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# ISOTOPIC EVIDENCE FOR THE PRIMARY PRODUCTION, PROVENANCE AND TRADE OF LATE BRONZE AGE GLASS IN THE MEDITERRANEAN

**J. Henderson*[1], J. Evans[2] and K. Nikita[1]**

[1]*Department of Archaeology, University of Nottingham, University Park, Nottingham, NG7 2RD, UK*
[2]*NERC Isotope Geosciences Laboratory, Keyworth, Nottingham NG12 5GG, UK*

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*Corresponding author: julian.henderson@nottingham.ac.uk*

## ABSTRACT

The earliest known man made glass comes from Mesopotamia and dates to the 23rd century BC. By the 16th century BC the first glass vessels appear in Mesopotamia, but the earliest evidence for the fusion of glass from raw materials has been found at the 13th century BC Egyptian site of Qantir. Chemical analyses of this elite Late Bronze Age material have produced compositional distinctions between glasses found in Mesopotamia and Egypt. It is however debatable whether trace element concentrations provide a (geological) provenance for the glasses. By using neodymium and strontium isotopes to fingerprint well-dated chemically analysed 15th to 11th century BC glass samples, we show that independent primary production probably occurred in both Egypt and Mesopotamia in the 14th century BC, and that both of these areas exported glass to Greece. We also discuss the technological implications for glass manufacture and colouring that these new data provide. The results add significant new scientific evidence for glass trade between Late Bronze Age palatial societies. Moreover, it is the first time that this methodology has been used to investigate Bronze Age glass.

**KEYWORDS:** glass, isotopes, provenance, production, trade, technology

## INTRODUCTION

The first man-made glass beads and pendants were made around 2300 BC in the area of modern Iraq and northern Syria (Mesopotamia) (Oppenheim 1973), with the first strikingly coloured (core-formed) vessels appearing there in the 16th/ 15th centuries BC (Moorey 1994, 193). Glass technology emerged from a background in metal and faience production, but, unlike these materials, its manufacture represents a fundamental transformation of raw materials (crushed quartz or sand, plant ash and colorants) into a very different material. This first truly synthetic material represented an important step change in ancient technology. Moreover, there must have been a reason why very detailed and extensive descriptions of the manufacture of glassy materials in cuneiform texts dating to between the 7th and 14th centuries, and possibly earlier, exist (Brill 1970, Moorey 1994, 211). There is nothing comparable for other technologies. The most likely interpretation seems to be that the *colour* produced in glass, in imitation of semi-precious stones, had strong ritual, social and political significance. Colour may therefore have been the primary driver for the production of this material in Late Bronze Age societies (Robson 2001, 51-53). It has been suggested that a technologically innovative group called the Hurrians, who controlled the state of Mitanni (Moorey 1989) in northern Mesopotamia and who were dominant among the Hittites and in Kizzuwatna (in south west Anatolia), were responsible for the production of the first glass vessels (Moorey 1994). If this was the case, they must also have invented special glass furnaces capable of accommodating large enough volumes of hot glass to make vessels. Up to that time, glass had only been used for making beads. The high ritual, social and political value of glass led to an increase in demand and to exchange and trade across the Mediterranean. Much of it was produced under Royal patronage in Mesopotamia within the ambit of palaces (Van der Mieroop 2004, 65).

By the mid 2nd millennium BC glass was being used by strongly hierarchical Late Bronze Age societies in three principle areas: Mesopotamia, Egypt and Greece (Nolte 1968, 154; Barag 1970). Only one (13th century BC) site, Qantir, has produced evidence for the fusion of glass from its raw materials (Pusch and Rehren 2007). Historical references to glass production give an indication of where it might have been fused from raw materials, but do not provide definitive evidence. Redford (1990, 46) argues that 'numerous stones-of-casting' imported from an unknown land (presumably Mesopotamia) after Tuthmosis III's seventh campaign is, in fact, glass. A 14th century BC request in the Amarna letters by an Egyptian Royal House for glass to be sent from a Mesopotamian royal house (Moran 1992, 235) has suggested to some that the Egyptians were dependent on the Mesopotamians for their glass. Excavations of an early 13th century shipwreck at Ulu Burun (Kaş) off the coast of Turkey have provided comprehensive evidence for trade in cobalt blue, manganese purple and copper-rich turquoise glass ingots (Bass 1986; Pulak 2001); it was probably travelling from east to west (Bachhuber 2006). Without evidence of primary production of glass in Egypt and Mesopotamia the sources of the glass *in the 14th century BC* remain somewhat speculative (Nicholson 2007).

Chemical analyses of Late Bronze Age glass from the eastern and southern Mediterranean indicate that it is of a soda-lime-silica composition (Sayre and Smith 1961). Plant ashes used to make the glass produce characteristically elevated levels of magnesium oxide (c. 2-6%) and, in many cases, potassium oxide (c. 0.5-4.0% $K_2O$) in it. A number of complex factors can determine the chemical compositions of such glasses:

(1) the chemical composition of the raw materials;
(2) the geological origins of (ashed) plants (Barkoudah and Henderson 2006) and silica (Turner 1956);
(3) the *genus* or species of the (ashed) plants used (Barkoudah and Henderson 2006);
(4) the raw material preparation and purification techniques used;
(5) the proportion of raw materials used;
(6) partial batch melting, whereby glass melts follow the course of the lowest eutectic trough and minimum temperature, so that glass compositions produced do *not* reflect the summed total chemical compositions of the raw material used (Rehren 2000). Rehren (2008) notes that alkali partitioning between co-existing silica and salt melts can over-ride the primary element signatures *and* ratios found in raw materials;
(7) the presence of buffering materials, such as lime in the crucibles used to melt the glass (Rehren and Pusch 2005, 1757);
(8) the addition of scrap glass (cullet);
(9) the addition of glass colorants, decolorants and clarifiers.

