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Tracing the Provenance of Unfired Ancient Egyptian Clay Figurines from Saqqara through Non-Destructive X-Ray Fluorescence Spectrometry

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Abstract

A collection of Prime Cultural Heritage artefacts consisting of Egyptian late Middle Kingdom figurines (c. 1850-1700 BCE), made of unfired clay and covered in inscriptions, is kept at the Royal Museums of Art and History (RMAH) in Brussels, Belgium. Several of these hieratic inscriptions curse enemies of the Egyptian state, including Canaanite, Nubian and Libyan entities; thus providing invaluable information for Middle Bronze topography in ancient Near Eastern studies. What makes the extensive Brussels group even more exceptional, is the fact that these figurines were discovered in a closed archaeological context in Saqqara, Lower Egypt. Defining and classifying clay and ancient ceramic provenance groups in Egypt is highly complex due to variability in Nile and marl clay deposits. Chemical characterization of this figurine assemblage (n=91) allows to effectively study the potential direct use of clays in the vicinity of Saqqara and characterize the nature of the raw materials used. Additionally, it aims to assess the validity of portable XRF spectrometry for this type of materials and its provenance resolution. Analyses show that very similar clays were used to produce the different morphological types of figurines in the assemblage. The chemical profile indicates the use of a mixed source at Saqqara rather than pure marl or Nile clays, which are common for ancient pottery production.

Keywords: clay figurine characterization; unfired clay objects; non-destructive chemical analysis; ancient Egypt; Portable XRF; clay provenance

1. Introduction

An exceptional collection of Egyptian late Middle Kingdom figurines (c. 1850-1700 BCE), made of unfired clay and covered in inscriptions, is housed at the Royal Museums of Art and History (RMAH) in Brussels (Fig. 1). The inscriptions express curses aimed at (potential) enemies of Egyptians or the Egyptian state as a whole, such as Levantine, Nubian or Libyan entities (Posener, 1940; Van der Perre et al., forthcoming). In general, such execration figurines are badly preserved because they were ritually broken, intentionally burned or buried, in order to symbolically neutralize the enemy and offer protection against dangerous forces (Ritner, 1993). The Brussels group of figurines was discovered in a closed archaeological context, namely a cenotaph near the Teti pyramid at Saqqara North, Lower Egypt (Fig. 2) (Posener, 1940; Abdalla, 1992).

Being crucial primary sources for our knowledge of ancient onomastica and the political geography of the ancient Near East and Egypt, they are regarded as Prime Cultural Heritage artefacts, but a significant part of the figurines is in a poor state, due to the fragility of the unfired clay. Therefore, urgent measures need to be taken to ensure their optimal further preservation, including a comprehensive analysis of the material and its surfaces. Facing the problem of a lack of reference frameworks for the study of unfired clay objects from Egypt (see below 2.), the present work offers the results of the initial analysis of the clay composition. As the clay is unfired, the objects have moreover the potential to directly correlate these materials with clay sources. Due to the nature of these objects, supposedly being disposable objects, it is generally assumed that local and easily accessible clay sources were used. Two research questions pertain to the material composition and require analytical techniques. Firstly, while the history of the discovery suggests that all figurines were discovered in a single place of origin, a characterization of the composition of the clay is required to determine whether the material confirms the use of a single clay source. The second research question concerns the homogeneity of the group. The Brussels figurines were divided in five groups based on a morphological typology and show potential for paleographical classification as well: their features are shaped differently and several handwritings can be distinguished in the inscriptions (Posener, 1939). It was therefore decided to determine whether the morphological typology subdivision is mirrored in the clay composition.

Since the figurines are museum objects and extremely brittle, the use of (micro)destructive methods is not an option. Non-destructive and in situ analysis by portable X-Ray Fluorescence (pXRF) was preferred as the most suitable method to assess this relative visible homogeneity, based on an analysis of mid to heavy Z elements in clay and ceramics studies (Padilla et al. 2006; Degryse & Braekmans 2014). As an additional concern, several conservation or consolidation treatments have been applied in the past but, unfortunately, these had been hardly documented (Van der Perre et al., forthcoming).

2. Clay raw materials and Egyptian ceramic provenance studies

In Egyptian archaeology, the raw clays used for ceramic production are generally divided into three groups: Nile silts, marl clays and kaolin clays. Both Nile silts and marl clays were used throughout the Pharaonic period in the entire Nile Valley, while kaolin clays were mainly used from the early Roman period onwards (Bourriau & Nicholson 1992; Bourriau et al. 2000; Nordström & Bourriau 1993). Nile silts, i.e. clays deposited by the river between the Upper Pleistocene and the present, are rich in silica and iron and appear grey to black in their raw state. Marl clays, originating from shales and limestone found along the river, are calcareous and rich in mineral salts (Bourriau et al., 2000). However, typical marl clays with good properties for pottery production are especially confined to the area around Qena, Upper Egypt (Arnold &Bourriau, 1993). These clay raw materials were used to create a wide variety of fabrics. They were often mixed, for example Nile silt mixed with carbonate grains or general carbonate/marl components. A non-site specific classification system, the so-called Vienna System, is generally used to identify the particular fabric type of ceramics (Arnold & Bourriau, 1993).

