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Non-destructive provenance study of cuneiform tablets using portable X-ray fluorescence (pXRF)

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ABSTRACT

Portable X-Ray Fluorescence (pXRF) apparatus of the last generation was tested to determine its potential for routine provenance determination of clay cuneiform tablets, which cannot be analyzed by "classical" intrusive methods. A group of tablets from Hattuša (Boğazköy) and from el Amarna, which were previously provenanced using optical mineralogy (OM) and instrumental neutron activation analysis (INAA), was analyzed by pXRF and the results were used to establish the grouping according to their elemental concentrations. These groups were compared with the previous results retrieved by OM and INAA in order to confirm their validity. The results corroborate the high potential of the pXRF for non-destructive study of well-defined, 'closed' assemblages of clay-derived, delicate artifacts, such as cuneiform tablets, bullae, and fine-ware pottery. Consequently, a group of previously unexamined tablets from Hattuša was analyzed by pXRF and the results are discussed with implications on future research.

1. Introduction

During nearly three millennia, the civilizations of the ancient Near East (ANE) produced the world's greatest archives of written texts in hieroglyphs, cuneiform, and alphabets containing vast numbers of legal codes, administrative accounts, contracts, rituals, epics, letters, historical narratives, songs, dictionaries and scholarly texts. The crystallization of the great empires of Egypt, Hatti, Mitanni, Babylonia and Assyria during the second millennium BCE (Fig. 1) brought about their rise as the main political and economic powers of their time. After centuries of military conflicts, these superpowers established peaceful relations through a series of treaties and a network of trade relations. Hence, by the second half of the 2nd millennium BCE, international commerce grew to unprecedented levels, spanning lands from the Anatolian Plateau to the Nile valley and from the Argolid to the Euphrates. Along with these interactions and the traffic in commodities, cross-cultural contacts such as international correspondence and the exchange of epics, narratives and scholarly texts advanced an unprecedented transfer of ideas, contributing to a high level of communication between distinct cultures.

Over a century of research into these archives has accumulated an enormous body of data concerning all related aspects. At the same time, the interpretation of many documents still remains disputed, as the archives contain abundant tablets whose origin is unknown. Letters often contain the name of the sender, but sometimes the letterhead is missing. In other cases we may have the name of the sender and still do not know his domicile. Further complicating the issue, the locations of many Near Eastern and Aegean countries and cities have not yet been clearly established. When it comes to documents other than letters, the situation is even worse. Though tablets might be assigned to an origin according to their style or location of discovery, some uncertainty still remains in such determinations. Hence, revealing the origin of documents by using quantitative physical methods brings potential to shed new light on the geographical history, the development and the transfer of syllabic information and the diffusion of language and literature, scribal habits, narratives and epics between agencies and cultures within the ANE and beyond.

In theory, this goal can be accomplished through systematic provenance studies of clay of documents from archives of different parts of the ANE. Indeed, solving the problem of origin of cuneiform tablets by their clay identification can mark a significant breakthrough in our understanding of these documents. The use of methods adapted from natural and exact sciences provides an independent witness to the origin of the tablets that may be compared

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Fig. 1. Map of the Ancient Near East with the localities mentioned in the text.

with the data extracted from the texts. Scientific provenance studies of clay-derived artifacts in archaeology are focusing on their mineralogical and elemental composition in order to identify their provenance and the technology used in their production. This is based on, but not restricted to, optical mineralogy (OM, often dubbed petrography) for defining the geological context of the clay and temper minerals, and/or on instrumental neutron activation analysis (INAA) for measuring the elemental concentrations of the clay (Mommsen, 2004). In practice, however, these methods face several difficulties resulting from their intrusive nature. In fact, fearing such intrusion, museum curators have always been extremely reluctant to allow such studies. Moreover, the antiquities laws of several countries are extremely strict when it comes to the export of archaeological materials, be they complete artifacts or meager samples taken from artifacts. As a result, apart from two pioneering but rather limited studies made during the 1970s using INAA (Artzy et al., 1976; Dobel et al., 1977), no attempt has been made until the turn of the 21st century to investigate the source of large numbers of tablets on the basis of their raw materials.

A significant step forward may be found in the comprehensive study of the much discussed and disputed 14th century BCE Amarna archive by Goren et al. (2002, 2003a,b, 2004). The Amarna tablets were retrieved in Egypt in the late 19th century and have been under investigation ever since, with many issues remaining unresolved (*e.g.,* Moran, 1992). For example, the locality of many Canaanite rulers and several kings of independent states who wrote to the pharaohs was unknown or was debated among scholars. The OM study made it possible to locate many of these places and consequently, to suggest an overall reconstruction of the territorial disposition of the ANE, particularly Canaan, during the 2nd millennium BCE.

As a sequel to this study, other research projects were planned, applying the same methodology. Part of the Amarna project involved the study of the Cypro-Minoan texts from Enkomi and Kalavasos in Cyprus at the Cyprus Museum in Nicosia (Goren et al., 2003a, 2004). Southern Levantine tablets and other texts on clay were also analyzed (Goren et al., 2004, 2007, 2009; Na'aman and Goren, 2009; Mazar et al., 2010). In addition, Goren, Cohen and Kaufman studied a collection of syllabic, legal, administrative and scholarly texts from the archive of Ugarit (Ras Shamra, on the north Syrian coast) along with a few letters now kept in the *Musée du Louvre* in Paris (Kaufman, 2008). At the *Vorderasiatisches Museum* (VAM) in Berlin, Goren and Mommsen also studied 65 documents from the Hittite archives at Hattuša (Boğazköy), the capital of the Hittite Empire in the late Bronze Age, consisting of official correspondence and contracts, legal codes, procedures for cult ceremony, oracular prophecies and literature of the ANE (this article, see Table 1 for details).

All of these studies employed OM and in most cases also INAA techniques. Jointly, they have demonstrated three methodological rules. The first was that sometimes cuneiform tablets were not produced of the same clay types as pottery of the same locality (Goren et al., 2004, pp. 316–318); hence, tablets should be treated separately from other archaeological ceramics. This means that the comparison between the clay of tablets and pottery fabrics should be made with caution. Second, clay selection for the production of cuneiform tablets by a given authority was not always consistent, and sometimes different clay sources were employed along a sequence of time. Such is, for example, the case of the letters sent from the Kingdom of Amurru to the Pharaohs of Egypt (Goren et al., 2003b). Third, while pottery can usually be studied using the routine mineralogical and chemical methods that involve destructive sampling, clay tablets are unique and delicate. Their sampling, if allowed at all, should be extremely minimal, thus often below the routine standards of the regular examination procedures. Therefore, going forward, it became clear that new methods should be introduced specifically for provenance studies of ancient clay documents, with the endeavor of enabling *in situ* application of scientifically based, non-destructive testing (NDT). Most significantly, this method must involve portable analytical apparatus which may be even carried as handbag in commercial flights, which would allow for their study in museums, departments of antiquities and collections, without the need to extract any samples and export them abroad for further analysis. Such method can offer new opportunities for the routine study of ancient clay documents, without violating any museological proviso or local antiquities law.

2. The portable X-ray fluorescence

In this context, it is only natural to consider the impressive development of portable X-Ray Fluorescence (pXRF) analyzers (Fig. 2A). The greatest advantages of pXRF for archaeology are twofold: first, it can be seen in many cases as a non-destructive method that under certain conditions does not require any extraction of samples. Moreover, the past few years have seen a meteoric and practical development of pXRF units, increasing the speed and efficiency of the testing process and making it available outside the research laboratory. A major development of the last decade was made in terms of the sensitivity of these units, as the limits of detection (LOD) of the previous generations were rather restricted, making them almost impractical for quantitative analysis of composite materials such as ceramics. Today, however, most manufacturers equip advanced models of pXRFs with Silicon Drift Detectors (SDD), lowering the LOD by an entire order of magnitude relative to the previous Silicon Pin Detectors and by up to four times relative to the HgI technology that existed over a decade ago (Goren, 2000). Given these developments, the use of an SDD-pXRF should now be tested again for *in situ* quantitative elemental NDT of delicate archaeological objects for provenance determinations (Padilla et al., 2006; Liangquan, 2008). Today, the pXRF procedure has become standard practice in the mining and natural resources industry, after tests have indicated that the pXRF instrument can give excellent correlation with laboratory-based reference methods such as atomic absorption spectrometry (Radu and Diamond, 2009).

A cautionary note is necessary here. While the industry and material sciences are witnessing an ever-growing interest in the development and application of NDT techniques, such methods often present problems when they are applied on non-

Table 1
List of tablets included in this study.