The colorant and opacifying materials added to the glass have been the subject of much discussion (Rehren 1997, Henderson 2000a, Lilyquist *et al* 1993, Mass *et al* 2002). Slight variations in production processes, the raw materials used (especially silica), and the addition of colorants, can provide separate chemical groupings for some Late Bronze Age Mesopotamian, Egyptian (Shortland *et al*. 2007) and Greek glasses (Nikita and Henderson 2006; Walton and Shortland 2009) with trace elements providing the clearest groupings. Whether these groupings can be regarded as providing a true *provenance* is still not completely clear (Rehren 2008). The key question is whether Bronze Age glass chemical compositions are a true reflection of the *local* soil geochemistry (Shortland 2005) of where the glasses were made (the compositions of silica and plant ash used to make the glass and, less likely, any soil incorporated in the melt) and therefore of the primary production sites involved. As noted recently (Degryse et al 2009, 68), glass samples falling into the same chemical group can have markedly different isotopic signatures suggesting that they were made in different geological zones. This underlines the interdependence of chemical and isotopic analyses and that we are only just beginning to understand the complexities involved.

Being able to show with confidence where this potentially valuable material was made provides a means of addressing several key questions about its place in Late Bronze Age society. For instance, it might eventually be possible to address the following questions: were there several glass production centres tied to individual 'states'?; is there evidence of specialisation in the glass colours produced?; in which direction(s) was the glass exported?; was Egypt dependent on Mesopotamia for *raw* glass?; which glass colours were in demand in Egypt, Mesopotamia and Greece, potentially leading to trade?; which patterns of trade involving glass existed with Mycenaean palatial states?

To address these crucial questions, we have, for the first time, determined isotopic signatures (neodymium and strontium) in some of the oldest preserved glasses, including glass ingots and raw glass from Mesopotamia, Egypt and Greece. These results provide new scientific evidence of provenance, the direction of glass trade, glass coloration and a range of technological information.

## WHERE WERE THE PRODUCTION CENTRES?

The distinction between primary and secondary glass production is an important one. The *primary production* of glass in the Late Bronze Age Mediterranean involved the fusion of plant ashes and a silica source; *secondary glass production* involved reheating and working raw or ingot glass into vessels, beads and pendants. The earliest glass appears in the Late Bronze Age, c. 24th -23rd century B.C. However, the archaeological evidence for centres of *primary glass production* is virtually absent, without any archaeological evidence in either Bronze Age Mesopotamia or Greece. Only recently has the 13th century Rameside site of Qantir on the Nile delta in Egypt (Fig. 1) provided the earliest archaeological evidence for the fusion of plant ash and silica to produce opaque red copper-coloured glass in the form of ingots (Rehren and Pusch 2005, Pusch and Rehren, 2007).

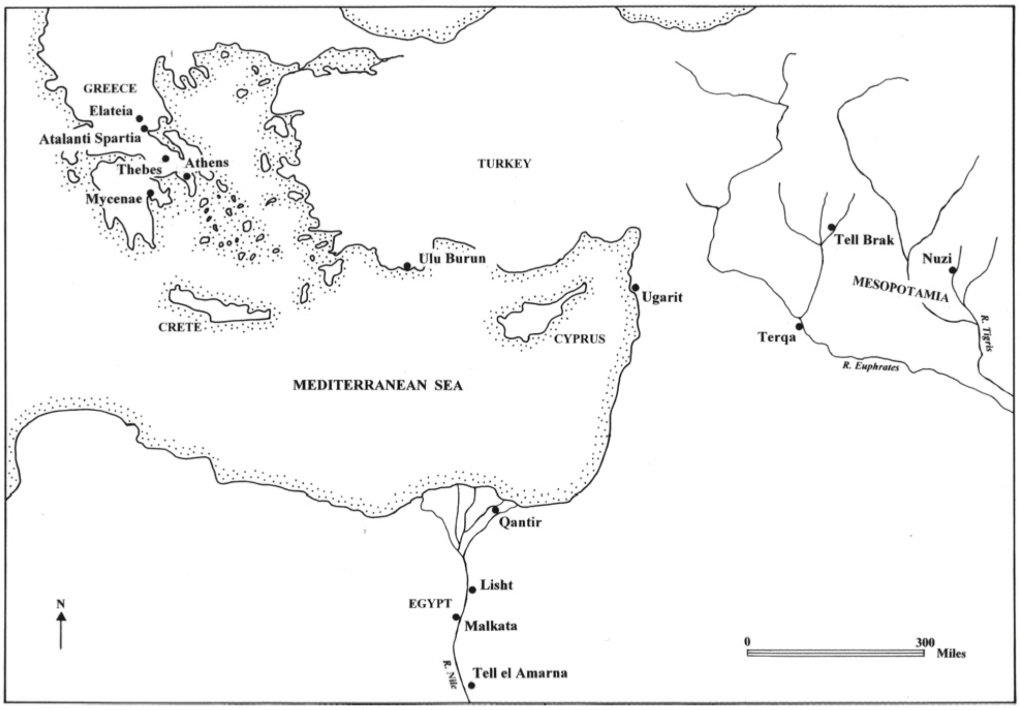

**Fig. 1. Map of the Mediterranean showing sites where evidence for Bronze Age glass production occurred, including those from which samples have been derived.**

There is no evidence for primary glass production from earlier periods, including the 14th century BC, the key period of social and political change in Egypt under the heretic Pharoah Akhenaten. The evidence suggests that secondary glass production occurred in the 14th century BC and, judging from the volume of glass that has survived, it was a time when primary glass production must have occurred on an increasingly large scale. A range of evidence for *secondary glass production* has been found at the 14th century BC site of Tel el-Amarna in Egypt including raw cobalt blue glass in a cylindrical vessel, a turquoise blue glass ingot, recently recognised fragments of raw (including ingot) glass (Fig 2b) and coloured glass canes (Nicholson 2007). Furnaces have been found, although it has not been possible to prove that they were used for primary glass production (Henderson 2000b, 40-41; Nicholson 2007, 127). This evidence suggests that glass working occurred at Tell el-Amarna, including ingot casting, and perhaps vessel production, but overall it does not (yet) constitute definitive evidence for primary glass production. Other Egyptian sites that have produced evidence of glass-working, dating to both before and after Amarna, are Malkata, Lisht, Menshiyeh and possibly Medinet Gurob (Keller 1983) (Fig.1). An Egyptian source of cobalt alum has been recognised for more than 20 years and its use in the coloration of Egyptian faience and glass (Kaczmarczyk and Hedges 1983; Kaczmarczyk 1986, Sayre and Smith 1974, Rehren 2001, Tite and Shortland 2003). Whilst this shows that this cobalt source was used, it does not by itself *necessarily* show that raw glass was made in Egypt in the 14th century BC.