The current challenge with ceramic provenancing, especially in Egypt, is related to the composition of the clay. On the one hand, most minor and trace elements are not distributed homogeneously in the clay beds. Secondly, clays were often selected from heterogeneous clay deposits (Rapp & Hill, 2006). Another issue is that mainly fired pottery samples are used for provenance studies, while it can be complicated to connect these to local clay beds. Clays were often prepared before being used; impurities were removed, different clays were mixed, or a temper was added. Moreover, the effects of firing can influence the compositional nature of the materials (Kilikoglou et al., 1988). Finally, the (surface) composition pattern of the clay can be influenced by reactions during the burial of the analyzed objects and cause mobility of certain elements. For example, many Egyptian ceramics exhibit a so-called scum layer (Ownby & Griffiths 2009). Whereas local characteristics and variations can often be detected by visual analysis, this generally cannot determine the exact place of production. Although provenancing clearly requires scientific analyses, these are still rarely applied in Egyptian pottery studies (Schiestl & Seiler, 2012a). The available targeted geochemical provenance studies on Egyptian objects and clay mineral resources treat various areas and periods and highlight the difficulty in sourcing these materials (Spataro et al. 2018). An increasing amount of studies succeed in determining an exact provenance of a ceramic assemblage in Egypt, especially when analyzing the output of a local production center (Allen et al., 1989; Hamroush & Zeid, 1990; Ballet & Picon, 1990).

By analyzing unfired clay objects, the ultimate objective of this research is to provide additional information that allows to identify the use of mineral resources in the past and that contributes to the characterization of locally and regionally produced ceramic materials and clay beds. Since we are dealing with unfired clay objects, the main challenges – as opposed to ceramic studies – are: 1) the lack of a significant reference framework compared to pottery studies and the latter less relevant due to changes that might occur during and after firing; and 2) the fragile state of unfired surfaces and, subsequently, their sensitivity to external influences and interventions (conservation and preservation). With this study, we therefore aim to evaluate if meaningful differences can be identified through non-destructive chemical analysis of the unfired objects, related to technology and, potentially, their provenance. Secondly, this study seeks to provide an assessment on the possibility to distinguish different sources of Nile clay and/or marl clay and to establish whether natural or intentional mixtures of clay were utilized for these particular unfired objects.

3. The geological setting of Saqqara

Geological investigations in the region of Saqqara have been the scope of several studies and described in detail (Said, 1975; 1990; Moustafa, 1988; Papa 2003; Youssef et al. 1984; Hamdan et al. 2018; Stanley & Jonathan G. Wingerath 1996; Hamdan 2000; Hamdan et al. 2014). The site of Saqqara itself is locatedon a plateau consisting of mainly Upper Eocene limestone, marl and claystone. The Eocene Limestone (Mokattam Formation) substrate is bordered by Pleistocene sediments to the north and south, while more Eocene limestone (Maadi Formation) and Pliocene sediments can be found to the west. The floodplain Nile alluvial sediments are located to the east. The Saqqara-Abu Sir Plateau – exposing the Upper Eocene Saqqara Member and overlooking the Nile floodplain – consists of laminated alternating layers of light yellow, hard limestone and yellow marl with layers of marly to sandy limestones and shale at the top. Older strata include Upper Eocene marly shale deposits including several gypsum bands, overlain by calcareous layers of the Saqqara Member (Yehia, 1985). Also, the Late Pliocene Helwan Formation (Said, 1975; 1990) consists of fine-grained sandstone and shale. Apart from the Nile floodplain and the limestone plateau, a cuesta scarp is located at the eastern side of the plateau. Attested quaternary sediments in the immediate area contain mostly sand and gravel with thin layers of Nile silt (Hamdan et al. 2014; Hamdan 2000). Especially floodplain sediments can be influenced by variations in paleoclimatic and paleoenvironmental developments throughout the Holocene, more notably around the collapse of the Egyptian Old Kingdom (ca. 2100 BCE). Recently, however, geochemical data from floodplain deposits at Saqqara has become available for this specific Middle to New Kingdom period. These deposits range from sandy to clayey silt and are enriched in carbonate and manganese concretions (Hamdan et al. 2018).

4. Material

Preliminary research on the clay was conducted using a trinocular microscope Leica M80 with 8:1 zoom range and magnification of 7.5x-60x, identifying the unfired clay as mainly Nile silt. In addition, selected fracture surfaces were examined in detail with a digital microscope with polarizer (Dino-lite Edge with 14:1 zoom ratio and magnification range of 10x-140x) (Fig. 3).

The general appearance of the clay suggests the use of a generic Nile silt for the manufacturing of these figurines (Van der Perre et al., forthcoming). The color of the unfired clay corresponds to Munsell codes 5YR 5/1-5/2-5/3. The figurines contain several inclusions in a fine-grained matrix, visible with the naked eye and approximately measuring 250-500 µm, suggesting temper might have been added to the raw material. All figurines contain mineral inclusions, while a vast amount contains inclusions of ashes and organic material, sometimes carbonized. Reed particles, chaff, carbonized barley and wood splinters are very common, even remnants of small (grape) and large seeds (Fabaceae family) are discernable (Van der Perre et al., forthcoming). Small voids in the clay are either the result of the degradation of organic inclusions or an irregular drying rate, indicative of a lack of preparation and blending of the clay. A large number of small, grey-white granules can be identified as carbonate particles but require further analyses. The edge of these inclusions appears smooth and rounded, lacking sharp corners, suggesting that the granules were not added as a temper but are part of the original raw material. Crushed sherds are also present, in some figurines only as small (~150-250 µm) orange-brown inclusions, while other figurines contain large fragments (c. up to 2 cm, Van der Perre et al., forthcoming, fig. 8). Although the identification is often difficult, several figurines have small shell fragments as well.

Based on the quantity of, and the variation in the inclusions, the composition of the clay categorizes as Nile B1 or Nile B2 in the Vienna System (Arnold & Bourriau, 1993). Even though the raw clay used for pottery is basically the same as the raw clay of the figurines, the former will have been modified by adding temper or by levitation to adapt it to the specialized needs of the potter. The figurines, however, did not require such preparation. The most relevant characteristic of the clay is its plasticity: the clay cannot be too plastic because that increases the risk of cracking due to excessive shrinkage (Hope, 1978). Although this resembles the adding of a temper by potters, the (intentional) addition of chaff and/or sand merely avoids cracking during drying, not during firing.