No.	VAT[a]	EA	CTH	Description	Reference to	OM definition/Ref[b]	INAA class[c]	pXRF group[d]
1	153	38		Letter of the King of Alašiya to Egypt.	Alašiya	Pachna marl from Cyprus (IiC: 51).	Alašiya	AlaA
2	1654	33		Letter of the King of Alašiya to Egypt.	Alašiya	Pachna marl from Cyprus (IiC: 50).	Alašiya	AlaA
3	6184		216	Fragments of an Akkadian docket mentioning Alašiya.	Alašiya	Pachna marl from Cyprus, as the EA Alašiya tablets (except EA 37).	Alašiya	AlaA
4	342	32		Letter of Tarhundurandu to Egypt.	Arzawa	Aegean "red clay" (IiC: 45).	East Aegean	Singular
5	148 + 2706	2		Letter of Kadašman-Enlil to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 34).	Unstudied	BabA
6	149	6		Letter of Burra-Buriyaš to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 35).	Unstudied	BabA
7	151 + 1878	11		Letter of Burra-Buriyaš to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 35-6).	Unstudied	Not BabA
8	152	8		Letter of Burra-Buriyaš to Egypt.	Babylonia	Euphrates sediment, coarser (IiC: 35).	Unstudied	BabA
9	1605	12		Letter of a princess to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 36).	Unstudied	BabA
10	1657	4		Letter of Kadašman-Enlil(?) to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 34-5).	Unstudied	BabA
11	1717	13		Inventory of gifts sent to Egypt.	Babylonia	Fine Euphrates sediment (IiC: 36-7).	Unstudied	BabA
12	6692		181	Letter to the king of Ahhiyawa (Tawagalawa).	East Aegean	Clayey, extremely micaceous illitic, chistose, with quartz & flysch, similar to Samos – Miletus amphorae.	East Aegean, Ephesus region	Singular
13	1583	340		Amarna scholarly text.	Egypt Es	Esna marl from Egypt (IiC: 76).	Unstudied	Singular
14	1611 + 1613 1614 + 2710	357		Myth of Nergal and Ereshkigal.	Egypt Es	Esna marl from Egypt (IiC: 83).	Unstudied	EgypA
15	1651 + 2711	14		Inventory of gifts sent from Egypt.	Egypt Es	Esna marl from Egypt (IiC: 25).	Unstudied	Singular
16	347	162		Letter of the King of Egypt to Amurru.	Egypt Es	Esna marl from Egypt (IiC: 25-6).	Unstudied	EgypA
17	1885	163		Letter of the King of Egypt to Canaan.	Egypt NS	Egyptian Nile silt (IiC: 26-7).	Unstudied	EgypD
18	1887	339		Letter written in Egypt.	Egypt NS	Egyptian Nile silt (IiC: 29).	Unstudied	EgypD
19	13067		169	Letter of Sutahapsap, son of Ramses II, to Hattušili III.	Egypt Ra	Egyptian marly clay.	Egyptian marl	EgypB
20	6156		156	Letter of Ramses II to Hattušili III.	Egypt Ra	Egyptian marly clay.	Egyptian marl	EgypC
21	6161		159	Letter of Ramses II to Hattušili III and Puduhepa.	Egypt Ra	Egyptian marly clay.	Unstudied	EgypC
22	6168		166	Letter of Ramses II to the king of Mira.	Egypt Ra	Egyptian marly clay.	Egyptian marl	EgypC
23	6169 + 7669		156	Letter of Ramses II to Hattušili III on the subject of the Syrian war.	Egypt Ra	Egyptian marly clay.	Egyptian marl Berk: Ela1	EgypB
24	6172		156	Letter of Ramses II to Hattušili III on the subject of the Syrian war.	Egypt Ra	Egyptian marly clay or Paleocene marl mixed with calcareous & quartz sand.	Egyptian marl	EgypC
25	7677		164	Letter of Ramses II to Puduhepa.	Egypt Ra	Egyptian marly clay.	Unstudied	EgypB
26	12887		68	Treaty with Kupanta-KAL of Mira and Kuwalia.	Hattuša	Hattuša fabric, nearly isotropic matrix, coarse quartzite inclusions.	Hattuša	HattB
27	12890		341	Gilgamesh fragment: Akkadian.	Hattuša	Hattuša fabric lightly fired. Inclusions: quartzite, quartz, some limestone.	Hattuša	HattB (Mn+, K+)
28	13007		8	Anecdotes (Palace Chronicle) of the Reign of Hattušili I.	Hattuša	Hattuša fabric.	Unstudied	HattA
29	13009		311	Naram-Sin in Anatolia.	Hattuša	Hattuša fabric.	Singular	HattB
30	13012		125	Šuppiluliuma II, Carchamish treaty(?).	Hattuša	Hattuša fabric, nearly vitrified by firing.	Hattuša	HattA
31	13059		284	Kikkuli.	Hattuša	Hattuša fabric.	Hattuša	HattA
32	13060		284	Kikkuli.	Hattuša	Hattuša fabric, nearly isotropic.	Hattuša	HattA
33	13064		6	Political testament of Hattušili I.	Hattuša	Hattuša fabric, nearly isotropic.	Hattuša	HattA
34	1655	42		Letter of a king of Hatti to Egypt.	Hattuša	Hattuša fabric (IiC: 31).	Unstudied	HattB
35	1656	44		Letter of Zita to Egypt.	Hattuša	Hattuša fabric (IiC: 31-2).	Unstudied	HattB (Fe+)
36	6163		53	Treaty with Tette of Nuhasse: Akkadian.	Hattuša	Highly fired, most likely Hattuša fabric, unlike EA 51 (assigned to Nuhasse in IiC: 91-2).	Unstudied	HattA
37	6165		61	Ten Year Annals of Mursili II.	Hattuša	Hattuša fabric, coarse version with quartzite and greywacke.	Unstudied	HattA
38	6207 + 13572		91	Silver Treaty with Ramses II (Akkadian).	Hattuša	Hattuša fabric, low firing.	Hattuša	HattA
39	6699		14	Fragments relative to the Syrian Wars: Mentioning Yarim-Lim, Atradu, Hammurabi, and Hattušili I.	Hattuša	Hattuša fabric, fine.	Unstudied	HattC
40	7423		52	Treaty of Šuppiluliuma I with Sattiwaza of Mitanni, Akkadian version.	Hattuša	Hattuša fabric.	Hattuša	HattA
41	7428		63	Treaty with Duppi-Teshub of Amurru.	Hattuša	Hattuša fabric. Low firing, matrix highly optically active.	Unstudied	HattB
42	7456		381	Muwatalli's Prayer to all Gods through the Storm-God of Lightning.	Hattuša	Hattuša fabric, isotropic matrix, inclusions: quartzite, quartz, decomposed carbonates.	Hattuša	HattA
43	7476		154	Letter of Šuppiluliuma I to a Pharaoh.	Hattuša	Hattuša fabric (fine) with much vegetal material (chopped grass), Low firing, matrix highly optically active.	Hattuša (Ta+, Sc+)	HattB
44	7487		124	Šuppiluliuma II, Carchamish treaty(?).	Hattuša	Hattuša fabric, fine. Low firing, matrix highly optically active.	Hattuša	HattA

Table 1 (*continued*).

No.	VAT[a]	EA	CTH	Description	Reference to	OM definition/Ref[b]	INAA class[c]	pXRF group[d]
45	7699 + 7701		402	Ritual of Alli.	Hattuša	Perhaps Hattuša fabric, but sample completely vitrified.	Hattuša	HattA
46	6180		833	Karum Hattuš.	Karum Hattuš	Very dark red-tan fine fabric, no inclusions, small sample.	Unstudied	KaHat
47	7674		833	Karum Hattuš Assyrian docket.	Karum Hattuš	Very dark red-tan, ferruginous, inclusions: quartz, augite, K-feldspar, chert.	Unstudied	KaHat
48	190	21		Letter of Tušratta to Egypt.	Mitanni	Mitanni clayey fabric (IiC: 41).	Unstudied	Not MitA
49	191	20		Letter of Tušratta to Egypt.	Mitanni	Mitanni marly fabric (IiC: 40).	Unstudied	Not MitA
50	2197 + 233	27		Letter of Tušratta to Egypt.	Mitanni	Mitanni clayey fabric (IiC: 42).	Unstudied	MitA
51	271 + 1600, 1618–20	29		Letter of Tušratta to Egypt.	Mitanni	Mitanni marly fabric (IiC: 43).	Unstudied	MitA
52	340 + 2191a–c	25		Inventory of gifts sent to Egypt.	Mitanni	Mitanni marly fabric (IiC: 42).	Unstudied	MitA
53	395	22		Inventory of gifts sent to Egypt.	Mitanni	Mitanni marly fabric (IiC: 41).	Unstudied	MitA
54	422	24		Letter of Tušratta to Egypt.	Mitanni	Mitanni marly fabric (IiC: 41).	Unstudied	MitA
55	1690	48		Letter of the Queen of Ugarit to Egypt.	Ugarit	Ugarit fabric (IiC: 90).	Unstudied	Ugar
56	1692	45		Letter of Ammishtamru to Egypt.	Ugarit	Ugarit fabric (IiC: 88).	Unstudied	Ugar
57	1693	47		Letter of the King of Ugarit to Egypt.	Ugarit	Ugarit fabric (IiC: 90).	Unstudied	Ugar
58	1694	46		Letter to Egypt.	Ugarit	Ugarit fabric (IiC: 89-90).	Unstudied	Ugar
59	7416b		309	Vocabulary	Vocabulary	Unstudied	Unstudied	HattC
60	7434a		299	Vocabulary	Vocabulary	Unstudied	Unstudied	HattB
61	7434b		304	Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
62	7434d		302	Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
63	7434f			Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
64	7437b		301	Vocabulary	Vocabulary	Unstudied	Unstudied	Singular
65	7440		304	Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
66	7441		304	Vocabulary	Vocabulary	Unstudied	Unstudied	Singular
67	7442		303	Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
68	7445			Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
69	7449		301	Vocabulary	Vocabulary	Unstudied	Unstudied	HattA
70	7450		301	Vocabulary	Vocabulary	Unstudied	Unstudied	Singular
71	13008		50	Treaty of Supiluliuma I with Sarri – Kusuh of Carchemish.	Singular	Marl with sand of basalt, dolerite, limestone, quartz, serpentinized minerals.	Unstudied	Singular
72	13049		123	Treaty of Tudhaliya IV with an unknown party (Isuwa?).	Singular	Extremely micaceous (illitic) clay with perfect optical orientation, sparse quartz, serpentine, phyllite, olivine.	Singular	Singular
73	1877	172		Letter fragment.	Singular	Marl with schistose minerals (IiC: 75).	Unstudied	Singular
74	348	356		Myth of Adapa and the South Wind.	Singular	Euphrates sediment(?), (IiC: 82-3).	Unstudied	Singular
75	6210		147	Madduwattas indicement.	Singular	Hattuša fabric(?) with inclusions of phyllite, plagioclase, quartz & granite.	Singular	Singular
76	6697		585	Vow of Puduhepa.	Singular	Dark red-tan, fine (like Karum Hattuš)?.	Unstudied	HattA
77	7412		105	Treaty of Tudhaliya IV with Sausgamuwa of Amurru.	Singular	Fine micaceous, undetermined.	Singular or Cyprus I (but Sc -)	Singular
78	7420		57	Recognition of Piyassili of Carchemish by Arnuwanda II.	Singular	Marl with river sand of basalt, dolerite, limestone, quartz, serpentinized minerals.	Unstudied	Singular
79	7454		191	Manapa-Tarhunta letter.	Singular	Ferruginous clay with abundant mica, plagioclase, chert and quartz.	Singular	Singular
80	7479		1	Proclamation of Anitta, King of Kussara.	Singular	Probably Hattuša fabric but containing coarser sand with ophiolitic components (serpentine, pillow basalt, schist).	Singular	Singular
81	7679		7	Akkadian version of the siege of Uršu.	Singular	Marl with river sand of basalt, augite, limestone, quartz, some serpentine.	Singular	Singular

[a] VAT: Vorderasiatisches Museum number. EA: Amarna number (see Moran, 1992 for details). CTH: The catalogued texts from Hattuša.

[b] IiC: Inscribed in Clay (Goren et al., 2004).

[c] Unstudied: Not examined by INAA. Singular: chemical single.