There is no archaeological evidence for *primary glass production* in Bronze Age Greece or Mesopotamia. Diagnostically Mycenaean jewellery has been found in Greece and one possibility is that primary glass making occurred there (Nikita and Henderson 2006). However, it is in Mesopotamia that the earliest glass vessels occur, dating to *c.* 1600 BC. So presumably it is only a matter of time before evidence for primary glass production will be found there. There is evidence of *secondary glass production* from a number of sites in Mesopotamia (Fig.1), such as raw and ingot glass and mis-shapen vessel fragments, sometimes accompanied by large concentrations of glass artefacts, at sites such as Nuzi, Kar-Tukulti-Ninurta (Iraq), Tell Brak (Syria) (Fig. 2), Alalakh/ Tell Atchana (Turkey), Ugarit (Syria) and Tell Ashara- Terqa (Syria) (fig. 1). In the Aegean, probable glass-working centres have been identified at Thebes, Tiryns and Mycenae, with other possible examples on Crete (Nikita 2003).

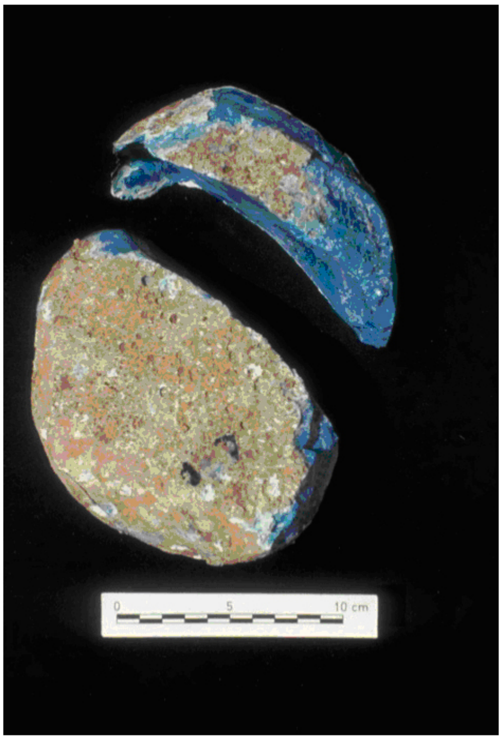

**Fig. 2a A 14th century BC opaque turquoise glass ingot from Tell Brak, in northern Syria 'Reproduced with kind permission of Dr. Joan Oates'**

## MATERIALS AND METHODS

### *Materials*

Samples of glass were removed from ingots, raw glass fragments, a vessel fragment and beads. The samples used in this study derived from sites in Mesopotamia, Egypt and Greece. Their origins, artefact types, date and colour are listed in (Table 1), along with the isotope results for each.

More detailed descriptions of each sample are given in the Appendix. The samples include some of ingots (Fig 2a) and raw glass, including previously unpublished raw glass from Tell el Amarna, Egypt (Fig 2b). The reason why raw and ingot glasses were sampled is that they are liable to be in their original manufactured state and therefore are least likely to consist of mixed glasses from a range of origins.

**Fig. 2b. Three chunks of newly identified highly coloured raw glass from Tell el Amarna from the Ashmolean Museum. The top two show clear conchoidal fractures and 'bulbs of percussions'. They have all been removed from larger pieces, probably ingots (the fragment shown in the top right measures 4.5cm across)**

Each sample has been analysed chemically using electron probe microanalysis (EPMA) and isotopically using thermal ionisation mass spectrometry (TIMS). These two techniques have been described in detail elsewhere (Henderson 1988 and Henderson *et al*. 2009 respectively) and a summary of each follows.

## METHODS

### (1) Electron probe microanalysis

Chemical analyses were primarily carried out using a Jeol JSM8200 Superprobe in the Department of Archaeology, University of Nottingham equipped with 4 wavelength-dispersive spectrometers, one energy-dispersive spectrometer and secondary and backscattered detectors. [The trace concentrations of strontium and neodymium were determined as part of the isotope determinations-see below]. The system was calibrated using a series of geological and multi-element glass standards. The glass samples were mounted in epoxy resin and polished flat using a series of increasingly fine grades of polishing powders, finishing with 0.25μm grade diamond paste. The samples were then coated with a thin layer of carbon so as to prevent distortion and deflection of the electron beam. Analyses were performed at at 20 K.V. accelerating voltage and 40nA beam current using a defocused electron beam of 40μm in diameter so as to prevent the volatilisation of low-Z components, such as sodium. Multi-element glass standards were analysed on a regular basis so as to detect any change in the performance of the system and so as to establish its accuracy and precision.

The following relative analytical accuracies were obtained using a Corning B standard as unknown: 4% for $Na_2O$; 1% for $SiO_2$; 3% for $K_2O$; 5% for CaO and 2% for PbO. For minor elements the accuracy was 2% for MgO, 1% for $Al_2O_3$, 3% for $P_2O_5$, 10% for $Fe_2O_3$ and CuO and up to 20% for Cl. The levels of detection varied from 170ppm for CaO to 1200ppm for CuO. A ZAF program was used to correct and quantify the results. The system allowed opacifying crystals in the glasses to be analysed separately and photographed under high magnification.