In regard to the surfaces, two types of pigment were used for the inscriptions. The majority of the figurines (90%) is inscribed with a red ochre ink (hydrated iron oxides), while a carbon-based black pigment was used on the other objects (Blom-Boër, 1994; Lee and Quirke, 2000; Van der Perre et al., forthcoming).

5. Methodology

Studies on the utilization of handheld (or portable) X-ray fluorescence (hXRF/pXRF) for geological and archaeological materials also point out the drawbacks of the technique (Forster et al. 2011; Goren et al. 2011; Hunt & Speakman 2015; Müskens et al. 2017; Young et al. 2016; Barone et al. 2011; Rowe et al. 2012; Pincé et al. 2018; Frahm 2018; Shackley 2011; Speakman et al. 2011; Shugar & Mass 2012). Furthermore, few of these studies have successfully employed pXRF to address other clay-based materials in Egypt (Emmitt et al. 2018; Morgenstein & Redmount 2005; Emery & Morgenstein 2007). X-ray fluorescence (XRF) entails the emission of characteristic "secondary" (or fluorescent) X-rays from a material that has been excited by bombarding with high-energy X-rays. The phenomenon is used for elemental analysis of solids, powders or liquids. While the advantages of its portability and non-destructive nature are clear for vulnerable museum objects, it is important to gauge the effectiveness of the technique for the various elements measured on the surface of the ceramic objects. Therefore, the analysis of matrix matched standards and an assessment of precision and accuracy are invaluable for any pXRF study. Nevertheless the application of an energy dispersive technique allows for a relative comparison with other previous publications that employed XRF-based analysis of both ceramics and clays in Egypt.

Handheld equipment (Bruker Tracer III-SD) was used to determine the chemical composition of the clay of the selected figurines directly in museum context in order to identify (in)consistencies in this possibly unique assemblage. The instrument is equipped with an Rh anode X-ray tube and a Peltier-cooled Silicon Drift Detector (~145 eV at MnKa). Spot size is approximately in the range of $5mm^2$. Because of homogeneity considerations and the spot size of a pXRF device, analysis was concentrated on the flat backs of the figurines to achieve the most consistent bulk chemical data. The location of the measurements was carefully chosen on each figurine: the selected surfaces were clean of any post-depositional encrustations, visible inclusions or applied painted decoration. In general, the figurines themselves do not show a macroscopically heterogeneous texture and occasional visible larger grains were avoided. Matrix identifications of the different figurines are selected and can be described as exceedingly fine, thus offering potential for an adequate chemical determination of these clay materials. Post-excavation treatment products include the acrylic resin Paraloid B-72, diluted in Paraxylene. In most cases, it had been applied locally on these figurines and could thus be avoided (Van der Perre et al., forthcoming). However, some samples (E.7441, E.7444, E.7448, E.7472, E.7480) had been fully submerged in this resin. These were marked for possible additional further analysis of the obtained spectra if flagged as outliers. Measurements were taken in air for 120 seconds (de Winter et al. 2017), using a 150 μm Cu, 25 μm Ti, and 300 μm Al filter, with beam conditions of 40 keV and 14μA. An empirical custom ceramic calibration (through multiple pressed pellet matrices) was used to semi-quantitatively assess the composition of the samples. Multiple readings were carried out on several objects to assess possible differences due to potential heterogeneity within the assemblage. Certified rock and soil standards are also analyzed periodically to check accuracy, precision and monitor for any machine drift during the analyses. Prior to quantifying the spectra, all data was evaluated through Bruker proprietary ARTAX software to determine the consistency of the matrices. The following set of international certified standards (powder in pressed pellet form) was used for quality control of the data: BIR-1 (basalt), SRG-1 (shale), GSP-2 (granodiorite), 98b (clay) and CRM667 (sediment) (supplementary table 2). Only elements with relative high squared correlation coefficients (R^2) in the calibration were retained for subsequent analysis: Ca=0.98, Sr=0.96, Y=0.98, Ti=0.94, Mn=0.96, Fe=0.94, Ni=0.99, Zn=0.94, Zr=0.94, Cr=0.96, Rb=0.99, Nb=0.95 and K=0.94. Precision (both repeatability and reproducibility) of the measurements was controlled by a replicate analyses of standards 98b and CRM667 and is commonly assessed through the calculation of the relative standard deviation (%RSD) (Abzalov 2008). All elements are below 10% RSD, except for Ni (14.50): i.e. Ca (2.55), Fe (1.84), K (6.71), Mn (4.99), Sr (1.18), Ti (5.60), Zn (9.79), Nb (8.04), Rb (2.94), Y (9.50) Cr (4.75) and Zr (2.06). Based on these values, especially Ni does not provide adequate results for quantification and therefore needs to be discarded for further interpretation.

The measurements were evaluated by an assessment of absolute data through bivariate and ternary diagrams, as well as by means of commonly applied multivariate statistical procedures such as hierarchical cluster analysis (HCA, for sample clustering) and principal component analysis (PCA). These statistical procedures are widely employed in a chemometric approach and provide a way to investigate complex datasets and thus isolate and also refine reference groups of pottery production. Prior to statistical processing the data is also log base transformed.