[d] See Table 3 for the average concentration patterns of these groups.

homogeneous materials such as ceramics or soils. Because NDT does not alter the article being inspected, it is regarded in material sciences as a highly-valuable tool in research, troubleshooting and product evaluation. For exactly the same reasons, one could expect this approach to be especially appreciated in the science of art and archaeology. Yet the pXRF has some important limitations that in certain cases undo the advantages of its non-destructiveness. Especially for a quantitative analysis certain assumptions have to be made which must be fulfilled, but which in practice cannot be always tested. The most important assumption is that the sample is homogeneous and has no layering. In such cases the absorption of the X-rays inside the sample to be analyzed can be considered and correct quantitative results are obtained. Good results can be also obtained, if the internal structure of a layered sample is known, which is rarely applicable. The absorption of X-rays depends on their energy and is strongest for the light elements that have the lowest characteristic X-rays energies. To give an example, a small silicon grain of 20 μm diameter absorbs already 75% of the intensity of the Ti–K X-ray radiation. Therefore to obtain quantitative X-ray measurements of the light elements in ceramics, often a sample is

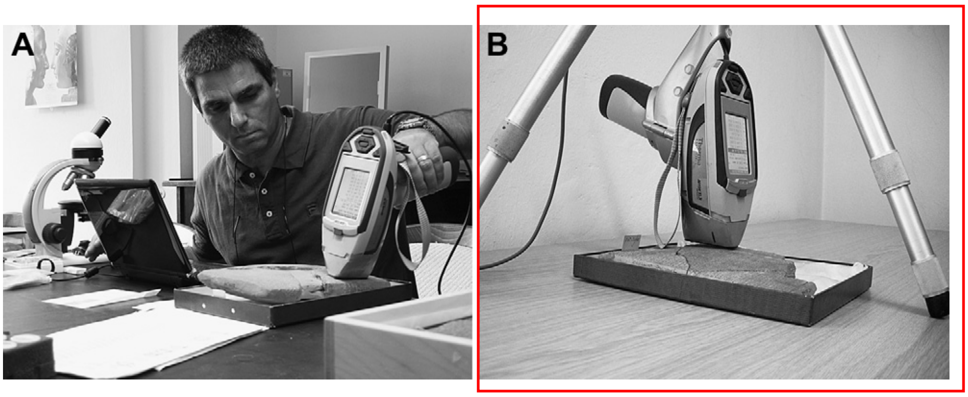

Fig. 2. Analysis of tablets in the VAM with the pXRF: A. Direct analysis of a tablet from Hattuša by holding the pXRF. B. The pXRF installed on an improvised accessory built from a photographic tripod and the unit holder of the optional Extend-a-Pole facility provided by Niton, showing the examination of a flat surface of a cuneiform tablet.

taken that is homogenized before measurement by melting it into a glass. With pXRF used in the NDT-mode without sample treatment, correct quantitative results are obtained only for homogeneous samples with no surface layer. To average over a possible varying internal structure it is advantageous to analyze a large sample area and perform repeated measurements at different positions to test for non-homogeneities. Since the directions of the exciting radiation from the X-ray tube entering the sample surface and the excited measured characteristic X-rays emitted into the detector are both approximately perpendicular to the sample surface (Fig. 2), a non-flat, structured surface can be tolerated, since for this measurement geometry the measured intensities do not depend on the surface structure. These known problems of pXRF were treated by the method to be discussed below.

The present article reports a study that aimed to test the application potential of the last generation of SDD-pXRF units for the routine study of clay cuneiform tablets. In view of the results, the possibilities and limitations of this method are discussed together with some preliminary outcome resulting from the analysis of a test group of documents of unknown provenance.

3. Material and methods

A group of 58 cuneiform tablets including 29 tablets from Hattuša and 29 letters and scholarly texts from the Amarna archive, was selected to form the reference group for this study. All tablets have been previously examined by the ‘classical’ intrusive methods of OM and, in most cases, also by INAA. The list of tablets is presented in Table 1 (nos. 1–58), together with their bottom-line OM and INAA results, indicating the logic for including each tablet as “reference” to a certain location. The references were made to Alašiya (Table 1: nos. 1–3), Arzawa (4), Babylonia (5–11), the east Aegean (12, the “Tawagalawa letter” discussed below), Egyptian texts written on Esna marl (13–16), Nile silt (17–18) and Marl D or marl-silt mixtures (19–25), Hattuša (26–45), Karum Hattuš (46–47), Mitanni (48–54) and Ugarit (55–58). In addition, 11 tablets from Hattuša, which were defined as “singular” according to the OM and/or INAA results, were added (Table 1: 71–81) in order to examine their relations with the reference groups. An additional group of 12 vocabulary tablets from Hattuša (Table 1: 59–70), which were not examined before by any natural scientific method, was added to form a test group attempting to assign them to the elemental clusters of the reference group. Hence, the pXRF examination covered a total sum of 81 tablets.

The SDD-pXRF apparatus used for this study was a Thermo Scientific Niton XLt-900 GOLDD equipped with a 50 kV X-ray tube with a Geometrically Optimized Large Area Drift Detector (GOLDD), 80 MHz real-time digital signal processing, and dual embedded processors for computation and data storage. As the Niton pXRF is set to use several company-preset matrices, we employed the “mining” matrix, which includes most of the relevant elements for ceramic studies (listed below). The apparatus uses up to four filters for each irradiation session, set to include the main, low, high, and light ranges of elements. The filters are set to include the following elements: Main: Sb, Sn, Cd, Pd, Ag, Mo, Nb, Zr, Sr, Rb, Bi, As, Se, Au, Pb, Hg, W, Zn, Cu, Re, Ta, Hf, Ni, Co, Fe, Mn, Cr, V, Ti. Low: Cr, V, Ti, Ca, K. High: Ba, Sb, Sn, Cd, Pd, Ag. Light: Al, P, Si, Cl, S, Mg. The irradiation time of each filter can be controlled by the software, as can be the display units (weight percent or ppm = mg/kg), and the calibration against standards (although Niton provides its pXRF with an internal, factory-set calibration program). The irradiation area is circular, 8 mm in diameter, making it efficient for relatively non-homogeneous surfaces such as ceramic earthenware. The Niton XLt-900 GOLDD is capable of detecting up to 32 elements (through the mining matrix), using the four different filters for the detection of the entire range of elements, from Mg ($Z = 12$) up to U ($Z = 92$).

With the above-mentioned limitations of the pXRF method in mind, a pilot testing was carried out on a group of pre-examined tablets from Hattuša. The analytical procedure for the main study that followed was set after some trial and error to the following: applying the mining matrix, the apparatus was set to the irradiation times of 60 s for each of the main and low filters and to 30 s for each of the high and light filters, with the measurement units set to ppm. Hence each measurement lasted for 180 s. The analyses were made on flat and smooth surfaces; visually clean of incrustation or dirt. The measured area was controlled and selected precisely by inspection through the internally installed video camera that this model of Niton pXRF is equipped with. In order to ease the use of the apparatus during the long sets of measurements, an improvised accessory was built in advance from a photographic tripod and the pXRF unit holder of the optional Extend-a-Pole facility provided by Niton (Fig. 2B). By manipulating the length of the tripod’s poles, the pXRF could be tilted and lowered or lifted in order to meet flatly with the measured surfaces.

For the reasons explained above, each tablet was tested in three different locations, resulting in approximately 10 min for the analysis of each tablet including data recording in the pXRF software and the selection of the appropriate surfaces for irradiation. As the scanning area is a circle of 8 mm in diameter, the total scanning area of the tree measurements together was about 150 mm^2. This is, in fact, nearly the standard size of a common thin section for ceramic OM. The results were also monitored as spectra using the Niton NDTr 6.5.2 software, to ensure accurate interpretation of the raw data.

The quantitative handling of the data included several stages. First, the three measurements and their given experimental uncertainties taken from each tablet were compiled on an Excel spreadsheet. To obtain the elemental composition of the tablet, the three values of each element (resulting from the three measurements at different locations) were averaged and the data subjected to a best relative fit for each case with respect to the average values using the standard Bonn statistical procedure (Beier and Mommsen, 1994a,b, Mommsen and Sjöberg, 2007). This was done to consider a possible varying inhomogeneity of the clays at the different locations by Si and/or Ca and/or other elements not measured like H and O (water). The concentration values and the given statistical measurement uncertainties of fifteen elements dependent on the intensities of the X-ray lines and of the underlying background, have been used for the calculation of the best relative fit factors (numbers in brackets indicate the averaged statistical measurement uncertainties of the three measurements in %): Al (8.1), K (1.6), Ti (1.3), V (8.4), Cr (9.5), Mn (4.9), Fe (0.6), Ni (17), Cu (4.8), Zn (1.8), Rb (1.5), Sr (14), Zr (1.9), Nb (8.3), excluding Si (1.0) and Ca (0.6). After the application of these factors, which only deviated from 1.00 by more than 5% in rare cases, the new average values (see Table 2) and standard deviations (= spreads = root mean square deviations *including* the experimental uncertainties) of each tablet were calculated and stored in a databank. Then these spreads of the three measurements were checked. Large spreads indicated either large statistical measurement uncertainties or large differences of the measured values at the three locations. Elements with spreads smaller than 20% included: Al (13.2%), Si (8.3%), K (9.0%), Ca (20%), Ti (6.3%), V (11.3%), Cr (12.9%), Fe (4.1%), Ni (18.5%), Rb (4.4%), Sr (9.6%), Zr (4.7%), Nb (8.3%), and Ba (12%). For these elements, strongly differing values for the three different locations of the measurement hardly ever occur.

At this stage, the elements having values below or near the LOD level of 2-sigma were cleared from the list. In addition, elements that are known to be affected by post-depositional processes or firing effects were also omitted. These include S, much affected by the presence of gypsum in arid soils (such as in el Amarna); Cl, which is increased by surface enrichment by salt; P, influenced by bones and ash in archaeological deposits; and Ba, which is enriched as barite in seismites within clay of lake deposits (Katz et al., 2009). In addition, due to the use of Cu and Zn filters by the mining matrix of the pXRF, these elements are subjected to inconsistent fluctuations in the measurement and they were also excluded from some of the statistical tests. Ca is also likely to be affected (either enriched or diluted) near the surface of archaeological ceramics due to its precipitation in groundwater and was also excluded from the successive statistical grouping using the Bonn statistical procedure for considering uncertainties and effects of dilution (Beier and Mommsen, 1994a,b), yet it was included in the PCA plots (below), as in practice, it seemed to have little effect on the datasets. The Bonn statistical grouping of the different tablets was done using the 12 elements: Al, K, Ti, V, Cr, Mn, Fe, Ni, Rb, Sr, Nb, and Zr. Although the spread of the values for Mn are larger than 20%, Mn could be included in the Bonn grouping procedure, since these errors were taken into account during the calculations. Only the pXRF data values have been used during the group forming procedure regardless of the OM and INAA definitions, in order to prevent any bias of the results. The assignment of each tablet to its pXRF group is given in Table 1, last column. The average concentration values of the groups and their spreads in % (=relative standard deviation, rms deviation, variance, coefficient of variation) are shown in Table 3 after application of the best relative fit factor (BRF) with respect to the average grouping values shown in the last column of Table 2.