### (2) Thermal ionisation mass spectrometry

Glass samples were broken into fragments, cleaned by placing in 2.5 molar HCl in an ultrasonic bath for 5 minutes and rinsed thrice in de-ionised water for the same length of time. The fragments, once dry, were milled to a fine powder in an agate micro ball mill. Standard silicate dissolution was then undertaken. The strontium and REE fractions were collected from Dowex resin columns, and the Nd was separated from the REE fraction using Ln resin (Horwitz and Bloomquist 1975).

Nd and Sr concentrations were determined by isotope dilution using enriched tracer solutions of $^{150}$Nd and $^{84}$Sr respectively. Neodymium concentration and isotope ratios were measured using a double filament assemblage with Thermo Triton multi-collector. Strontium was measured using a rhenium single filament with TaF activator after the method of Birck (1986). The reproducibility of the data is based on standard data runs during sample analysis using the NBS987 international strontium standard and the international La Jolla standard for Nd. The results were: NBS987 = 0.710282 ± 0.000005 ($2\sigma$, n=9), La Jolla = 0.511848 ± 0.000005, ($2\sigma$, n=3). $^{87}$Sr/$^{86}$Sr data were normalized to NBS987 = 0.710250. Samples were run to a precision of better than ± 0.00001 (2SE) for Sr and better than 0.00002 (2SE) for Nd. The lower Nd precision was due to the very small samples and low concentrations in some of the material. All data are corrected to a $^{146}$Nd/$^{144}$Nd ratio of 0.7219 and a $^{86}$Sr/$^{88}$Sr ratio of 0.1194. Blank values were *c.* 100pg Nd, and c. 50pg Sr.

The EPMA results are given in the Appendix. They provide the relative levels of major, minor and some trace components in glasses. The technique provides evidence for the raw materials used to make glasses, including colorants. Thermal ionisation mass spectrometry is used to produce isotope ratios in glasses. The aim is to highlight contrasts in the geological ages of the raw materials used to make glasses (as reflected in e.g. strontium and neodymium isotope ratios) and, by comparing the results with environmental samples, provide information about *where* the glasses were fused from raw materials, and therefore produce a *geological provenance*.

## THE ISOTOPE RESULTS (SEE TABLE 1)

The isotope results plotted in Figs. 3a-d show that Mesopotamian glass is distinctive and can be separated from Egyptian and Greek glasses (Table 1); Egyptian and Greek glasses form a single isotopic field of data in the diagrams. The Mesopotamian glass is characterised by higher $^{143}$Nd/$^{144}$Nd ratios, of between 0.51233 and 0.51253, a relatively wide range of $^{87}$Sr/$^{86}$Sr ratios of between 0.70798 and 0.70871 (Fig. 3a), very low neodymium concentrations of between 0.72 and 1.71 ppm and restricted strontium concentrations between 244 and 449 ppm (Figs. 3b, 3c and 3d). There is no correlation between the strontium and neodymium concentrations in Mesopotamian glass.

In contrast, the Greek and Egyptian glasses have lower neodymium isotope ratios, of between 0.51215 and 0.51224, lower and more restricted strontium isotope compositions of between 0.70788 and 0.70825 (Fig. 3a), and their variable strontium and neodymium concentrations are correlated, forming two elongated fields (Fig. 3d). Therefore, it is clear that the Mesopotamian glass was made using raw materials with different isotopic characteristics from the Egyptian and Greek glass.

**Table 1: Strondium and neodymium isotope and concentration results for Mesopotamian, Egyptian and Greek samples**

| Sample | Glass object | Sr ppm | $^{87}Sr/^{86}Sr$ | Nd ppm | $^{143}Nd/^{144}Nd$ |
|---|---|---|---|---|---|
| Mesopotamia (14th century BC) | | | | | |
| Brak-01 | Brak Pale opaque turquoise ingot | 412 | 0.70849 | 0.72 | 0.51235 |
| Brak-02 | Brak Translucent brown raw fragment | 449 | 0.70798 | 1.03 | 0.51233 |
| Brak-08 | Brak Translucent turquoise bead | 340 | 0.70829 | 1.14 | 0.51233 |
| Brak-09 | Brak Translucent brown raw fragment | 308 | 0.70813 | 0.84 | 0.51237 |
| Brak-12 | Brak Pale opaque turquoise ingot | 407 | 0.70871 | 1.67 | 0.51235 |
| Brak-14 | Brak Opaque cobalt blue fragment | 280 | 0.70856 | 1.66 | 0.51240 |
| Brak-16 | Brak Pale opaque turquoise ingot | 330 | 0.70867 | 0.73 | 0.51253 |
| Brak-17 | Brak Translucent brown bead fragment | 324 | 0.70859 | 1.71 | 0.51236 |
| Egypt | (14th century BC) | | | | |
| Am-03 | Amarna (AM 1893.1-41= 403J) Translucent cobalt blue ingot fragment | 1068 | 0.70791 | 5.95 | 0.51223 |
| Am-04 | Amarna (AM403K) Translucent cobalt blue flake, possible ingot fragment | 332 | 0.70798 | 3.99 | 0.51215 |
| AM-05 | Amarna (AM403L) Translucent purple raw glass, possible ingot fragment | 346 | 0.70817 | 2.15 | nd |
| AM-06 | Amarna (AM403M) Translucent brown raw glass fragment. One flat face | 756 | 0.70803 | 3.38 | 0.51221 |
| AM-07 | Amarna (AM403N) Nearly colourless raw glass flake with pale green tint | 526 | 0.70805 | 2.40 | 0.51224 |
| AM-09 | Amarna (AM 1924.92) Translucent deep purple vessel fragment (with combed opaque white and yellow decoration). | 1275 | 0.70788 | 3.65 | 0.51224 |
| Greece | | | | | |
| KN 1 | Athens Translucent turquoise green bead, so-called Nuzi type, 15th century BC | 295 | 0.70825 | 1.70 | 0.51232 |
| MK1966 | Thebes Translucent dark cobalt blue plaque 13th century BC | 367 | 0.70793 | 5.30 | 0.51216 |
| KN-98-224 | Elateia Translucent dark cobalt blue plaque 11th century BC | 308 | 0.70795 | 4.303 | 0.51215 |
| KN-P-134-251 | Thebes Translucent dark cobalt blue pendant 13th century BC | 880 | 0.70803 | 11.320 | 0.51222 |
| KN-96-216 | Elateia Translucent dark cobalt blue bead 14thcentury BC | 516 | 0.70793 | 7.073 | 0.51216 |
| KN-138-121 | Thebes Translucent turquoise calf 13th century BC | 922 | 0.70790 | 6.541 | 0.51222 |
| KN-I-69-132* | Elateia Translucent dark cobalt blue plaque 12th century BC | 597 | 0.70800 | nd | nd |
| KN-I-65-126* | Elateia Translucent dark cobalt blue plaque 12th century BC | 390 | 0.70800 | nd | nd |
| KN-18-39* | Atalanti-Spartia Translucent dark cobalt blue plaque 12th century BC | 392 | 0.70790 | nd | nd |
| KN-M-100-363* | Thebes Translucent dark cobalt blue bead 13th century BC | 3035 | 0.70795 | nd | nd |