This transformation allows for equal contribution of each element in chemometric procedures (Aitchinson 1986; Baxter & Freestone 2006). As a linkage method and distance presentation in HCA, it was opted to use hierarchical Ward's clustering and squared Euclidian distance (as described by Johnson & Wichern 2007). In contrast to other common methods, the distance between clusters is evaluated by an analysis of variance. Although cluster creation generally tends to be small-sized, this method endeavors to minimize the 'sum of squares' of any two (theoretical) clusters, formed at each step (Ward 1963). Principal Component Analysis (PCA) is widely applied as a pattern-recognition technique, by means of identifying subgroups in the compositional data. After PCA a visual output is often displayed in the form of a biplot. These represent reference axes as computed by the statistical procedure arranged by a decreasing order of variance. Both 'objects' and variables can easily be evaluated on the same set of axes. Therefore the contribution of specific elements to group separation can be observed together with the degree of variability. PCA is commonly both used as a tool to discover such subgroups or to assess the coherence of hypothetical groups suggested by other criteria, e.g. petrographic groups, archaeological context or decorational patterns,. For PCA, the log transformed data were analyzed through the covariance matrix. The optimal number of principal components can be assessed through the Krzanowski cross-validation (Eastman and Krzanowski, 1982). Factor analysis is also a main technique used for statistical analysis. This technique can simplify a complex data set by identifying several variables that might explain the dimensions associated with data variability as an added tool (Howarth & and Govett, 1983). The contribution (or loading) of each factor is evaluated on the basis of associations between each variable. A varimax rotation was applied to the initial factor loadings in order to maximize the variance of the squared loadings.

The obtained cluster data and variability assessment can subsequently be utilized as a comparison to other XRF studies on Egyptian (raw) materials. Recently few studies were successfully conducted with pXRF or ED/WD-XRF, and clusters can be verified and put in context with published reference data from clays and ceramics from the region of Saqqara and beyond (Goren et al. 2011; Hamdan et al. 2014; Hamdan et al. 2018; Redmount & Morgenstein 1996). Discriminant analysis will be further used to assess whether the identified typological groupings to which the chemical observations belong are distinct and if a systematic division occurs.

6. **Results**

pXRF analysis was conducted on 91 individual artefacts. Fragments too small or fragmentary to be assigned to a typological group were not taken into account for statistical analyses. First, the variability and consistency of the chemical composition of the dataset itself is provided through both statistical cluster procedures and bivariate diagrams. Reference studies dealing with chemically fingerprinting ceramics from Egypt utilized variable resolution techniques (ranging from ED-XRF to Neutron Activation Analysis - NAA) (Hancock et al. 1986; Morgenstein & Redmount 2005), which provided an extensive set of variable elements to explore for provenance fingerprints. In the past, both major element ratios such as SiO_2/Al_2O_3 as well as patterns involving heavy elements such as Th, Hf and U have been applied. Other studies pointed especially at the importance of strontium, titanium and zirconium for distinguishing Egyptian clays (McGovern 2000) as well as rubidium, strontium and iron for differentiating various Nile silt and marl deposits from other marine, volcanic or desert substrates (Morgenstein & Redmount 2005; Emery & Morgenstein 2007/1). Both light and very heavy elements are often difficult to detect and/or to quantify adequately by pXRF technology. As a result, focus will be drawn towards the feasibility of the Mid-Z elements (a.o. Sr, Rb, Zr, Ti, Y) for characterization of the assemblage. All statistical operations are based on log10 normalized data (Bishop & Neff 1989; Baxter 2003).

To provide an initial overview of potential compositional groups within this figurine assemblage, a hierarchical cluster diagram (Fig. 4) in combination with factor analysis was constructed incorporating all measured figurines. In this way, groupings as well as the characterization of elements responsible for this variability can be analyzed (Degryse & Braekmans 2014). Ward's agglomerative method was followed using squared Euclidean linkage distance. Only elements with relative consistent accuracy and precision results are retained: Ca, Fe, K, Sr, Ti, Zn, Nb, Rb, Y, Cr and Zr. As a result, three compositional groups are clustered together, labelled SEF (Saqqara Execration Figurines) I, SEF-II and SEF-III, representing three potential groups. Table 1 reports the mean and standard deviations of these groups.

A second step is to evaluate which elements contribute to this differentiation and are responsible for the proposed group classification. Subsequently, reliability of these groups is assessed through factor analysis. Each factor represents a potential cluster of correlated variables within the dataset and is used to detect the structure of variance gain information about the interdependencies. This information can be utilized to identify associations of elements and their relation to the geological substrate. Retained factors are those with eigenvalues greater than 1, as a factor needs to extract at least as much as the equivalent of one original variable, or is to be removed (Hakstian et al., 1982). As such, four factors pass this criterion in our dataset. The factor loadings, communalities and proportion of the variance explained by all determined factors are presented in table 2.

The factor analysis yielded four factors, explaining 72.6% of the total variance in the data. Factor 1 is comprised of high by positive loadings (> 0.70) for Fe_2O_3(T), TiO_2, Y and Nb. Small positive contributions from Zr and Rb are given. Factor 2 contains high positive loadings for CaO and Sr. Factor 3 is dominated by positive loadings for Cr and K_2O, while Factor 4 shows positive loadings for Zn and MnO. The variability of elements in the first factor seems to be controlled mainly by the presence of Fe-bearing minerals. This factor accounts for 33.7% of the total variance. Factor 2 and 3 are mainly controlled by the presence of respectively limestone and carbonate content, and feldspar and clay mineralogy. Since the study concerns unfired clay figurines, the alteration of Rb, Ca, K and Na would be less affected than it would be for ceramics fired above 800°C (Buxeda I Garrigós, 2001; Picon, 1991) and it could in this case also provide a direct link to the available clay raw materials.