Another statistical processing was made by manipulating the data with several multivariate statistical procedures to scrutinize its validity and clustering. The spreadsheet with the averaged three measurements taken from each tablet was loaded on a statistical package (SAS-JMP release 8.0.2). The mean concentrations of the 14 selected elements (Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Rb, Sr, Nb, and Zr) were tested by cluster analysis (Ward's method), discriminant, and factor and principal component analysis (PCA) to establish the grouping of the tablets according to their elemental composition of these elements for comparison. Of these, PCA proved to be the most useful method. By plotting the factor loadings, the most significant elements could be selected. This was done in an attempt to condense the resulting clusters according to the previous OM and INAA results. Better clustering was achieved when the following seven most significant elements were used: Al, Si, K, Ti, Rb, Zr, and Nb.

A very simple way for monitoring the results involved producing a scatterplot matrix for the entire dataset and calculating the correlation matrix between each pair of the elements involved. Elements that were not internally correlated were plotted by simple X–Y or ternary diagrams. This method proved useful for quick observation of the general clusters prior to the more detailed statistics. The best results were achieved by plotting K, Ti, Rb, and Nb. Especially significant were the X–Y plots of K and Ti (for which we coined the term "K–Ti test"), which enabled quick monitoring of the results during the pXRF analysis and the tentative attribution of an unknown tablet into a possible provenance cluster of the reference group prior to the more advanced, statistical development of the data.

4. Results

The groups obtained and assigned to the tablets are given in Table 1, column "pXRF group". The individual best relative fit factors with respect to the average grouping values are shown in Table 2, column BRF. Table 3 records the pXRF elemental concentration patterns of these groups including the values of Ca and Si not used for the grouping calculations. The number of six elements measured by both the pXRF and INAA elemental analytical methods is considered to be too low for a reliable comparison of the average absolute concentrations of the groups. The full INAA data and the grouping will be published in a future report; the INAA pattern of the group for Hattuša has been published already in Goren et al. (2007). Absolute values for trace and minor elements are difficult to measure and tend to disagree sometimes in different laboratories without a laborious inter-laboratory study due to the choice of different standards and calibration procedures. Using a fixed analytical method, groups of samples having similar composition can be reliably formed, even if not absolute, but only relative concentration values are measured.

As expected, the different groups formed repeat the grouping results of the OM and INAA evaluations for most of the tablets. The pattern for Alašiya, Babylonia, Egypt, Hattuša, Mitanni, and Ugarit are all well separated. Especially useful are the elements Fe, K, Rb, Ti, and Zr, since the average concentrations of these elements all have small standard deviations only slightly larger or comparable to the measurement uncertainties of the pXRF method included in the calculations (see above). The good separability of the groups is shown in Fig. 3, where the result of discriminant analysis is depicted. The overlapping groups Alašiya and Babylonia are due to the projection into the plane, the values for the elements Rb and Ni are quite different as can be seen in Table 3a. In higher projections, both groups are separated. The OM subgroups of the different clays of Egypt (Esna marl, Nile silt and Egyptian marly clay) can be resolved also with pXRF (Table 3b). Two tablets made of 'Esna marl' have pattern EgypA. Pattern EgypD belongs to the two 'Nile silt' tablets. The group 'Egyptian marly clay' is subdivided by the pXRF data into two groups EgypB and EgypC. Group EgypC (Ca is high, see Table 3b) is diluted by 15% (best relative fit factor 1.15) with respect to EgypB and differs after correction mainly in Mn and K. Since this is not seen with OM or INAA (Mn is not measured by the Bonn laboratory, but K is), this subdivision might be needless and obtained only due to a given too small experimental uncertainty for

Table 2
Elemental composition (as determined by pXRF) of the analyzed tablets, averages of the measurements at three different positions (14 significant elements only, values are in ppm (mg/kg), for the average statistical measurement uncertainties and for the root mean square deviations of the three measurements see text).[a]

No.	Museum No.	Reference to	Al	Si	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Rb	Sr	Zr	Nb	BRF
1	VAT 153	Alašiya	50,012	208,763	16,612	121,189	2461	186	125	676	31,926	62	28	292	95	14	1.00
2	VAT 1654	Alašiya	30,571	137,784	14,900	133,004	2139	175	104	634	33,078	66	27	338	93	13	1.04
3	VAT 6184	Alašiya	42,077	185,981	14,333	154,817	2390	223	140	1531	33,820	54	26	365	105	16	0.96
4	VAT 342	Arzawa	35,941	90,256	14,622	191,009	1706	201	130	723	41,417	45	42	200	116	17	–
5	VAT 148 + 2706	Babylonia	48,318	190,526	13,467	93,632	2787	224	298	867	44,868	209	19	184	91	12	0.99
6	VAT 149	Babylonia	42,282	180,540	12,550	87,917	2785	223	281	1025	45,310	165	19	749	105	12	0.95
7	VAT 151 + 1878	Babylonia	39,133	176,010	12,992	115,829	2531	195	196	673	35,712	114	21	237	92	10	–
8	VAT 152	Babylonia	33,699	149,759	11,085	107,058	2468	182	231	923	39,453	158	17	536	87	9	1.10
9	VAT 1605	Babylonia	43,569	181,255	11,029	96,416	2827	216	276	968	45,775	218	16	273	96	11	0.99
10	VAT 1657	Babylonia	53,288	191,134	12,847	92,491	2897	232	300	884	45,718	202	20	188	93	11	0.97
11	VAT 1717	Babylonia	46,893	175,699	11,002	98,037	2570	196	250	1021	43,891	211	17	466	93	12	1.02
12	VAT 6692	East Aegean	70,598	205,649	26,953	27,637	3658	367	256	1241	61,045	119	44	152	140	17	–
13	VAT 1583	Egypt Es	35,937	138,094	8201	213,703	2200	134	84	440	23,550		8	164	215	12	–
14	VAT 1611 + others	Egypt Es	41,255	150,933	8690	155,484	3160	192	143	673	34,985		10	182	147	14	1.05
15	VAT 1651 + 2711	Egypt Es	34,295	136,213	6989	145,769	2804	179	101	443	31,769		10	155	186	14	–
16	VAT 347	Egypt Es	51,912	171,151	8067	136,631	3496	214	131	773	40,140		10	463	155	19	0.98
17	VAT 1885	Egypt NS	62,092	203,988	8805	18,089	6660	395	212	1108	73,997	73	18	138	201	23	1.0
18	VAT 1887	Egypt NS	62,382	243,264	12,691	20,571	6250	305	227	1202	59,520	70	20	96	340	28	0.99
19	VAT 13067	Egypt Ra	39,221	288,451	11,912	35,826	6973	360	182	1422	456,98	45	9	299	174	21	1.09
20	VAT 6156	Egypt Ra	50,436	218,267	11,390	90,540	6246	346	210	1060	59,012	64	13	280	225	25	0.91
21	VAT 6161	Egypt Ra	35,872	221,316	8831	86,121	4795	363	166	888	49,519	75	8	323	219	23	1.01
22	VAT 6168	Egypt Ra	36,146	180,451	9640	85,175	4266	291	151	949	54,153	82	9	262	190	22	1.06
23	VAT 6169 + 7669	Egypt Ra	56,116	279,687	13,871	33,522	7181	317	196	1497	59,706	92	10	283	212	23	0.98
24	VAT 6172	Egypt Ra	46,628	212,408	9969	68,120	4905	294	177	736	52,279	91	9	262	175	21	1.07
25	VAT 7677	Egypt Ra	53,561	295,497	16,851	35,165	6580	410	207	1613	58,244	84	11	335	194	24	0.94
26	VAT 12887	Hattuša	75,562	238,788	25,536	25,651	4272	324	252	989	49,064	104	45	199	166	19	0.96
27	VAT 12890	Hattuša	83,175	278,680	29,067	23,061	4399	335	329	3197	46,665	167	41	153	149	18	0.99
28	VAT 13007	Hattuša	68,275	254,782	27,578	11,444	4274	303	219	910	43,839	65	52	115	170	19	0.99
29	VAT 13009	Hattuša	69,386	268,229	27,392	11,960	4358	341	253	1106	46,117	113	45	144	156	18	1.00
30	VAT 13012	Hattuša	58,887	289,041	26,859	33,154	3716	254	206	756	38,972	99	49	192	164	19	1.06
31	VAT 13059	Hattuša	89,247	235,912	31,048	28,819	3872	319	184	980	39,068	60	59	206	176	20	0.99
32	VAT 13060	Hattuša	70,938	218,935	28,405	39,874	3849	297	187	1332	38,984	54	60	205	180	20	0.99
33	VAT 13064	Hattuša	72,287	230,951	26,541	23734	3994	298	208	1506	41,435	59	54	194	169	19	1.01
34	VAT 1655	Hattuša	72,902	205,990	24,946	73,139	3975	319	262	793	49,893	125	44	234	154	19	0.98
35	VAT 1656	Hattuša	67,624	212,375	21,896	52,665	3562	293	206	848	45,873	91	39	216	161	18	1.04
36	VAT 6163	Hattuša	75,712	223,807	25,119	32,392	3929	293	203	1325	41,063	58	55	208	179	20	1.00
37	VAT 6165	Hattuša	75,673	236,068	26,357	25,604	4144	293	205	3169	45,973	82	47	178	163	19	1.00
38	VAT 6207 + 13572	Hattuša	89,202	251,720	30,260	26,526	4235	293	214	1323	45,664	101	49	210	171	20	0.96
39	VAT 6699	Hattuša	85,708	240,010	29,793	10,307	4231	358	192	770	33,745	82	56	146	176	19	1.03
40	VAT 7423	Hattuša	59,926	219,868	23,677	19,298	3811	282	201	890	45,010	56	44	149	164	18	1.06
41	VAT 7428	Hattuša	83,409	274,955	26,051	14,553	4660	325	249	1407	46,500	105	41	118	162	18	0.98
42	VAT 7456	Hattuša	84,147	256,756	24,837	31905	4255	296	250	833	44,192	68	43	174	171	18	1.00
43	VAT 7476	Hattuša	64,598	255,960	23,038	11652	4269	299	245	718	47,636	118	35	119	150	17	–
44	VAT 7487	Hattuša	89,278	281,286	27,676	6600	4284	334	212	1066	44,046	86	50	128	160	18	0.99
45	VAT 7699 + 7701	Hattuša	70930	238,264	26,167	30,159	3872	320	246	825	43,320	76	52	221	166	19	1.00
46	VAT 6180	Karum Hattuš	55,641	219,947	17,380	53,322	4459	296	240	1348	65,852	107	19	161	128	14	0.99
47	VAT 7674	Karum Hattuš	58,921	240,701	17,404	43,287	3807	289	224	3473	55,351	86	24	135	142	14	1.00
48	VAT 190	Mitanni	33,788	63,559	10,904	184,430	1167	134	180	587	34,511	193	22	227	86	12	–
49	VAT 191	Mitanni	19,068	96,567	10,971	127,201	2612	333	279	716	42,342	241	19	351	92	11	–
50	VAT 2197 + 233	Mitanni	31,898	146,558	11,619	82,265	2174	180	341	761	46,012	302	19	397	76	10	0.99
51	VAT 271 + others	Mitanni	39,722	172,084	13,586	98,224	2112	179	372	705	45,733	312	18	484	68	10	0.98
52	VAT 340 + 2191a-c	Mitanni	37,464	162,463	12,071	91,539	2419	195	331	745	47,431	294	20	194	86	11	0.94
53	VAT 395	Mitanni	43,665	163,273	11,513	99,437	2394	200	331	717	45,043	285	18	252	71	9	1.00
54	VAT 422	Mitanni	23,977	98,892	8747	91,811	1979	174	263	773	41,108	268	18	293	67	9	1.08
55	VAT 1690	Ugarit	21,469	111,661	6680	187,347	3633	1139	653	1308	31,200	162	13	278	100	16	1.00
56	VAT 1692	Ugarit	31,764	158,379	10,287	158,969	2574	248	1125	989	36,920	184	15	268	113	17	0.90
57	VAT 1693	Ugarit	19,626	93,911	4818	218,843	2221	478	496	1024	28,244	153	13	294	95	14	1.02
58	VAT 1694	Ugarit	23,031	109,921	6321	210,597	1903	211	567	1013	30,623	158	14	285	98	16	1.01
59	VAT 7416b	Vocabulary	97,722	264,016	36,817	17,794	4368	342	194	729	36,044	53	67	221	186	21	0.97
60	VAT 7434a	Vocabulary	67,602	221,632	23,344	49,207	4405	304	302	1024	46,826	124	42	247	155	19	1.00
61	VAT 7434b	Vocabulary	63,740	217,059	25,918	55,901	3720	309	227	1312	40,998	125	50	203	155	18	1.04
62	VAT 7434d	Vocabulary	78,249	256,613	22,789	35,970	4433	333	270	1271	41,434	96	50	207	159	18	1.01
63	VAT 7434f	Vocabulary	80,695	218,419	25,786	33,122	4291	309	268	831	45,703	96	55	213	167	20	0.96
64	VAT 7437b	Vocabulary	63,949	265,261	20,961	39,921	4263	313	361	710	48,003	231	24	335	94	12	–
65	VAT 7440	Vocabulary	83,384	252,086	28382	32,305	4240	312	222	1288	44,106	82	51	203	176	20	0.96
66	VAT 7441	Vocabulary	68,144	263,501	23,528	59,313	3113	315	179	613	40,653	55	27	384	83	10	–
67	VAT 7442	Vocabulary	69,878	245,364	25,011	13,959	3831	306	214	658	43,157	69	46	125	154	17	1.06
68	VAT 7445	Vocabulary	88,435	265,405	30,138	5998	4333	357	214	719	40,037	72	51	121	161	18	0.99
69	VAT 7449	Vocabulary	65,117	207,294	24,673	62,501	3583	301	214	1267	42,340	80	54	224	172	20	0.98
70	VAT 7450	Vocabulary	45,514	288,682	9191	92,741	3355	231	291	1032	44,751	163	19	337	132	17	–
71	VAT 13008	Singular	31,275	27,7770	14,642	27,505	4553	231	406	948	56,319	314	14	270	100	11	–
72	VAT 13049	Singular	68,680	241,037	23,487	18,652	5958	396	333	1086	65,819	165	33	198	145	21	–
73	VAT 1877	Singular	37,356	134,994	9405	126,726	3274	198	127	415	36,767		12	454	132	19	–