* The samples were too small to enable reliable Nd data to be determined; nd= nt determined

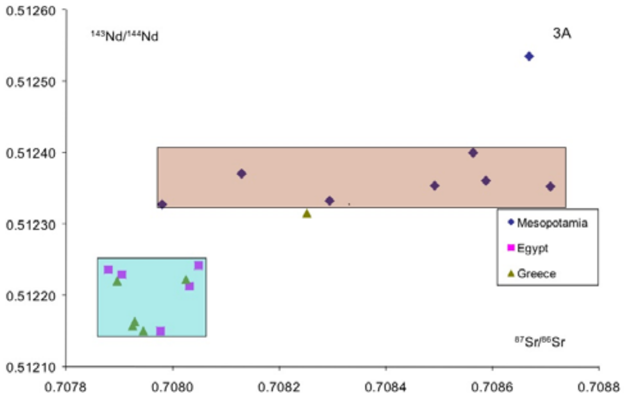

**Fig. 3a. A plot of $^{87}$Sr/$^{86}$Sr vs. $^{143}$Nd/$^{144}$Nd isotope measurements in Bronze Age glass from Mesopotamia (lozenges), Egypt (squares) and Greece (triangles).The plot shows that Mesopotamian glasses have different $^{87}$Sr/$^{86}$Sr and $^{143}$Nd/$^{144}$Nd signatures from Greek and Egyptian glasses. The latter form a tight group.**

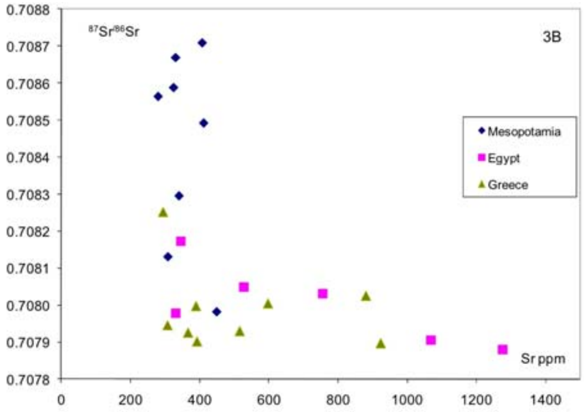

**Fig. 3b. A plot of $^{87}$Sr/$^{86}$Sr vs. Sr concentrations in Bronze Age glass from Mesopotamia (lozenges), Egypt (squares) and Greece (triangles). Mesopotamian glasses contain lower concentrations of Sr than Greek and Egyptian glasses, and are also characterised by a wider range of $^{87}$Sr/$^{86}$Sr values.**

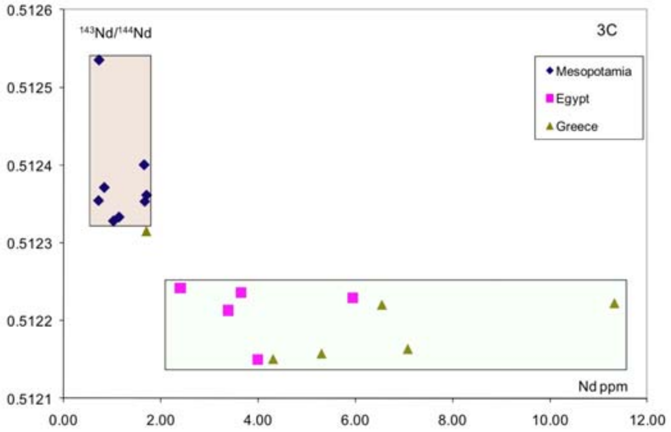

**Fig. 3c. A plot of $^{143}$Nd/$^{144}$Nd vs. Nd concentrations in Bronze Age glass from Mesopotamia (lozenges), Egypt (squares) and Greece (triangles). Mesopotamian glasses contain lower and more constrained concentrations of Nd than Greek and Egyptian glasses and are also characterised by a wider range of $^{143}$Nd/$^{144}$Nd signatures.**

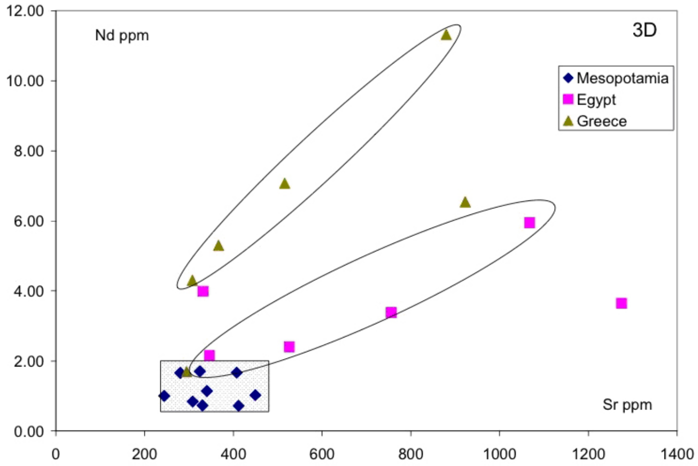

**Fig. 3d. A plot of Sr vs. Nd concentrations in Bronze Age glass from Mesopotamia (lozenges), Egypt (squares) and Greece (triangles). Mesopotamian glasses contain lower concentrations of both Nd and Sr than most Greek and Egyptian glasses. Some of the Greek and Egyptian glasses form two positively correlated groups.**

In contrast, the Greek and Egyptian glasses have lower neodymium isotope ratios, of between 0.51215 and 0.51224, lower and more restricted strontium isotope compositions of between 0.70788 and 0.70825 (Fig. 3a), and their variable strontium and neodymium concentrations are correlated, forming two elongated fields (Fig. 3d). Therefore, it is clear that the Mesopotamian glass was made using raw materials with different isotopic characteristics from the Egyptian and Greek glass.

