Ş-P2L	27.027	0.266	0.008	0.009		0.521	0.004		0.261	0.039	29.987	0.059	0.073	0.733		
3Ş-P2L	25.300	0.292	0.010	0.016	0.046	0.705		0.016	0.266	0.053	33.225	0.075	0.104	0.772		
3Ş-P2L	26.551	0.271				0.489	0.004	0.013	0.254	0.069	30.546	0.069	0.062	0.881		
3Ş-P2L	26.622	0.261			0.310	0.440	0.003		0.253	0.058	30.670	0.058	0.063	0.692		
3Ş-P2L	26.839	0.261				0.406			0.258	0.089	30.721	0.058	0.057	0.741		

<b>Turquoise</b>	Si	Fe	Cu	Cr	Co	Ni	Mn	Zn	V	Sn	Pb	Ba
S7-green	20.523	0.2	1.771	0.136								
S8-green	25.916	0.328	2.039	0.092								
S9- contour	26.486	0.247	2.144	0.037	0.011		0.016	0.006	0.23	2.368	27.423	0.071
S25	23.106	0.358	2.687		0.041	0.009	0.026	0.006	0.262	2.16	32.221	0.062
S-bordure	27.556	0.208	1.367				0.015	0.003	0.232	2.332	26.359	0.084
M1	23.058	0.265	2.475						0.25	7.814	28.573	0.043
M5	22.043	1.589	1.858			0.016	0.756			8.972	26.52	0.09
M6-green	16.979	1.845	0.813									
M6	20.465	1.119	1.386				0.555		0.257	8.61	29.68	0.051
M6	19.377	4.622	1.31				3.074		0.203	6.486	26.937	

<b>Turquoise</b>	Si	Fe	Cu	Cr	Co	Ni	Mn	Zn	V	Sn	Pb	Ba
M7-green	19.34	0.315	1.32									
M7-green	19.356	0.311	1.309	0.037								
M7	20.731	1.05	1.728				0.152	0.007	0.245	7.587	30.915	0.033
S M P1-2	25.394	0.197	2.448				0.014		0.222	6.148	25.352	0.032
S M P1-2	25.026	0.201	2.522		0.009		0.02		0.221	6.378	26.006	0.042
S M P1-1	18.058	0.758	2.234			0.008	0.477			9.883	27.951	0.052
S M P2-1	18.812	1.176	1.871	0.03			0.541		0.229	9.452	30.049	0.043
S M P2-2	20.321	0.256	3.356	0.037						8.426	28.206	
S M P2-2	30.365	0.231	1.645		0.008		0.018	0.011		5.083	15.005	0.056
S M P2-2	23.701	0.273	3.4		0.011	0.005	0.015		0.224	7.223	26.847	0.041
S M P2-2	21.077	0.23	4.057	0.037	0.01				0.228	8.936	28.689	0.048
3Ş-P1L	25.53	0.266	3.243						0.258	0.036	31.363	0.059
3Ş-P1R	25.026	0.264	4.278		0.006		0.015		0.253	0.048	30.565	0.041
3Ş-P1R	27.264	0.258	0.533		0.012	0.014			0.254	0.049	29.866	0.058
3Ş-P2L	25.091	0.275	3.421						0.256	0.052	30.748	0.07
2008-82	13.278	0.505	2.14	0.061		0.008	0.042	0.012	0.207	11.047	39.937	0.235
2008-83	2.289	0.593	3.455	0.064	0.023	0.011	0.049	0.01	0.299	16.984	59.089	0.229

<b>Green</b>	Si	Fe	Cu	Cr	Ni	Mn	Zn	V	Sn	Pb	Ca
S7	20.523	0.2	1.771	0.136		0.03	0.006	0.348	0.055	41.949	0.881
S8	25.916	0.328	2.039	0.092	0.006	0.033	0.004	0.23	2.232	28.016	0.785
M6	16.979	1.845	0.813		0.01	0.951			5.844	34.365	3.593
M7	19.34	0.315	1.32			0.027	0.004	0.219	4.083	38.187	0.954
M7	19.356	0.311	1.309	0.037		0.021	0.003	0.223	4.115	37.949	0.995