Table 2 (*continued*).

No.	Museum No.	Reference to	Al	Si	K	Ca	Ti	V	Cr	Mn	Fe	Ni	Rb	Sr	Zr	Nb	BRF
74	VAT 348	Singular	28,578	119,200	9152	149,063	3544	352	134	685	39,015		12	198	163	18	–
75	VAT 6210	Singular	50,775	215,858	25,125	25,288	4058	364	163	333	37,847		57	145	211	22	–
76	VAT 6697	Singular	85,522	240,118	28,413	30,333	4202	313	219	972	40,818	69	56	204	177	21	0.96
77	VAT 7412	Singular	54,663	211,846	17,952	87,060	2833	280	307	1830	49,197	272	20	206	89	12	–
78	VAT 7420	Singular	31,775	150,276	9631	18,3181	2680	285	312	1043	38,805	152	14	366	87	12	–
79	VAT 7454	Singular	42,489	177,161	19,280	10,4222	2692	312	173	3026	34,579	127	33	338	145	12	–
80	VAT 7479	Singular	63,147	253,240	22,738	10,491	4394	283	280	803	59,134	149	33	76	153	18	–
81	VAT 7679	Singular	28,856	182,296	10,329	52,204	2866	451	714	804	45,026	339	10	231	59	8	–

[a] Averaged from the three measurements of each tablet. BRF = individual best relative fit factors with respect to the average grouping values given in Table 3.

Mn and K. Adding all 7 members to one group EgypB + C results in spread values for Mn and K of 24% and 16%, respectively. For the large group of reference tablets from Hattuša, the pXRF data are also statistically sub-dividable into 3 groups: HattA, HattB, and the pair HattC. The two tablets from Karum Hattuš form a seperate group. The main difference in composition between HattA and HattB is in the Rb and Zr values, whereas a lower Fe value than in both the other groups is seen in the tablets of the pair HattC (see Table 3c). The pattern Hatt-sum is obtained if all 30 reference items for Hattuša are merged into one group. The pair from Karum Hattuš differs from group HattA after application of a best relative fit factor of 1.22 in lower Rb and K and a higher Fe concentration.

Not all the reference tablets are members of their expected group; some tablets are statistically outliers either due to the choice of different clay or due to a contamination of the elements considered or due to a wrong concentration value caused by the limitations of the pXRF method. In the set of the 58 reference tablets, there are only five of such chemical loners: tablet VAT 151 + 1878 (EA 11, Tables 1 and 2: No. 7) does not match BabA (Babylonia); VAT 1583 (EA 340, Tables 1 and 2: No. 13) and VAT 1651 + 2711 (EA 14, No. 15) are both different from each other and from the pair of the two other Esna marl tablets; VAT 190 (EA 21, Tables 1 and 2: 48) and VAT 191 (EA 20, Tables 1 and 2: No. 49) are different and do not match MitA (Mitanni). Yet it was already noticed in the past based on INAA and OM, that two distinctive clay types were used by the Mitannian scribes: a marly type (including EA 20) and a clayey type (including EA 21) (Goren et al., 2004, pp. 38–44). These are probably the different INAA 'chemical profiles' referred to by Dobel et al. (1977). All other tablets match the former results. The Arzawa letter (VAT 342, No. 4) and also the Tawagalawa letter (VAT 6692, No. 12) are single items representing clays from northern Ionia/Aiolis (Artzy et al., 2004; Mommsen and Kerschner, 2006) and an Ephesos region INAA group (Kerschner and Mommsen, forthcoming), respectively, and their pXRF concentration patterns are also singles.

Table 3a
pXRF elemental concentration patterns measured for Alašiya (AlaA), Babylonia (BabA), Mitanni (MitA), and Ugarit (Ugar).[a]

	AlaA (3 samples, factor 1.00)		BabA (6 samples, factor 1.00)		MitA (5 samples, factor 1.00)		Ugar (4 samples, factor 1.00)	
	M	σ (%)	*M*	σ (%)	*M*	σ (%)	*M*	σ (%)
Al%	4.36	16	4.35	12	3.49	22	2.29	20
Ca%	12.3	7.8	9.23	8.6	9.00	8.4	19.0	19
Cr	124	19	273	9.3	325	9.5	670	31
Fe%	3.19	3.0	4.40	1.7	4.50	1.4	3.09	6.0
K%	1.53	9.0	1.19	6.9	1.14	12	0.63	23
Mn	671	9.3	950	8.2	721	8.0	1050	17
Nb	14.5	9.8	11.1	12	9.67	14	15.2	9.3
Ni	60.7	31	193	12	289	8.4	161	13
Rb	27.5	4.4	18.0	5.9	18.5	5.4	13.5	7.2
Si	18.6	16	17.7	4.7	14.7	17	11.2	8.9
Sr	330	11	395	58	311	39	290	2.8
Ti%	0.24	2.8	0.27	2.7	0.21	4.1	0.22	11
V	193	11	211	10	185	10	223	30
Zr	97.5	4.0	92.8	3.4	73.0	7.9	99.2	2.8

[a] Averages *M* in μg/g (ppm), if not indicated otherwise, and spreads σ in percent of *M*. The individual datasets have been corrected for dilution effects by the best relative fit factor with respect to *M* (see Table 2).

The results from the 12 vocabulary tablets are discussed below. Finally, the 11 tablets that have been singulars with OM and, if studied, with INAA, are also singulars according to the pXRF data with the exception of VAT 6697 (No. 76, Vow of Puduhepa), which belongs to group HattA and should have been made in Hattuša. This tablet was not studied with INAA, and OM places it in the neighborhood of Karum Hattuš but without certainty.