Statistical clustering procedures defined the presence of three discrete cluster groups. Factor analysis determined the presence of three influential factors related mainly to a heavy mineral component, a limestone component, and the clay materials themselves. Chromium clearly separates SEF-III from the other two groups based on the higher abundance of Cr, generally concentrated above 200 ppm. SEF-I on the other hand is mainly defined on the bases of lower strontium values, situated below 300 ppm. Unfortuantely, when combined with TiO_2 for example, no clear pattern can be distinguished. Egyptian sediments are frequently enriched in Ti relating to the presence of detrital anatase and rutile (Takla & Arafa 1975; Schneiderman 1995).

In this graph, a large variability can be seen regarding the content of TiO_2, which is equally disparate for all groups ranging approximately between 1.0 to 2.2 wt.%. Apart from the Sr values, SEF-I also has systematically lower Zn values than encountered in the other two groups. Zinc content can vary widely in Egyptian soils, but is generally concentrated in heavy alluvial soils. In Egypt, the lowest amounts are found in calcareous and especially in sandy sediments (El-Kadi et al. 1973). While some overlap is clearly visible, both SEF-II and SEF-III contain generally more Rb and Zr than group SEF-I. Nile silt can contain a higher amount of Zr relating to a presence of aeolian sand into the floodplain silts. Elements such as iron (and the other major elements apart from CaO) show a close coherence between the various groups with only minor differences. The highest values are concentrated in group SEF-II and SEF-III while the lowest are related to group SEF-I.

In summary, three groups are established on the basis of all measured and semi-quantified data. A detailed analysis of the influence of the different elements show that SEF-III is separated on the basis of the consistent relative high Cr abundances and to a lesser extent Fe_2O_3 and TiO_2. Overall, SEF-I is more depleted in trace element content, while SEF-II has the highest CaO, K_2O and Sr values. Differentiation in trace and major elemental composition can thus be observed, especially regarding Fe_2O_3, CaO, Cr and Sr. Consequently, three statistical groups can be explained but they remain very closely connected and cannot be easily discriminated on the basis of sets of elemental bivariate diagrams. Therefore, the elemental composition of the assemblage itself points at one main resource group with several variations based on different ratios of clay and inclusions. These results will subsequently be compared to existing compositional and typological reference data.

7. Discussion

7.1 A comparison to Egyptian clay raw materials

The ceramics and clays of Egypt are well known for their relative homogeneity, belonging to one of the three large groups (Nile silt, marl or kaolin clay). Nile alluvium or Nile silt clays are generally calcium poor and have a fairly consistent composition throughout the entire Nile floodplain. These similar deposits are present since the Upper Pleistocene and therefore not only present in the current Nile valley, but in the entire floodplain (Bourriau et al. 2000; Michelaki & Hancock 2013). Other major deposits for clay production are lime-rich marl clays, which are more concentrated in Upper Egypt and especially at sites such as Qena and El-Ballas, alongside several silty Nile muds (Bakr 1956; Bourriau et al. 2000; Shortland 2000; Alfred & Harris 1962). Some studies on Egyptian (raw) materials were successfully conducted with pXRF and ED/WD-XRF and the obtained data from the figurines can be tentatively compared to these reference data of both Egyptian clays and ceramics (Goren et al. 2011; Hamdan et al. 2014; Hamdan et al. 2018; Redmount & Morgenstein 1996).

Reference values for the mineral resources and ceramics originate from various locations in the Saqqara-Memphis floodplain itself. In addition, selected clay cuneiform tablets from Amarna are added to the dataset as they are both concerned with XRF analysis of Egyptian clay sediments and ceramics (Hamdan et al. 2014; Goren et al. 2011). Relevant comparison materials to assess a local production of the figurines at Saqqara include ceramic reference data obtained from mixed "silt and gebel" and mixed silt-related modern ceramic material from Cairo and Badrashein respectively (Redmount & Morgenstein 1996), contemporary clay reference material from the recent SAQA 22 floodplain core at Saqqara (more specifically Unit IV) (Hamdan et al. 2018), and reference clays from the Saqqara-Memphis area (Hamdan et al. 2014) and Amarna (Goren et al. 2011).

The first two principal components explain ~75% of the total variance. The score plot of the PCA shows that all three figurine groups are correlated with each other (Fig. 5). A clear distinction can be made between all marl and Nile silt raw materials in Saqqara and beyond. When compared to the figurines studied here, all SEF groups are situated between the Nile silt and marl deposits at Saqqara, but especially seem to show similar values to the clays in the recent floodplain core and modern ceramics from nearby Badrashein. Based on the absolute values, the general characteristics of this cluster group would rather point in the direction of a Nile silt containing elements of marl and fits with the use of the deposits in the immediate surroundings of Saqqara. Hence, all figurines clustered in varieties SEF I-III appear to point at a (mixed) Nile silt origin of raw materials, while a pure marl source would be rather unlikely. The attested variability within the assemblage however, signifies the absence of a dedicated preparation of the clay bed utilized for ceramic production but might also incorporate different local varieties of Nile silt from the Saqqara floodplain. However, as noted by Riederer (1989), Bourriau et al. (2000), and more recently also discussed by Spataro et al. (2018), Old Kingdom Nile silt pottery at Dashur and Saqqara have been characterized by a high calcium carbonate content, which seems to be replicated here as well. Interestingly, contrary to those from Amarna, marl deposits from Saqqara are significantly enriched in rubidium, and the modern mixed ceramics from Cairo are heavily influenced by calcium carbonates.

Zn or Cr are not incorporated in these graphs because these elements were not provided in all the reference studies. Chromium, however, is the major element that distinguished SEF-III from the other two groups. Despite these differences, the general build-up of the figurines does not point at different clay sources, but rather at the use of one source with some inherent variability.