<b>Violet</b>	Si	P	K	Ca	Ti	Mn	Fe	Rb	Sr	Zr	Sn	Sb	Ba	Tl	Pb	Al
M6	20.552	0.048	0.865	1.032	0.233	1.644	2.006	0.054	0.046	0.042	5.167	0.023	0.043	0.103	32.188	0.482

M7	26.543	0.049	0.807	1.446	0.118	2.021	0.509	0.039	0.067	0.031	4.103	0.015	0.05	0.068	22.974	0.502
M7	23.445	0.023	0.766	1.575	0.257	0.815	0.382	0.044	0.031	0.036	7.901	0.024	0.059	0.076	26.486	0.61
3S-P2L	27.121	0.028	0.772	0.76	0.086		0.266	0.048	0.008	0.04	0.039	0.066	0.102	0.101	30.083	0.46

<b>Yellow</b>	Mg	Al	Si	P	K	Ca	Ti	Mn	Fe	Sn	Sb	Pb	Bi	Se	Rb	Sr	Zr
M5	1.273	0.546	16.63	0.02	0.764	6.205	0.416	0.661	1.374	5.803	0.021	34.138	0.1	0.011	0.05	0.026	0.042
M7	0.808	0.437	18.492	0.048	0.719	1.354	0.433	0.18	0.529	5.737	0.031	39.016	0.131	0.012	0.059	0.014	0.054
SM-1	0.671	0.71	15.427	0.164	0.712	0.381	0.519	0.519	1.081	8.154	0.068	34.496	0.098	0.016	0.057	16	0.048
SM-2	1.364	0.718	17.144	0.329	0.623	1.643	0.43	0.16	0.609	7.376	0.063	38.269	0.107	0.008	0.058	0.014	0.045
SM-2		0.734	18.1	0.298	0.786	2.169	0.39	0.645	1.314	7.964	0.067	35.227	0.093	0.015	0.058	0.019	0.047

<b>White</b>	Mg	Al	Si	K	Ca	Ti	Rb	Sr	Zr	Sn	Ba	Pb	P	Se	Tl
S7	1.075	0.288	21.692	0.52	0.823	0.133	0.067	0.015	0.058	0.048	0.043	41.286		0.016	0.106
S7	0.834	0.244	21.454	0.512	0.839	0.117	0.069	0.017	0.059	0.066	0.057	42.204			0.111
S8	0.474	0.158	25.694	0.858	0.775	0.152	0.054	0.012	0.048	2.446	0.058	30.966			0.096
S9	0.444	0.338	25.902	0.858	0.781	0.145	0.052	0.011	0.041	2.327	0.047	30.363	0.019	0.007	0.094
S10	0.38	0.31	26.161	0.772	1.157	0.172	0.044	0.014	0.04	2.278	0.06	29.41	0.131	0.008	0.091
S22		0.163	27.861	0.643	0.579	0.13	0.049	0.008	0.043	2.337	0.058	28.009		0.008	0.082
S23	0.896	0.26	20.582	0.546	0.951	0.164	0.06	0.012	0.057	0.075	0.067	40.762		0.013	0.107
M3		0.35	29.116	0.691	0.676	0.053	0.046	0.006	0.037	0.2	0.04	26.191		0.004	0.076
M4	0.363	0.326	31.734	0.489	0.545		0.037	0.006	0.031	0.061	0.056	21.543		0.008	0.069
M4	0.492	0.389	31.89	0.477	0.613		0.033	0.007	0.03	0.062	0.043	20.996		0.005	0.059
M5	0.729	0.331	19.489	1.199	0.997	0.401	0.057	0.013	0.046	12.269	0.115	30.332	0.019	0.007	0.078
M5	0.801	0.364	20.846	1.266	0.606	0.345	0.05	0.012	0.041	12.623	0.053	29.11	0.013	0.008	0.079
M7	0.855	0.426	21.922	0.874	1.012	0.348	0.049	0.015	0.04	10.99		27.861	0.013	0.015	0.078
M10		0.618	31.425	0.705	0.519		0.037	0.008	0.034	0.056	0.054	22.15		0.003	0.069
<b>White</b>	Mg	Al	Si	K	Ca	Ti	Rb	Sr	Zr	Sn	Ba	Pb	P	Se	Tl
SM-1	0.68	1.011	23.88	0.769	2.993	0.235	0.037	0.01	0.032	7.141	0.046	22.802	0.149	0.008	0.064