Fig. 4A presents the PCA score plot of the international letters from el Amarna and the Hattuša tablets of the reference group, using the 14 selected elements listed above. The clusters retrieved represent the overall true groupings of the tablets according to their known provenance. Especially significant is the clustering of the Hattuša group, the separated Egyptian cluster, and the Syro-Mesopotamian cluster where an internal clustering is visible between the Babylonian, Mitannian, and Ugaritic documents. The Alašiya letters form a separate cluster, but they also include the single tablet from Arzawa. Another problem of this kind occurs with the so-called "Tawagalawa letter" (VAT 6692, No. 12), which in the PCA score plot of the 14 elements falls within the Hattuša cluster. However, these problems are corrected when the factor loadings of the elements are used to select only the most significant elements. Fig. 4B demonstrates the PCA results using only the seven most significant elements (Si, Al, K, Ti, Rb, Zr, and Nb). In this case, the Tawagalawa letter is separated from the Hattuša cluster, the Egyptian cluster is divided into two distinct groups of tablets made of Esna marl (from Amarna, as identified by OM), and Nile sediments including the Ramses II letters from Hattuša that were made of Egyptian "Marl D" clay (as identified by OM) and two Egyptian dockets from Amarna made of Nile Silt. Better definition is also seen among the Mesopotamian and north Syrian clusters of the Babylonian, Mitannian, and Ugaritic letters. Hence the use of these elements on their own provides a more refined clustering. Still, while this grouping was significant enough to form the reference clusters for Hattuša, Karum Hattuš, Egypt, Mitanni, Babylonia, Alašiya and Ugarit, other possible provenances remained undefined. These will be established in the future by further analyses of pre-examined tablets from the Amarna and other archives.

The K–Ti test of the reference tablets (Fig. 5) yielded nearly similar results. Especially significant is the distinction of the Hattuša cluster, which is by far higher in K contents than any other group. At the same time, Egyptian tablets made of Nile alluvial sediments (including both Nile Silt and Marl D categories), are distinctive by their high Ti and low K contents. The Mesopotamian and north Syrian tablets demonstrate relatively low values of both elements. The reasons for this can be found in the mineralogy of the sediments. As Morgenstein and Redmount (2005, p. 1621) point

Table 3b
pXRF elemental concentration patterns measured for Egyptian clays (see text).

	EgypA (2 samples, factor 1.00)		EgypB (3 samples, factor 1.00)		EgypC (4 samples, factor 1.00)		EgypD (2 samples, factor 1.00)		EgypBC (7 samples, factor 1.00)	
	M	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)
Al%	4.62	14	4.87	13	4.16	15	6.19	8.1	4.47	14
Ca%	14.9	14	3.29	5.9	8.30	19	1.92	12	5.31	53
Cr	138	18	192	14	180	18	218	11	185	16
Fe%	3.82	4.8	5.51	8.1	5.24	4.4	6.64	16	5.39	5.5
K%	0.85	10	1.34	6.3	0.97	8.4	1.07	26	1.15	16
Mn	733	6.7	1519	6.0	924	7.6	1140	5.2	1136	24
Nb	16.8	13	22.7	6.7	22.8	7.4	25.5	12	22.6	7.1
Ni	–		69.5	31	73.7	26	71.4	27	72.0	28
Rb	10.2	10	10.2	11	9.74	16	18.6	6.6	9.92	11
Si	16.6	6.5	27.6	1.1	20.8	5.7	22.3	12	24.3	16
Sr	322	58	301	8.3	276	9.2	116	26	286	8.0
Ti%	0.34	4.9	0.70	10	0.51	11	0.64	4.7	0.59	16
V	207	12	357	13	324	9.3	330	18	336	9.9
Zr	152	4.2	187	3.1	197	7.5	269	36	193	9.3

out, Anatolian and Aegean “red clays” are derived from geological terrains that contain volcanic minerals loaded with potassium-rich minerals such as sanidine, alteration products of volcanic glass such as potassium and rubidium-adsorbed montmorillonite, and alteration products of potassium feldspar such as illite. Therefore, they are rich in K and have low Ti concentrations. On the other hand, Egyptian sediments are known to be enriched by Ti because of the abundance of detrital anatase and rutile in them (Takla and Arafa, 1975; Schneiderman, 1995). At the same time, Euphrates sediments are composed mostly of smectite and palygorskite (Ali, 1976; Berry et al., 1970; Philip, 1968), having relatively lower concentrations of both K and Ti.

5. Some case studies

One outcome of this study is the analysis of the twelve previously unexamined vocabulary tablets, and the nine pre-examined tablets from Hattuša and two from Amarna, which were defined as “singular” by INAA and/or OM. The pXRF analysis made it possible to assign these tablets to a provenance, or at least decipher whether a tablet is indeed local to Hattuša or not (Fig. 6). In theory, this method could be also used prior to sampling tablets for one of the “classical” archaeometric methods, in order to minimize the number of tablets selected for sampling. Yet in practice, many of the tablets that were found to be external to the Hattuša cluster could be attributed to a certain provenance by their proximity to other known clusters within the reference group. The following examples demonstrate such cases:

5.1. The Hittite correspondence with Egypt in the time of Suppiluliuma I

Within the four letters EA 41–44 there is one definitely written from Suppiluliuma I (EA 41, unstudied by pXRF), one is written by his brother Zida (EA 42, Tables 1 and 2: No. 34) and another one is probably another letter sent by Suppiluliuma I (EA 44, No. 35). The interesting point is that all these letters do not show the normal sign forms typical for texts in the Hittite archives of this period. OM analysis of EA 42 and EA 44 was aimed at supplying the available analytical data for similar analyses on other Hittite texts (Goren et al., 2004, pp. 31–32). CTH 154 (No. 43) is a Hittite draft of a letter sent to Egypt by Suppiluliuma I (van den Hout, 1994, without the additional fragment 154/s = KBo 49.13). A comparison of the clay of the letters found in Amarna with that of the Hattuša tablet confirmed the similarity between the letters from both sites (Table 1). INAA study of CTH 154 assigned it to the Hattuša group (though with somewhat higher values of Ta and Sc unmeasured by pXRF, see Table 1). Hence this case can serve as a model test for the efficiency of the pXRF clustering, because the three tablets, obviously sharing the same provenance, were found in two different sites (Amarna and Boğazköy), where they were exposed over millennia to different post-depositional processes resulting from

Table 3c
pXRF elemental concentration patterns measured for Hattuša (HattA, ~B, ~C) and Karum Hattuš (KaHat).

	HattA (20 samples, factor 1.00)		HattB (8 samples, factor 1.00)		HattC (2 samples, factor 1.00)		HattABC (30 samples, factor 1.00)		KaHat (2 samples, factor 1.00)	
	M	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)	*M*	*σ* (%)
Al%	7.73	10	7.39	8.8	8.98	7.2	7.71	9.7	5.68	8.2
Ca%	2.45	46	3.11	72	1.53	45	2.38	52	4.82	14
Cr	216	11	258	13	193	11	225	14	232	12
Fe%	4.24	5.8	4.75	3.2	3.49	2.2	4.40	7.7	6.05	12
K%	2.67	6.1	2.50	6.1	3.28	11	2.67	7.6	1.74	3.0
Mn	898	26	1242	64	709	8.6	870	20	2411	63
Nb	18.9	7.6	18.1	8.0	19.8	7.2	18.7	7.7	14.2	10
Ni	74.2	28	114	18	66.7	35	84.7	30	95.3	21
Rb	51.2	7.6	41.1	6.3	61.2	8.4	49.2	12	21.9	16
Si	24.5	9.0	25.2	10	24.9	5.2	24.7	8.4	23.0	7.0
Sr	184	19	178	29	187	24	182	22	147	12
Ti%	0.41	3.5	0.43	6.7	0.43	2.2	0.41	4.4	0.41	11
V	305	8.2	318	8.0	351	7.2	311	8.0	292	8.3
Zr	167	3.7	157	3.7	180	1.9	165	5.2	134	8.1

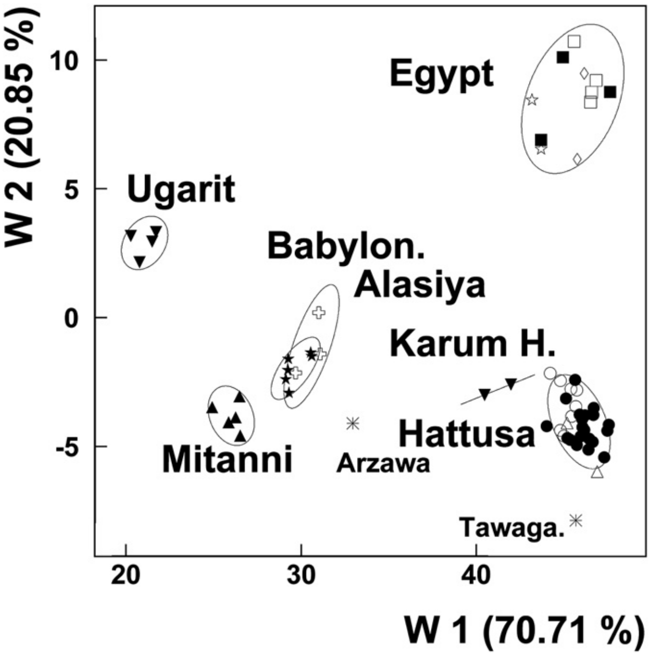

Fig. 3. Result of a discriminant analysis assuming 7 clusters of the grouped 61 tablets using all elements given in Table 2 except Ca and Si that might be part of possible diluents (and except Ni because of zero data). The 4 pXRF subgroups assigned petrographically to Egypt [EgypA (open star), ~B (full square), ~C (open square), and ~D (diamond) are added to one cluster 'Egypt'. Also the 2 subgroups of the tablets from Hattuša [HattA (full circle), HattB (open circle) and the pair HattC (open triangle, up) are assumed to form one group 'Hattuša'. The other clusters shown are described in the text. The Arzawa and the Tawagalawa tablets, both ungrouped, are drawn as single points (+ and × superposed). Plotted are the discriminant functions W1 and W2 describing 70.7% and 20.9%, respectively, of the between group variance. The ellipses are the 2 sigma boundaries of the groups. All concentration patterns are well separated in this diagram except the overlapping clusters of tablets from Alašiya (open plus) and Babylonia (full star), but they are resolved in higher projections.

dissimilar sediments and climatic conditions. Indeed, in all tests EA 42, EA 44 and CTH 154 fall neatly within the Hattuša cluster. Obviously the script of the letters EA 42 and 44 written in Akkadian are clearly different from the letter draft CTH 154, which is written in the Hittite language. An explanation for that could be that there were specialist scribes for the international correspondence working in Hattuša as well, but they were trained in a different scribal tradition then the normal Hittite scribes (Klinger, 2003, p. 239, n. 10, 11).