There is a significant consistency of the figurine data with the reference data obtained for Nile silt clays. However, the figurines itself prove to be enriched in calcium carbonate content, contrary to what is considered generic Nile silt clay geochemistry. Potters commonly mix and prepare Nile clays , but in the case of unfired figurines the shape might need to be consolidated by an additional temper or a mixture of Nile silt clay with calcium carbonates. Therefore, it would not be surprising that a clay from a deposit enriched in carbonate materials is added in minor quantities to an otherwise finer Nile silt composition. Alternatively, in the Nile valley to the south of Cairo, marl clays are found that relate to the cliffs and desert that encompasses the Nile valley (Redmount & Morgenstein 1996). These can provide a direct influx of carbonate material and thus a more natural mixture. As pointed out by other authors as well (Riederer 1989, Bourriau et al. 2000; Spataro et al. 2018), this data would also support the observation of a more carbonate enriched Nile silt material in the vicinity of Saqqara. This additionally has the potential to be a good marker for provenancing. Given the close geochemical consistency with both modern ceramics from Badrashein and contemporary Nile floodplain deposits, the data obtained for the figurines would suggest a localized production in the close vicinity of Saqqara within the Nile floodplain itself.

7.2 Typological considerations versus composition

At first sight, the RMAH assemblage contains two groups: large figurines (> 30 cm), carrying the nearly complete version of the execration texts (including references to non-Egyptian enemies), and a second group of smaller figurines (c. 10-15 cm), bearing references to Egyptian individuals and 'negative things' in general (Fig. 1). Based on the research conducted during the project (Delvaux et al., 2017), this general division was refined into five types: Types A-E, of which E contains two subtypes (E1-E2). This division in subtypes is based on morphological differences and, as such, not directly relevant for the analysis of the chemical composition of the assemblage. Therefore, the two subgroups were analyzed together as a homogeneous type, dubbed Type E.

As stated above, an additional research axis was to analyze whether the proposed typology is mirrored in the clay composition of the assemblage. The following analyses used the absolute compositional values and discriminant analysis to assess whether there is any differentiation in composition related to typological group attribution. A ternary diagram using iron, strontium and rubidium (Fig. 6) shows the composition of these typological groups opposed to published reference values from Hamdan et al. (2014; 2018), Redmount & Morgenstein (1196) and Goren et al. (2011). These elements are selected due to their previous successful compositional discrimination abilities in other parts of Egypt. All groups show very similar and consistent values. It is apparent that the general resources of all different typological groups are quite homogenous and point at a Nile silt with elevated amounts of carbonate materials. Nile silt clays and ceramics generally contain around 2.5-4 wt.% of CaO, while marl clays and ceramics around Saqqara contain above 10 wt.% CaO. While a marl source can be excluded, this diagram points at a (Saqqara) Nile silt origin, but richer in calcium carbonates than expected. Strikingly, it has a very close consistency with signatures obtained from modern pottery produced at nearby Badrashein as reported by Redmount and Morgenstein (1996) (fig 6).

Finally, discriminant analysis assesses whether these typological group denominations can be distinguished from each other based on the analyzed elements in this dataset (Fig. 7). Based on Fig. 7, it can be concluded that the morphological typology is not clearly mirrored in the clay composition. The only exception seems to be Type A, which has a chemical pattern that slightly differs from the others. These figurines, being noticeably larger (> 30 cm) and produced with greater care and skill, are thus not only a clearly identifiable type based on morphological characteristics, but also in clay composition. It is possible that the clay was processed in a slightly different way, since the size of the figurines required an adapted fabric plasticity. Differentiation between the other types is not as evident as in the case of the Type A figurines. A certain overlap is noticeable between the chemical composition of Types B and C, while the values of Type E are more spread out. Figurines in Type A can be distinguished from those in Types B and D, but not as clearly from certain figurines attributed to Types C and E.

There are several possible explanations for these variations, related to the composition of the raw material itself. Even within a single clay bed the chemical composition of the clay is never entirely homogeneous. When the clay was collected from another zone within the same clay bed, this is potentially mirrored in the composition. Another possible factor influencing the composition might be the time of collecting. It is possible that the production of the figurines took place at different moments in time, depending on certain ritual prescriptions. On the other hand, the minor differences may also reflect a different approach in clay preparation and processing, indicating the existence of different "workshops" or creators. These explanations need further research and present new avenues for research.

8. Conclusions

The overall goal of this project was to determine whether a single clay source was used for the production of these figurines, whether the morphological typology was mirrored in the clay composition and whether the raw materials are consistent with the production of local ceramics. The very nature of these figurines, being unfired, allows a direct comparison with clay raw materials and addresses the suitability of non-destructive XRF analysis on Egyptian ceramic materials in a museum setting.

In general, the chemical fingerprint of the figurines shows a fairly homogenous picture of similar (Nile) sediments, especially those analyzed in the Saqqara floodplain itself. Minor variations within the assemblage point at three different subgroups, but these variations appear limited and rather indicate that a similar type of clay raw material was used. The variations are moreover too discrete to clearly indicate distinct provenances. The chemical differentiation of groups SEF I-III can be explained by changes or modifications in clay preparation, mixing and processing, or by natural variations within the raw materials due to influx of carbonate deposits into the floodplain clays. The chemical characteristics based on discriminating elements as iron, rubidium and strontium indicate it is not likely that common marl or Nile silt clay sources for pottery production were utilized. Instead, the consistency in these compositions rather point at an untreated floodplain or 'mixed' resource.. This implies a mixture of Nile silt and marl clay, that is to sayNile silt to which calcium carbonates/marl clays have been intentionally added or which have naturally precipitated in the sediment. Whether this clay recipe is natural or man-made is difficult to assess at this point, but it must be kept in mind that extensive preparation or mixture procedures for these materials were probably deemed unnecessary because of the intrinsic nature of the figurines themselves, namely as objects to be ritually discarded. More likely, the raw materials derived from the periphery of clay sources, such as Nile silt clay with washed-in desert and sedimentary soil debris. Sources in the area of Saqqara can be enriched in carbonates. Recent analyses by Hamdan et al. 2018 was conducted within the floodplain on clay strata (Unit IV) contemporary with the production period of the figurines . The analyses show a high geochemical consistency and suggest that this resource might have been the main source for the production of the figurines under study. At this point, since chemical analyses are not frequently used in Egyptian pottery studies and the differences between the three groups are not visible with the naked eye, the clay used for all figurines remains classified as a Nile B1/B2 in the Vienna system.