## THE GEOLOGY: $^{143}$ND/$^{144}$ND AND $^{87}$SR/$^{86}$SR VARIATIONS

If the Egyptian and Greek glasses were produced in the same region, as is suggested by common chemical *and* isotopic features, then the question is: was this in Egypt or in Greece? In order to address this question, the *isotopic* characteristics of potential silica sources (a reflection of the age and formation histories of the silica) in the areas concerned need be considered.

The sediment run off from the Aegean region of Greece and Turkey is dominated by the erosive products of the Pindus and Taurus Mountains. These mountains contain considerable quantities of basaltic and associated materials (Asch 2005) and this is reflected in isotope systematics of rocks from this area which tend to give εNd values above -5, (>0.5124) and low $^{87}$Sr/$^{86}$Sr values (Lease and Abdel-Fattah 2008, Pe-Piper and Piper 2001, Cullen *et al.* 2000). The majority of isotope data available for Egyptian sediments come from studies of the Nile and its sediment load. This is dominated (95%) by, basalt-derived, volcanogenic material from the Ethiopian Highlands (Grousset et al. 1988, Freydier *et al*. 2001) and has εNd values of *c*. -3.3 (Goldstein *et al*. 1984). Scrivener *et al.* (2004) cite a εNd range of -1 to -1.5 as being typical of the Nile; Tachikawa *et al.* (2004) document a value of Nd -1.9 from a leached sediment. The best current indication of neodymium sand composition from Egypt is probably the two sand samples from Wadi el Natrun (Degryse and Schneider 2008), which give values of 0.512289 and 0.512199 (εNd = -6.8 and -8.6 respectively).

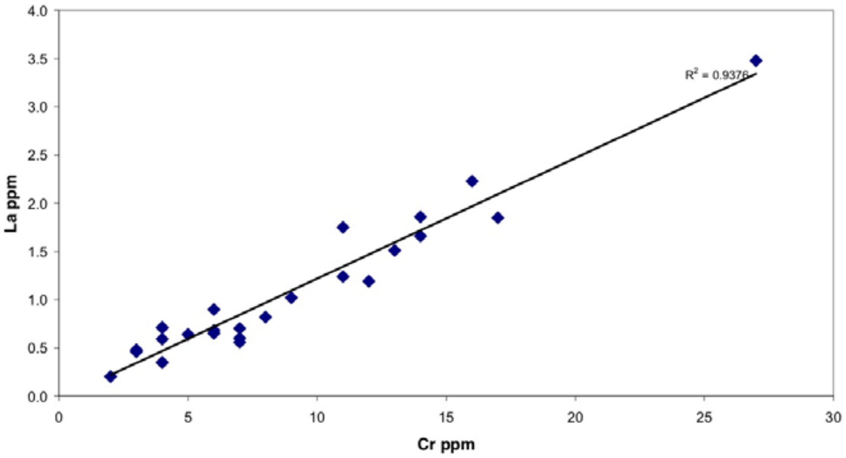

**Fig 4: Positively correlated levels of La and Cr in halophytic plant ashes of a range of genera from Syria based on data published in Barkoudah and Henderson 2006.**

The next thing to consider is whether the Sahara desert contributes to Egyptian silica sources. The εNd for aerosols derived from the Sahara give a range of -14.2 to -10.5 (Grousset etal 1988) providing evidence for a source of un-radiogenic sedimentary material that may have influenced the Egyptian soils and sediments, which incorporate sands. Based on a comparison of the neodymium isotope compositions of the glass with possible quartz sources, it is likely that glass found at Amarna was made in Egypt. In support of this observation, Levantine sands (Degryse and Schneider 2008) are too radiogenic to provide a source for the Greek/Egyptian glass, whereas one of two Wadi el Natrun sand samples (100kms north-west of Cairo) plot within the Greek/Egyptian field showing that the appropriate values can be obtained from inland Egyptian sources.

## MESOPOTAMIAN GLASS TECHNOLOGY

The very low neodymium concentrations in Mesopotamian glass (Table 1 and Fig. 3D) are consistent with the use of a very clean silica source, such as (crushed) quartz pebbles rather than a less pure sand source as suggested by Shortland *et al.* (2007, 788). The silica used would have introduced the dominant Nd component; Nd levels in plants of the genus *Salsola*, which are likely to have been used to make glass, are mainly below 1ppm (Barkoudah and Henderson, Table 2). No mixing line is apparent in the $^{87}Sr/^{86}Sr$ data (Fig. 3c) that might indicate that two sources of silica of different geological ages have been mixed. The use of quartz is probably referred to in (Bronze Age) cuneiform texts that describe the production of a vitreous material, probably glass, as *immanakku* (Brill 1970, 109-10). The translation of one passage from tablet C§4 (Oppenheim 1970, 35) refers to its use in the following way:

'If you want to produce *zagindurû*-coloured glass, you grind finely, separately, ten minas of *immanakku*-stone, fifteen minas of naga-plant ashes (and) 1²/₃ minas of "white plant." You mix these together. You put them into a cold kiln which has four fire openings (literally: eyes)'.