5.2. *"The Anitta proclamation" (CTH 1, Tables 1 and 2: no. 80)*

The text of the tablet KBo 3.22 (=CTH 1.A), one of the three fragmentary preserved copies, documents events leading up to the founding of the kingdom of the so-called Pitḫana dynasty, reporting the earliest genuinely historical events in Hittite language. Pitḫana's regime dates to the time of the old Assyrian merchant colonies in Anatolia, some three or two generations earlier then the time of the first written text to be found in the archives of the later Hittite capital Hattuša. The script of the tablet displays the typical old Hittite ductus, showing a number of grammar and writing features typical of the early stage of the Hittite scribal tradition. However, the text testifies that the original version of it was carved on a stele which was erected at the gate of the king's city of Kaneš/Neša, probably inscribed in Hittite or in Assyrian. The document records the deeds of Anitta's father Pitḫana, the beginning of Anitta's career, the rescue of ^{D}Siu-summin from the king of Zalpuwa, and Anitta's destruction of the city of Hattuša. While INAA classified this tablet as "singular", OM suggested that it may still be seen as a representative of the Hattuša fabric, yet coarser than the usual for this group in terms of the sand added to it as temper, thus including more diverse rock fragments and minerals. The pXRF data places it on the fringe of the Hattuša cluster, or somewhere near it but in the direction of the Karum Hattuš cluster (Fig. 6).

As far as we know the Hittite writing tradition starts at least in Hattuša with Hattušili I but the events unrolled have to be dated nearly a century earlier than the founder of the Hittite kingdom in Hattuša – is it possible to think of an older Hittite writing tradition starting in a different place may be in Kuššara, the hometown of Hattušili?

5.3. *The so-called "Tawagalawa letter" (CTH 181, Tables 1 and 2: no. 12)*

This document was thought to be written by a Hittite king, most likely Hattušili III, to a king of Ahhiyawa around 1250 BC. This letter, of which only the third tablet has survived, concerns the activities of a certain Piyamaradu against the Hittites, requesting his exile to Hatti with safe escort. The document refers to a certain Tawagalawa, a brother of the king of Ahhiyawa. However, the common name for this tablet may be a misnomer, as Singer (1983) has demonstrated that in fact, it was Piyamaradu who was in the focus of the document while Tawagalawa had a minor role in it. The Tawagalawa letter further mentions "Millawanda" better known under the name Miletus and its dependent city Atriya, as does the

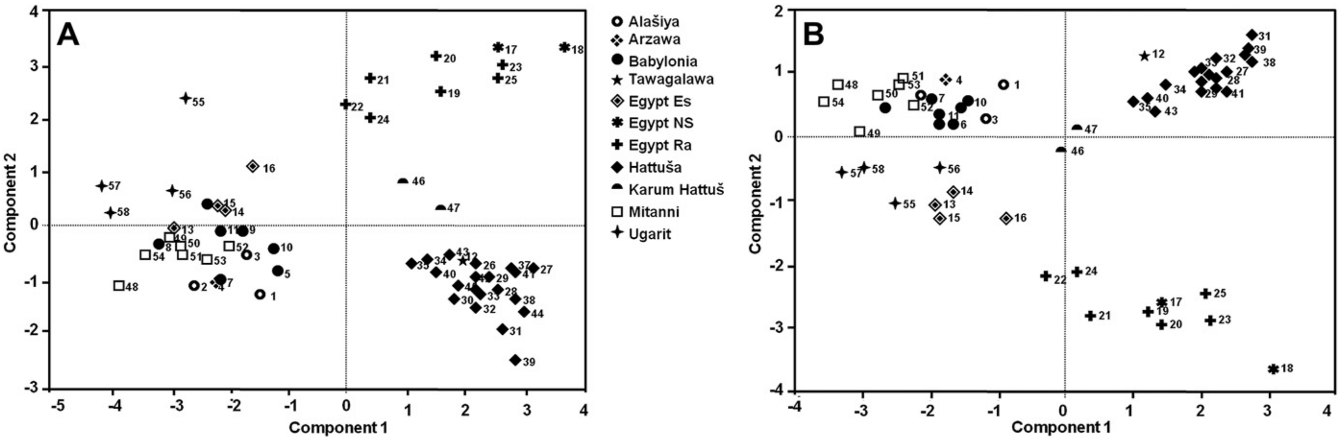

Fig. 4. Principal component analysis (PCA) of the pXRF results from the reference group of tablets (case numbers refer to the serial numbers in Table 1). A. Using 14 elements (Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Rb, Sr, Zr, and Nb). B. Using seven elements (Al, Si, K, Ti, Rb, Zr, and Nb).

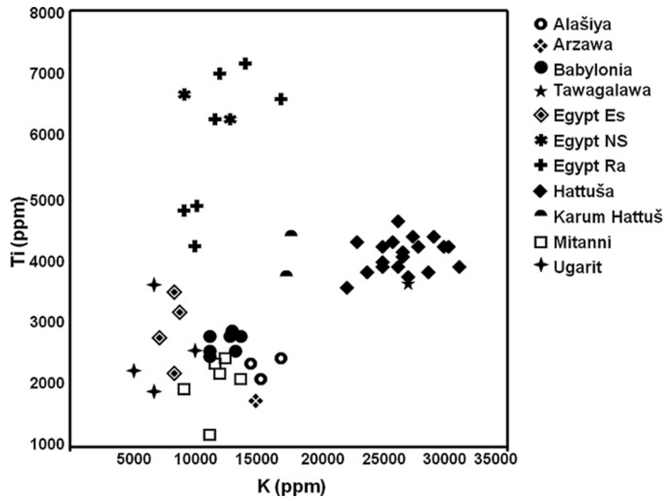

Fig. 5. K–Ti test of the reference tablets. The axes are plotted in ppm (mg/kg).

Milawata letter (CTH 182), and its governor Atpa, as does the Manapa-Tarhunta letter (CTH 191).

The results of both INAA and OM are very significant. In the first method, CTH 181 concurs with the reference material from the Eastern Aegean coastal area south of Ephesus. In terms of OM, if the Eastern Aegean is taken as the general source area, it is similar to the Samian amphora fabric presented by Whitbread (1995, pp. 122–133), which are known to be produced also at Miletus and the Samian Peraia. These results fit very well with the text itself because the Hittite word *kā* "here" (col. i, line 73) refers to the place, where the letter is written i.e. "Millawanda".

As discussed above, when using the pXRF data, CTH 181 can serve as good indication for the advantage of the "seven significant elements" over the "14 elements" testing.

5.4. *"The Siege of Uršu" (CTH 7, Tables 1 and 2: no. 81)*

"The Siege of Uršu" (CTH 7, Tables 1 and 2: No. 81) is a fragmentary tablet with a historical narrative concerning a military campaign of a Hittite king whose name isn't mentioned against the city of Uršu. The city of Uršu is already attested in the Ebla texts from the 3rd millennium and in the texts from Mari in the early 2nd millennium; a possible localization could be the region of Samsat, north of Karkemiš at the Upper Euphrates. The text is written in Akkadian language using a variant of the cuneiform script typical for the late Old Babylonian signs and ductus forms of Northern Syria, but the historical event could be dated to the time of Hattušili I on the basis of a campaign against the city of Uršu which is mentioned in his annals. Because of this coincidence and some literary features of the text (Hoffner, 1980; Beckman, 1995) with parallels in other old Hittite literature texts like the so-called "Palace Chronicle" the narration is considered of a Hittite origin. But there are no indications where the text was written and composed and who was its author. While INAA placed this tablet as "singular", OM suggests that by its petrographic affinities, it may be linked with the Upper Euphrates fabrics. The pXRF data clearly separates this text from the Hattuša cluster. Using the "seven significant elements" method, the tablet falls within the Mittanian cluster or on the fringe of the Babylonian group (Fig. 6). According to this result the tablet should be written in one of the late Old Babylonian centers in the region of Upper Euphrates, obviously not in a scriptorium of the Hittite capital Hattuša – this should be a very good explanation why the tablet is written in a non-Hittite ductus type using the Akkadian language.

5.5. *A group of vocabularies from Hattuša*

Twelve of the documents from the Hattuša archive which were examined by pXRF are classified as vocabularies. This assemblage was most likely part of the local school for scribes, where students were trained in producing tablets and inscribing them in cuneiform script. The vocabulary texts discovered at Hattuša are for the most part students' exercises; only few are stored in tablet collections. In order to achieve a better understanding of the work of the Hattuša school of scribes, we attempted to establish whether a given tablet is of foreign origin (Western Asiatic or other), hence most likely serving as a textbook, or was written in Hattuša, perhaps as an exercise.

Only one vocabulary tablet (VAT 7416c) has been previously studied by OM and was classified as belonging to the Hattuša fabric. In the present study, other twelve vocabularies were analyzed by pXRF (Tables 1 and 2: Nos. 59–70). When the Bonn statistical procedure is applied, seven of the unstudied vocabulary tablets are made of clay from Hattuša and are members of group HattA. One vocabulary (VAT 7434a, No. 60) belongs to group HattB. VAT 7416b (No. 59) forms a pair with tablet VAT 6699 (No. 39) with pattern called HattC. The remaining three tablets are chemical singles and

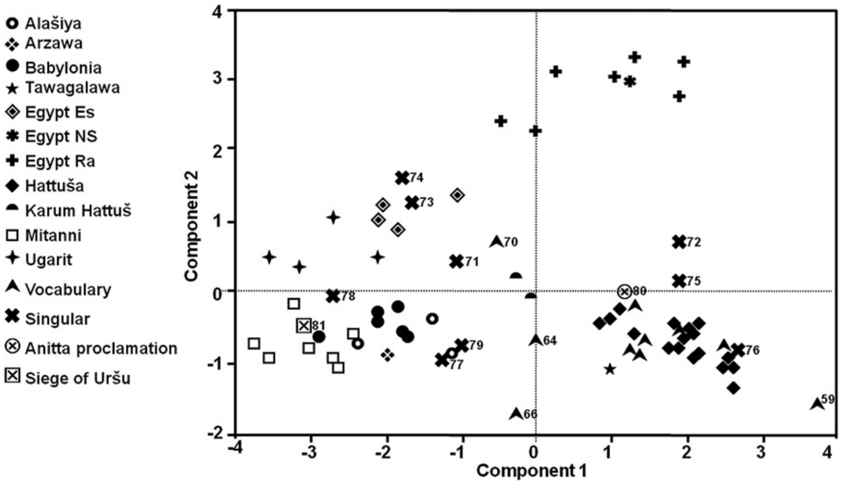

Fig. 6. Principal component analysis (PCA) of the reference group, compiled with tablets defined as "singular" by OM and INAA, and a group of vocabulary texts from Hattuša (case numbers refer to the serial numbers in Table 1), using the 7 significant elements (see Fig. 4B).

have each a different composition not otherwise represented in the dataset and are of unknown provenance.