SM-2	0.617	0.411	22.677	0.901	1.295	0.337	0.047	0.01	0.038	8.532	0.04	29.313	0.022	0.01	0.074
SM-2	0.412	0.424	23.027	0.932	0.933	0.252	0.044	0.01	0.042	8.658	0.036	29.32	0.015	0.007	0.071
3Ş-P1L		0.219	27.779	0.708	0.569	0.068	0.053	0.007	0.042	0.067	0.045	30.054		0.008	0.083
3Ş-P1L		0.175	25.691	0.869	0.675	0.092	0.059	0.011	0.047	0.07	0.053	33.616		0.008	0.083
3Ş-P1R		0.199	27.887	0.777	0.573	0.072	0.052	0.009	0.043	0.051	0.074	29.499		0.005	0.094
3Ş-P1R		0.135	27.049	0.731	0.555	0.061	0.054	0.009	0.046	0.044	0.048	31.42		0.01	0.103
3Ş-P1R	0.494	0.238	27.797	0.672	0.524	0.047	0.046	0.01	0.042	0.059	0.054	29.643		0.005	0.081
3Ş-P2L	0.606	0.363	27.452	0.744	0.594	0.066	0.05	0.009	0.043	0.032	0.044	29.759		0.014	0.09
3Ş-P2L	0.368	0.336	27.611	0.718	0.684	0.074	0.056	0.01	0.042	0.046	0.077	29.794		0.009	0.075
3Ş-P2L	0.401	0.541	27.154	1.017	0.859	0.124	0.048	0.013	0.044	0.086	0.093	29.418	0.033	0.009	0.073
3Ş-P2L		0.284	27.943	0.67	0.573	0.052	0.051	0.01	0.042	0.062	0.06	29.615		0.014	0.088
3Ş-P2L		0.273	27.259	0.815	0.592	0.075	0.047	0.01	0.043	0.073	0.056	30.861		0.01	0.088
3Ş-P2L	0.545	0.47	26.936	0.885	0.872	0.095	0.049	0.011	0.044	0.063	0.075	30.02	0.022	0.015	0.083
3Ş-P2L	0.566	1.178	29.106	0.844	1.341	0.143	0.035	0.014	0.037	0.055	0.081	21.448	0.082		0.071

Red	Si	Mg	Fe	Sn	Pb	Cr	Cu	Se				
S7	21.834	0.551	0.18	0.085	41.95		0.121					
S8	27.065	0.381	0.291	2.202	28.401	0.034	0.116	0.007				
S12	21.417		0.291	3.576	38.551	0.038	0.129	0.006				
S12	26.115		0.223	2.562	30.142		0.085	0.007				
S13	25.532	0.419	0.276	1.973	32.12		0.174	0.015				
S13	25.428		0.26	1.954	32.216		0.317	0.015				
S22	27.356	0.568	0.24	2.258	28.091		0.106	0.006				
S23	21.592	0.921	0.182	0.047	41.022		0.13	0.014				
S24	23.801	0.514	0.329	2.669	33.001		0.199	0.014				
S24	27.214	0.422	0.272	2.308	27.548		0.203	0.007				
S24	5.22	1.651	1.509	0.154	57.62	0.065	0.074					
Black line	Mg	Al	Si	P	Cr	Fe	Mn	Co	Ni	Cu	Zn	Ca
S12	0.542	0.427	25.354	0.016	0.058	0.342	0.018			0.255	0.01	0.912