Apart from the large figurines (Type A), which had been produced with more care, the chemical composition of the clay is hardly mirrored in the morphological typology. While for Type A figurines this might be linked to different clay processing due to the size of the figurines, this is not valid for the other types. The majority of the assemblage was made with a very similar clay, but even within the same morphological type the chemical pattern alters. On the one hand, this suggests that various types were made from the same batch of clay, and on the other hand that likely not all figurines were created simultaneously by one individual or workshop.

The paper also confirms the applicability of the presented approach for the characterization of unfired clay and the possibility for provenance determinations of these extremely fragile clay artefacts, especially those housed in museums, where sampling is not an option. While the restricted set of reference data for this particular assemblage currently impedes a direct one-on-one provenancing of the execration figurines, the applied approach does prove that a proper grouping of objects based on their chemical composition and general provenance is feasible and that it can point in the direction of a potential and suitable clay source. In this particular case all data confirms a consistent local production in Saqqara. Additionally, the data correlates well with the specific Saqqara clay deposits and modern Saqqara ceramics as previously determined, which contributes to the determination of a Saqqara geochemical signature.

In a later stage, when additional data might become available, e.g. more geological samples of specific regions, the obtained data can be compared and linked to more precise locations of the newly analyzed samples. Therefore, the importance of the presented assemblage for potential future ceramic and clay studies is evident.

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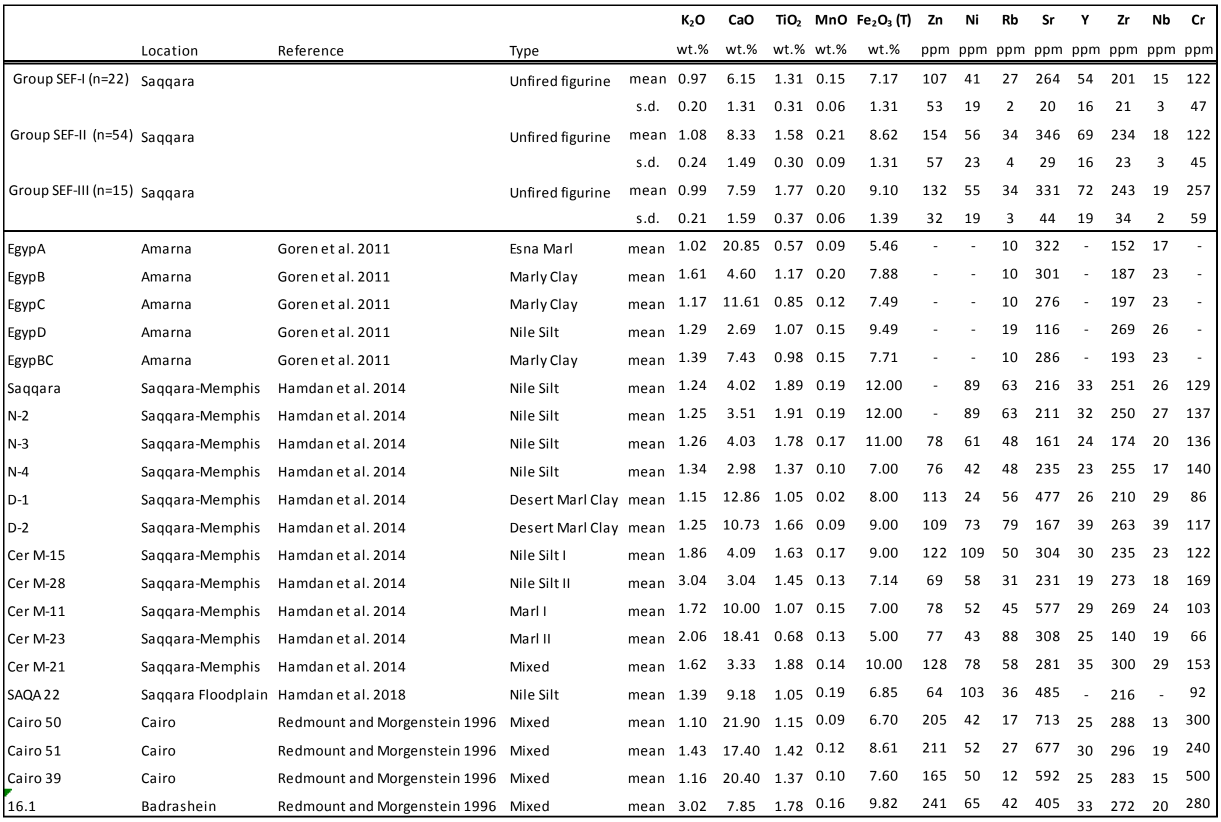