Shortland *et al.* (2007, 788) have suggested that the relatively high levels of chromium found in Mesopotamian glasses are due to the use of chromium-bearing sands derived from the erosion of ultra mafic rocks at the headwaters of the Euphrates and Tigris rivers. However, trace element analysis of Syrian halophytic plants shows that their ashes could be the source of the chromium instead (especially if quartz was used), providing one of the compositional characteristics of late Bronze Age glass found in Mesopotamia.

A plot of relative levels of La and Cr in halophytic Syrian plants of a range of genera (data from Barkoudah and Henderson 2006) shows a positive correlation with an $R^2$ value of 0.9376 (Fig.4), also found in Mesopotamian late bronze age glasses (Shortland et al 2007, Fig. 4).

Variations in lanthanum and chromium concentrations in plant ashes are a reflection of geochemical variations in the semi-desert soils in which the plants grow. The range of chromium concentrations detected in *northern* Syrian plant ash samples of the genus *Salsola*, a plant genus thought to have been used for making ancient glass (Ashtor 1992; Henderson and Barkoudah 2006), is between 4 and 14 ppm. These levels fall within the range detected in Mesopotamian glasses, of between <0.83 and 24.8 ppm (Shortland et al 2007). Therefore, a Cr contribution from plant ashes, could have provided a proportion of the chromium in the glass, or possibly all of it if an especially pure silica source was used. Therefore, chromium can not necessarily be used as a means of characterising the silica source used to make Bronze Age plant ash glasses. An apparent lack of published trace element data for Egyptian halophytic plant ashes makes it impossible to compare their chromium contents with Syrian plant ashes and to compare chromium levels in them with late Bronze Age Egyptian plant ash glasses.

The low neodymium levels in Mesopotamian glasses reported here indicate that silicates were *not* introduced in other components used in the glass-making, such as in the plant ash or in the melting crucibles. The geologically "young" radiogenic $^{143}$Nd/$^{144}$Nd signature shows that silicates from a source such as the river Euphrates were used, deriving from erosion of the Turkish mountains to the north. Furthermore, the restricted *range* of both neodymium and strontium concentrations in Mesopotamian glass suggests that a tightly controlled technique was employed to make it, both in terms of raw material selection and the processes involved in selecting and ashing the plants to produce the flux. On the other hand, the diversity of the $^{87}$Sr/$^{86}$Sr ratios suggests that plant material was collected from a number of geologically different sites. This raises the question as to whether separate production zones within Mesopotamia can be suggested using this isotopic data.

Within the Mesopotamian group in Fig. 3a the *opaque* turquoise ingots from Tell Brak, Syria (Brak 01, 12 and 16) (such as in Fig 2a), form part of the group with the highest strontium isotope values, with a range of between 0.70849 and 0.70871, close to a north Syrian strontium signature (Henderson *et al*. 2005; Henderson *et al.* 2009, Table 3.1, Figure 3.3). This group also includes an opaque cobalt blue glass fragment, which contains lower alumina levels than found in many Egyptian cobalt blue glasses (see appendix), and a brown bead. The 15th century BC *translucent* turquoise bead found in Athens, which, on typological grounds can be considered to have been made in Nuzi, Iraq, is close to another *translucent* turquoise bead from

Tell Brak (strontium values of 0.70825 and 0.70829) and must have therefore been imported from Mesopotamia. Strontium values for two translucent brown fragments from Tell Brak have the lowest strontium values (0.70798 and 070813).

Overall, these results for Mesopotamian glass may indicate that the translucent turquoise glass and some translucent brown glasses were made in a separate production centre from the group containing opaque turquoise ingots. There are no sub-groupings apparent amongst the trace element data currently available for Mesopotamian glasses so Shortland *et al.* (2007, 787), have suggested that 'very similar raw materials, perhaps even the same raw materials from the same place' were used to make glasses from different Mesopotamian sites on the basis of such analyses. However our strontium data clearly show that plant ashes from contrasting geological locations have been used to make Mesopotamian glasses. This again underlines the interdependence of chemical and isotopic analyses, that the results of each technique provide us with different kinds of information and how important it is to conduct both types of analyses.

In all of these diagrams a 13th century BC cobalt blue plaque from Thebes, Greece, plots within or very close to the Mesopotamian field suggesting that this glass was exported to Greece from a Mesopotamian source.

## EGYPTIAN GLASS TECHNOLOGY

The isotopic similarities between the Greek and Egyptian glass suggest that similar raw materials with the same geological origin were used to make them, and possibly that they were made at the same site: given the range of archaeological evidence for glass production Tell el Amarna is one possible production centre. Restricted *sources* of plants and sand were used to make Egyptian/Greek glass as shown by the restricted range in both strontium and neodymium isotope values (Figs. 3A-3C). However, the manufacturing process seems to have been less controlled than that for Mesopotamian glass, something also suggested using trace elements (Shortland et al 2007). The neodymium concentrations are much higher and more variable than in Mesopotamian glasses. This could attest to the use of a much more heterogeneous silica source, such as sand with varying quantities of other minerals within it, such as feldspar rather than hydrothermal quartz as suggested by Shortland et al (2007, 788). Nevertheless, the relative chromium levels in Mesopotamian glasses, especially those from Nuzi, are also generally higher, and they may well have been introduced in the plant ashes (see above). The variability of the strontium concentrations in Egyptian glasses suggests that plants with a range of chemical compositions were used. However, the positive correlation between the neodymium and strontium concentrations (Fig. 3d) shows that the two are linked. This is not a natural geological relationship; in fact the reverse would be expected (increasing neodymium with decreasing strontium). The most obvious explanation for this correlation is that both neodymium and strontium were introduced in plant ashes. Hence, in the case of Egyptian glass, the neodymium was *not only* introduced with the silica but also in the plant ash. It has been shown that the trace and minor element chemistry of a halophytic plant species varies according to contrasting geology (Barkoudah and Henderson 2006) though the precise reasons for the balance of chemical elements in plants and plant parts is complex, and not completely understood (Watanabe *et al*. 2007).