S14			24.954	0.019		0.225		0.051	0.015	0.462		0.783
S15	0.715	0.279	22.871	0.056	0.19	0.361	0.32		0.013	0.115	0.014	1.015
S15		0.907	17.525	0.119	0.1	0.44				4.256	0.012	4.283
S17	0.466	0.156	27.286	0.102		0.214		0.014	0.008	0.177		0.652
S19		0.305	23.625	0.069		0.346	0.025			1.172	0.007	1.139
S19	0.426	0.349	24.014	0.043	0.058	0.291	0.033		0.005	1.073	0.007	0.823
S19	0.743	0.231	21.331		0.036	0.192	0.029	0.017	0.007	0.313	0.006	0.833
S19	1.076	0.218	21.234		0.036	0.181	0.028			0.29	0.005	0.875
S19	0.604	0.21	25.051	0.112		0.292	0.0027		0.007	0.282	0.019	0.956
S19	0.401	0.329	25.956	0.171		0.295	0.02	0.012		0.294	0.012	0.876
M5	0.479	0.443	25.79	0.04		1.951	0.645	0.25	0.274	0.558	0.004	1.049
M7	0.688	0.831	19.64	0.052		3.121	1.73			0.389		2.215
SM-1		0.43	15.048	0.092	0.039	0.834	0.312	0.025	0.013	2.112		5.037
SM-2	0.474	0.807	17.328	0.239		2.493	1.39		0.012	0.433		2.616
SM-2		0.808	19.867	0.154	0.034	3.152	1.644	0.304	0.141	0.5		3.806
SM-2	0.455	0.784	20.12	0.236		3.005	4.406	0.022	0.03	0.36	0.003	2.239
SM-2	0.875	1.172	17.886	0.264		4.299	3.537		0.021	0.183	0.005	3.664
3S-P1R		0.196	27.422			0.239				0.396	0.004	0.565
3S-P2L		0.367	27.39			0.259				0.178		0.571

Glaze	Mg	Al	Si	K	Ca	Ti	Rb	Sr	Zr	Pb	Sn
S4	0.48	0.213	26.219	0.801	0.881	0.144	0.051	0.013	0.045	29.767	2.351
S7	0.751	0.243	21.259	0.533	0.859	0.124	0.068	0.014	0.057	42.175	0.064
S8	0.454	0.203	25.916	0.856	0.787	0.137	0.052	0.012	0.045	30.067	2.38
S9	0.518	0.24	25.736	0.909	0.737	0.135	0.049	0.013	0.042	29.998	2.394
S10	0.47	0.391	25.521	0.784	1.445	0.183	0.044	0.014	0.041	29.829	2.262
S11	0.627	0.19	22.704	0.949	1.051	0.194	0.059	0.016	0.054	36.047	2.341
S12	0.539	0.282	24.337	0.884	0.833	0.186	0.055	0.014	0.048	32.905	2.828
S13	0.419	0.229	25.48	0.706	0.697	0.135	0.048	0.012	0.046	32.168	1.963

S14			24.954	0.787	0.783	0.184	0.06	0.01	0.048	32.832	2.846
S22	0.59	0.177	27.474	0.66	0.6314	0.125	0.047	0.009	0.04	28.087	2.301
S23	0.916	0.285	21.227	0.531	1.02	0.154	0.065	0.012	0.058	41.308	0.058
S24	0.468	0.214	25.507	0.845	1.035	0.2	0.049	0.008	0.04	30.275	2.489
S25	0.656	0.392	23.106	0.68	1.102	0.207	0.054	0.012	0.046	32.221	2.16
M1	0.435	0.336	23.058	0.587	0.535	0.173	0.05	0.009	0.041	28.573	7.814
M2-M4	0.427	0.355	30.913	0.552	0.611	0.053	0.039	0.006	0.032	22.91	0.108
M5	0.765	0.344	20.793	1.221	0.889	0.346	0.049	0.015	0.042	28.654	7.936
M6	0.655	0.521	19.343	1.128	1.242	0.336	0.047	0.04	0.043	30.792	6.527
M7	0.665	0.524	22.939	0.819	1.389	0.264	0.045	0.024	0.038	27.725	7.166
M9	0.491	0.493	23.928	0.877	0.745	0.092	0.048	0.011	0.04	28.472	0.235
M10		0.53	30.175	0.826	0.592		0.044	0.009	0.036	24.224	0.061
M12		0.372	31.704	0.513	0.779		0.033	0.008	0.032	21.782	0.049
SM-P1 Multi color	0.625	0.689	18.868	0.958	4.124	0.267	0.044	0.014	0.039	27.246	8.29
SM-P1 Mono turq		0.237	25.21	0.489	0.955	0.209	0.045	0.01	0.037	25.679	6.263
SM-P2 Mono turq	0.51	0.399	23.866	0.976	1.396	0.235	0.043	0.011	0.035	24.687	7.417
SM-P2 Multi color	0.636	0.738	21.609	1.278	2.385	0.281	0.042	0.028	0.036	26.228	6.144
3Ş-P1L	0.689	0.214	26.819	0.782	0.589	0.079	0.052	0.011	0.044	30.928	0.057
3Ş-P1R-1	0.66	0.209	27.428	0.78	0.564	0.07	0.052	0.009	0.045	30.141	0.052
3Ş-P1R-2	0.494	0.166	27.148	0.731	0.557	0.067	0.055	0.01	0.045	31.128	0.049
3Ş-P1R-3	0.722	0.23	26.693	0.785	0.555	0.075	0.054	0.009	0.042	30.22	0.052
<b>Glaze</b>	Mg	Al	Si	K	Ca	Ti	Rb	Sr	Zr	Pb	Sn
3Ş-P2L-1	0.474	0.385	26.949	0.755	0.571	0.081	0.051	0.009	0.042	30.128	0.045
3Ş-P2L-2	0.443	0.354	25.827	0.934	0.857	0.119	0.057	0.011	0.047	32.459	0.067
3Ş-P2L-3	0.514	0.537	27.352	0.877	0.848	0.102	0.047	0.011	0.042	28.744	0.068
2008-82	0.949	0.431	13.278	1.438	3.077	0.311	0.079	0.022	0.068	39.937	11.047
2008-83			2.289	1.413	1.642	0.34	0.12	0.028	0.093	59.089	16.984