					K_2O	CaO	TiO_2	MnO	Fe_2O_3 (T)	Zn	Ni	Rb	Sr	Y	Zr	Nb	Cr
	Location	Reference	Type		wt.%	wt.%	wt.%	wt.%	wt.%	ppm	ppm	ppm	ppm	ppm	ppm	ppm	ppm
Group SEF-I (n=22)	Saqqara		Unfired figurine	mean	0.97	6.15	1.31	0.15	7.17	107	41	27	264	54	201	15	122
				s.d.	0.20	1.31	0.31	0.06	1.31	53	19	2	20	16	21	3	47
Group SEF-II (n=54)	Saqqara		Unfired figurine	mean	1.08	8.33	1.58	0.21	8.62	154	56	34	346	69	234	18	122
				s.d.	0.24	1.49	0.30	0.09	1.31	57	23	4	29	16	23	3	45
Group SEF-III (n=15)	Saqqara		Unfired figurine	mean	0.99	7.59	1.77	0.20	9.10	132	55	34	331	72	243	19	257
				s.d.	0.21	1.59	0.37	0.06	1.39	32	19	3	44	19	34	2	59
EgypA	Amarna	Goren et al. 2011	Esna Marl	mean	1.02	20.85	0.57	0.09	5.46	-	-	10	322	-	152	17	-
EgypB	Amarna	Goren et al. 2011	Marly Clay	mean	1.61	4.60	1.17	0.20	7.88	-	-	10	301	-	187	23	-
EgypC	Amarna	Goren et al. 2011	Marly Clay	mean	1.17	11.61	0.85	0.12	7.49	-	-	10	276	-	197	23	-
EgypD	Amarna	Goren et al. 2011	Nile Silt	mean	1.29	2.69	1.07	0.15	9.49	-	-	19	116	-	269	26	-
EgypBC	Amarna	Goren et al. 2011	Marly Clay	mean	1.39	7.43	0.98	0.15	7.71	-	-	10	286	-	193	23	-
Saqqara	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt	mean	1.24	4.02	1.89	0.19	12.00	-	89	63	216	33	251	26	129
N-2	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt	mean	1.25	3.51	1.91	0.19	12.00	-	89	63	211	32	250	27	137
N-3	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt	mean	1.26	4.03	1.78	0.17	11.00	78	61	48	161	24	174	20	136
N-4	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt	mean	1.34	2.98	1.37	0.10	7.00	76	42	48	235	23	255	17	140
D-1	Saqqara-Memphis	Hamdan et al. 2014	Desert Marl Clay	mean	1.15	12.86	1.05	0.02	8.00	113	24	56	477	26	210	29	86
D-2	Saqqara-Memphis	Hamdan et al. 2014	Desert Marl Clay	mean	1.25	10.73	1.66	0.09	9.00	109	73	79	167	39	263	39	117
Cer M-15	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt I	mean	1.86	4.09	1.63	0.17	9.00	122	109	50	304	30	235	23	122
Cer M-28	Saqqara-Memphis	Hamdan et al. 2014	Nile Silt II	mean	3.04	3.04	1.45	0.13	7.14	69	58	31	231	19	273	18	169
Cer M-11	Saqqara-Memphis	Hamdan et al. 2014	Marl I	mean	1.72	10.00	1.07	0.15	7.00	78	52	45	577	29	269	24	103
Cer M-23	Saqqara-Memphis	Hamdan et al. 2014	Marl II	mean	2.06	18.41	0.68	0.13	5.00	77	43	88	308	25	140	19	66
Cer M-21	Saqqara-Memphis	Hamdan et al. 2014	Mixed	mean	1.62	3.33	1.88	0.14	10.00	128	78	58	281	35	300	29	153
SAQA 22	Saqqara Floodplain	Hamdan et al. 2018	Nile Silt	mean	1.39	9.18	1.05	0.19	6.85	64	103	36	485	-	216	-	92
Cairo 50	Cairo	Redmount and Morgenstein 1996	Mixed	mean	1.10	21.90	1.15	0.09	6.70	205	42	17	713	25	288	13	300
Cairo 51	Cairo	Redmount and Morgenstein 1996	Mixed	mean	1.43	17.40	1.42	0.12	8.61	211	52	27	677	30	296	19	240
Cairo 39	Cairo	Redmount and Morgenstein 1996	Mixed	mean	1.16	20.40	1.37	0.10	7.60	165	50	12	592	25	283	15	500
16.1	Badrashein	Redmount and Morgenstein 1996	Mixed	mean	3.02	7.85	1.78	0.16	9.82	241	65	42	405	33	272	20	280

Table 1: Mean and standard deviations of the statistically defined groups in addition to selected reference materials from Goren et al. (2011), Hamdan et al. (2014, 2018) and Redmount and Morgenstein (1996).

	Factor - 1	**Factor - 2**	**Factor - 3**	**Factor - 4**
K_2O	0.317556	0.137589	0.755983	0.028258
CaO	-0.113705	0.931006	0.052177	0.04483
TiO_2	0.915737	-0.049439	0.073581	-0.027508
Cr	0.343731	0.110067	-0.702187	-0.073091
MnO	0.053622	0.111364	-0.095655	0.778606
Fe_2O_3 (T)	0.946322	0.00489	0.038389	0.135418
Zn	0.12317	0.172467	0.251327	0.734501
Rb	0.704904	0.231607	0.000074	0.371147
Sr	0.324723	0.753607	-0.027315	0.395016
Y	0.825307	0.158985	0.021735	-0.023666
Zr	0.585845	0.513799	-0.042001	-0.153702
Nb	0.79021	0.077895	-0.146456	0.17055
Expl.Var	4.235177	1.85933	1.170934	1.520024
Prp.Totl	0.352931	0.154944	0.097578	0.126669

Table 2: Eigenvalues of the factor analysis corresponding to the first four factors.