When the results are plotted against the reference group as PCA graph using the "seven significant elements" (Fig. 6), it clearly indicates again that while the same seven tablets match the Hattuša cluster, four vocabularies are of different composition. None of the latter matches any cluster of the reference group. A very interesting result is that two of these fragments are KBo 1.40 (VAT 7441, No. 66) and KBo 1.55 (VAT 7416b, No. 59), which – regarding the content and the script – could be parts of one and the same tablet, representing a Boğazköy version of the acrographical list type Proto-Kagal with four columns: one for the sign form, one for the spelling of the sign, one for an Akkadian and one for a Hittite equivalent. While the pXRF results clearly indicate that these are in fact fragments of two different tablets, having different chemical compositions, it is very unlikely that such type of a lexical list was brought to Hattuša from other localities. There are not so many different sign forms preserved on the fragments but there is a tendency to use older sign forms, may be indicating that the tablets were written relatively early, earlier than the empire period, perhaps at the end of the fifteenth century BC. As such, they could represent another selection of raw materials that are different from the type used for the main body of the reference group. In fact, according to the Bonn statistical procedure VAT 7416b might be assigned to HattC. Still, this option seems highly unlikely in view of the homogeneousity of the other Hattuša reference tablets and the local vocabularies. Therefore, this hypothesis still requires further investigation.

The other two fragments are KBo 1.50 (VAT 7437b, No. 70) and KBo 1.44 (VAT 7450, No. 64), belonging to different tablets of the Boğazköy version of Erimḫuš, showing the typical script of the empire period and having both a column in Hittite language. They look like standard exercise texts and it is difficult to explain, why the pXRF results are indicating that they come from another locations. It may be assumed that these texts originate within the Hittite scribal tradition, but not from Hattuša. Obviously, this opens new possibilities for future research concerning the transfer of scholarly texts between different centers within the confines of the Hittite Empire. In terms of methodology, the pXRF results can minimize the study of an assemblage of tablets by intrusive analyses, limiting the sampling only to the tablets that do not match the local cluster of raw materials.

6. Conclusion

The primary conclusion of this study is that new generations of pXRF analyzers can yield proper grouping of clay tablets (and other ceramics) according to their provenance and serve as a non-destructive method for assigning more tablets into these cluster-groups. Although it should be emphasized that the method presented here cannot substitute INAA as a general elemental provenancing procedure for ceramics, or OM as a mineralogical provenancing tool also capable of exploring technological processes, it can become extremely powerful in cases where internal groupings of "closed" populations of delicate items are needed. Namely, measurements by pXRF of the element concentrations of tablets whose provenance has been already determined by OM and/or INAA, can create a database for further pXRF examination of unstudied tablets from the same archives in collections where intrusive sampling is not allowed. For example, by establishing the 'pXRF grouping' for the local Hattuša fabrics of the tablets that were studied by intrusive methods in the VAM, other tablets from Hattuša can be studied in Turkey (where sampling is often prohibited) and matched statistically with the clusters of tablets of known provenances in the database. Other 'pXRF groups' can be established by the same methodology for other significant ANE landmarks, such as Ugarit, Cyprus, Waššukanni (Mitanni's capital), Carchemish, Assyria, Babylonia, and many Canaanite cities, based on the pXRF analysis of tablets that were previously studied by OM and INAA. This enables the non-destructive study of the Amarna tablets in the Cairo Museum (where sampling was not allowed) or the Canaanite tablets in the Museum of the Ancient Orient in Istanbul. The same approach can be applied also in other cases, such as the study of bullae, figurines, intact pottery vessels and other items of high museological value. Under these conditions, pXRF can become an extremely powerful tool (and the only one available) for provenance determinations of such delicate clay-based artifacts.

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References

Ali, A.J., 1976. Heavy mineral provinces of the recent sediment at the Euphrates. Journal of the Geological Society of Iraq 10, 33–46.

Artzy, M., Perlman, I., Asaro, F., 1976. Alašiya of the Amarna letters. Journal of Near Eastern Studies 2, 171–182.

Artzy, M., Mommsen, H., Asaro, F., 2004. Arzawa: neutron activation analysis of EA32. In: Goren, Y., Finkelstein, I., Na'aman, N. (Eds.), Inscribed in Clay: Provenance Study of the Amarna Tablets and Other Near Eastern Texts. Monograph Series of the Institute of Archaeology, Tel Aviv University, Tel Aviv, pp. 45–47.

Beckman, G., 1995. The siege of Uršu text (CTH 7) – an old Hittite historiography. Journal of Cuneiform Studies 47, 23–34.

Beier, Th., Mommsen, H., 1994a. Modified Mahalanobis filters for grouping pottery by chemical composition. Archaeometry 36, 287–306.

Beier, Th., Mommsen, H., 1994b. A method for classifying multidimensional data with respect to uncertainties of measurement and its application to archaeometry. Naturwissenschaften 91, 546–548.

Berry, R.W., Brophy, G.P., Nagash, A., 1970. Mineralogy of the suspended sediment in the Tigris, Euphrates and Shatt Al-Arab rivers of Iraq and recent history of the Mesopotamian plain. Journal of Sedimentary Petrology 40, 131–139.

Dobel, A., Asaro, F., Michel, H.V., 1977. Neutron activation analysis and the location of Waššukanni. Orientalia 46, 375–382.

Goren, Y., 2000. Provenance study of the cuneiform texts from Hazor. Israel Exploration Journal 50, 29–42.

Goren, Y., Finkelstein, I., Na'aman, N., 2002. The seat of three disputed Canaanite rulers according to petrographic investigation of the Amarna tablets. Tel Aviv 29, 221–237.

Goren, Y., Bunimovitz, S., Finkelstein, I., Na'aman, N., 2003a. The location of Alašiya: new evidence from petrographic investigation of Alašiyan tablets from el-Amarna and Ugarit. American Journal of Archaeology 107, 233–255.

Goren, Y., Finkelstein, I., Na'aman, N., 2003b. The expansion of the kingdom of Amurru according to the petrographic investigations of the Amarna tablets. Bulletin of the American Schools of Oriental Research 329, 1–11.

Goren, Y., Finkelstein, I., Na'aman, N., 2004. Inscribed in Clay: Provenance Study of the Amarna Tablets and Other Near Eastern Texts. Monograph Series of the Institute of Archaeology, Tel Aviv University, Tel Aviv.

Goren, Y., Na'aman, N., Mommsen, H., Finkelstein, I., 2007. Provenance study and re-evaluation of the cuneiform documents from the Egyptian residency at Tel Aphek. Ägypten und Levante 16, 161–171.

Goren, Y., Mommsen, H., Finkelstein, I., Na'aman, N., 2009. Provenance study of the Gilgamesh fragment from Megiddo. Archaeometry 50, 763–773.

Hoffner, H.A., 1980. Histories and historians of the Ancient Near East. Orientalia Nova Series 49, 283–332.

Katz, A., Agnon, A., Marco, S., 2009. Earthquake-induced barium anomalies in the Lisan formation, Dead Sea rift valley. Israel Earth and Planetary Science Letters 286, 219–229.

Kaufman, M., 2008. The content and origin of the Sumero-Akkadian documents found at Ugarit-Ras Shamra, Unpublished M.A. dissertation, Tel Aviv University (Hebrew, English summary).

Klinger, J., 2003. Zur Paläographie akkadischsprachiger Texte aus Ḫattuša. In: G. Beckman et al. (Eds.), Hittite Studies in Honor of Harry A. Hoffner Jr. on the occasion of His 65th Birthday, Winona Lake, pp. 237–248.

Liangquan, G., 2008. Geochemical prospecting. In: Potts, P., West, M. (Eds.), Portable X-Ray Fluoroscence Spectrometry, Capabilities for In Situ Analysis. RSC Publishing, Cambridge, pp. 141–173.

Mazar, E., Horowitz, W., Oshima, T., Goren, Y., 2010. A cuneiform tablet from the Ophel in Jerusalem. Israel Exploration Journal 60, 4–21.

Mommsen, H., 2004. Provenancing of pottery – the need for an integrated approach? Archaeometry 46, 267–271.

Mommsen, H., Kerschner, M., 2006. Chemical provenance determination of pottery. The example of the Aiolian pottery group G. In: Schlotzhauer, U., Villing, A., (Eds.), Naukratis: Greek Diversity in Egypt. The British Museum Research Publication Number 162, London, pp. 105–108

Mommsen, H., Sjöberg, B.L., 2007. The importance of the 'best relative fit factor' when evaluating elemental concentration data of pottery demonstrated with Mycenaean sherds from Sinda, Cyprus. Archaeometry 49, 357–369.

Moran, W.L., 1992. The Amarna Letters. Johns Hopkins University Press, Baltimore and London.

Morgenstein, M., Redmount, C.A., 2005. Using portable energy dispersive X-ray fluorescence (EDXRF) analysis for on-site study of ceramic sherds at El Hibeh, Egypt. Journal of Archaeological Science 32, 1613–1623.

Na'aman, N., Goren, Y., 2009. The inscriptions from the Egyptian residence: a reassessment. In: Gadot, Y., Yadin, E. (Eds.), Aphek-Antipatris II, the Remains on the Acropolis. The Moshe Kochavi and Pirhiya Beck Excavations, Monograph Series of the Institute of Archaeology, Tel Aviv University, Tel Aviv, pp. 460–471.

Padilla, R., Van Espen, P., Godo Torres, P.P., 2006. The suitability of XRF analysis for compositional classification of archaeological ceramic fabric: a comparison with a previous NAA study. Analytica Chimica Acta 558, 283–289.

Philip, G., 1968. Mineralogy of recent sediments of Tigris and Euphrates rivers and some of the older deposits. Journal of Sedimentary Petrology 38, 35–44.

Radu, T., Diamond, D., 2009. Comparison of soil pollution concentrations determined using AAS and portable XRF techniques. Journal of Hazardous Materials 171, 1168–1171.

Schneiderman, J.S., 1995. Detrital opaque oxides as provenance indicators in River Nile sediments. Journal of Sedimentary Research 65, 668–674.

Singer, I., 1983. Western Anatolia in the thirteenth century B.C. according to Hittite texts. Anatolian Studies 33, 205–218.

Takla, M.A., Arafa, E.H., 1975. The mineralogy of the sand dunes between Minya and Dairut, Egypt. Mineralogy and Petrology 22, 164–173.

van den Hout, Th.P.J., 1994. Der Falke und das Kücken: der neue Pharao und der hethitische Prinz? Zeitschrift für Assyriologie und Vorderasiatische Archäologie 84, 60–88.

Whitbread, I., 1995. Greek Transport Amphorae, a Petrological and Archaeological Study, Athens, The British School at Athens (Fitch Laboratory Occasional Paper 4).