**Table S2.** Ratio of Sn versus Pb comparing the tiles of the study with the literature [2, 3, 41]

Tiles studied [2, 3]	Sn/Pb	Sn/Pb	Literature study [41]
Üç Şerefeli, S7, S23, M2-M4, M9, M10	~ 0	0.169	Fatimid (960-1160 AD): Egypt-Fustat
17 <sup>th</sup> century Ottoman tiles (ref)	0.033	0.242	Seljuk (1050-1200 AD): Iran-Kashan
Selimiye	0.081	0.508	Timurid (1400-1440 AD): Uzbekistan
16 <sup>th</sup> century Ottoman tiles (ref)	0.175	Sn: n.d.	Turcoman (1380-1500 AD): Iran Tabriz
Muradiye & Şah Melek Paşa (Ş M P2-1, Ş M P1-2)	0.234	0.215	Ottoman (1480-1700 AD): Iznik
Şah Melek Paşa (Ş M P1-1 & Ş M P2-2)	0.305		

**Table S3.** Ratio of the elements (Co versus Mn, Cu, Ni and Bi versus Co) found in the blue decor (n.a.: not available)

	Co/Mn	Co/Cu	Co/Ni	Bi/Co
Selimiye	S7, S23: <b>0.4</b>		S7, S23: <b>n.a.</b>	S7, S9, S23: <b>4.5-6</b>
	S18, S22: <b>n.a.</b>	S18, S22: <b>~1</b>		
	S: <b>1.5-3</b>	S: <b>0.1-0.3</b>	S: <b>3-4</b>	S: <b>1.5-3</b>
Muradiye	M5, M8: <b>0.65</b>	M5, M7: <b>0.4</b>	M2, M10, M12: <b>n.a.</b>	M5, M7, M11: <b>0.2-0.6</b>
	M7: <b>5</b>	M8, M9, M11: <b>0.01-0.004</b>		M8: <b>6.5</b>
			M: <b>0.6-1.1</b>	M9: <b>13</b>
Şah Melek Paşa	S M P1-1: <b>0.1-0.9</b>	S M P1-1: <b>0.6</b>	S M P1-1: <b>1.9</b>	S M P1-1: <b>1.9</b>
	S M P2-1: <b>1-8</b>	S M P2-1: <b>0.7</b>	S M P2-1: <b>2.1</b>	S M P2-1: <b>2.1</b>
Üç Şerefeli	3Ş P1-L: <b>n.a.</b>	3Ş P1-L: <b>0.1</b>	3Ş P1-L: <b>0.8</b>	3Ş P1-L: <b>0.8</b>
	3Ş P1-R: <b>n.a.</b>	3Ş P1-R: <b>n.a.</b>	3Ş P1-R: <b>n.a.</b>	3Ş P1-R: <b>n.a.</b>
	3Ş P2-L: <b>n.a.</b>	3Ş P2-L: <b>0.01</b>	3Ş P2-L: <b>0.8</b>	3Ş P2-L: <b>7.5</b>