Plant ashes also provide a source of lime (linked to the strontium isotope results) in glass production. It has been suggested that the interaction of the crucible lining with the glass would contribute calcium to the glass (Rehren 2008) which could potentially modify the strontium isotope signature of the glass. However, in glasses where the concentrations of strontium and neodymium are correlated, as in this case, the dominant source of calcium must be the plant ash. Similarly, the suggestion that the glass melt was contaminated by the implement used to crush the quartz would not lead to this correlation.

The positively correlated group of four Greek and one Egyptian glasses in Fig. 3d, that includes glass samples with the highest neodymium concentrations (max. 11.32ppm), are all of a cobalt blue colour, the other positively correlated group being formed from a range of glass colours. It could be suggested that the correlated group of blue glasses was made from a different species of ashed plant from that used to make the other correlated group. However, *all* of the Egyptian blue glasses for which isotope data are given (including those falling outside the correlated group) are united because they contain low potassium oxide concentrations, a characteristic of some, but not all, Egyptian cobalt blue glasses (Rehren 2001). Therefore, although the isotopic results show that the Egyptian cobalt blue glasses were made in the same or in a similar geological zone (according to their isotopic characteristics) as for other colorants, the elevated neodymium and strontium concentrations nevertheless show that the raw materials used to make the two correlated groups into which blue glasses fall (in Fig. 3d) were of slightly different compositions. They were probably made using plant ashes growing in slightly different geological environments but not necessarily different species.

The isotopic results can also be used to shed light on putative production centres and to investigate whether they specialised in making specific glass colours. Two cobalt blue raw glass fragments from Amarna (Am03 and AM 04) have isotope values (and strontium concentrations) within the Egyptian Sr/Nd group that are almost as wide as the maximum variation observed for all Amarna samples *of a range of colours*. This shows that a range of glass colours was made in the same area as defined by the isotope signatures and *could* suggest that a range of glass colours was made at Amarna. However, one group of isotopic data for *Mesopotamian* glasses, that consists of opaque cobalt blue, opaque turquoise and brown glass, exhibits similar variations of strontium isotope values as found in the Egyptian group, with maximum variations within each group of 0.00029 and 0.00022 respectively. This suggests that these two groups represent the products of individual sites or small regions within Egypt and Mesopotamia respectively.

## DISCUSSION & CONCLUSION

This first set of Nd and Sr isotope results for Late Bronze Age glasses provide increasingly firm evidence that both ingot and non-ingot glass artefacts were made separately in Mesopotamia and Egypt during the Bronze Age. The results appear to confirm the hitherto somewhat tentative archaeological evidence for primary glass production at Amarna in the 14th century BC because they provide new locational information. As noted above, there is certainly evidence for glass working, but evidence of glass making at Amarna is less certain (Nicholson 2007). Furthermore, the results show that none of the 14th century Egyptian glasses from Amarna that were tested were imported from Mesopotamia, or, for that matter, exported from Amarna to Mesopotamia. This indicates that in the 14th century BC, Egypt was *not* completely dependent on Mesopotamia for glass as has been suggested using historical information. Moreover, the clear separation of the isotope results indicates that none of the glasses were mixed, or that glass raw materials were exported from Mesopotamia to Egypt, or *vice-versa*. The samples tested include newly identified raw glass from the Ashmolean Museum, which almost certainly formed parts of ingots (Fig 2b). Such glass would probably have derived directly from the glass making furnace, with minimal risk of 'foreign' glass being mixed with it.

The isotope results for Mesopotamian glasses reveal a relatively wide range of (strontium) values, indicative of variations in the geological ages of the one raw material (plant ash) used to make them. This, in turn, suggests one centre did not have a 'monopoly' over glass production and that separate palatial areas within the Kingdom of Mitanni (or perhaps further afield) controlled primary glass production. It is hoped that more isotopic results for both glass and environmental samples, will provide a more statistically coherent basis for this.

Compared to the Mesopotamian results, those for Egypt show a much more restricted set of (strontium) values, suggesting that one production site was involved in the 14th century BC, Tell el Amarna being a strong candidate. An alternative interpretation is that there was more than one primary production centre, but that variations in the isotope values are insufficient to show this. The only way to investigate this further would be to establish the variations in isotopic signatures for plants and silica across the Egyptian landscape, including for plants growing near centres where evidence for glass production has been found, such as Amarna and Qantir. This would then reveal whether the variation in isotopic results obtained for Amarna glasses match those for the environment near Amarna or across a broader area of northern Egypt.

The importance of colour to ancient society can not be underestimated (e.g Bianchi *et al*. 2002). Our isotope results confirm what has long been suspected, that cobalt blue glass made in imitation of lapis lazuli was probably fused from primary raw materials in 14th century BC Egypt. Therefore we can concur that this was probably *one* Egyptian 'specialisation' using cobalt from Egypt (Jackson and Nicholson 2007, 115); the production of red glass appears to have been another one (Rehren 1997). These isotope results provide further solid evidence that the base glass to which colorants were added was fused in 14th century Egypt. It is clear that Mycenaean Greece was importing raw cobalt glass from Egypt (see also Walton et al 2009). Indeed, it is very likely that the cobalt blue ingots found on the Ulu Burun shipwreck off the coast of Turkey were manufactured in Egypt, especially since that some 'ingot moulds' found at Amarna were probably used to manufacture some of the ingots found at Ulu Burun (Nicholson et al 1997). We have also provided scientific evidence of the primary manufacture of cobalt blue glass in Mesopotamia, so this could have been a second/alternative source for some of the cobalt blue Ulu Burun glass ingots. The determination of isotopic signatures of the Ulu Burun glass ingots would address the question of whether just Egypt or Egypt *and* Mesopotamia were the sources of the blue glass (Nicholson et al 1997). The early 13th century cobalt blue and turquoise glass ingots found on the Ulu Burun shipwreck have different shapes, potentially indicating that both shape and colour are a reflection of the respective origins of the glasses.

A predominant proportion of 14th century glass found at Tell Brak in Mesopotamia is translucent or opaque turquoise and it was therefore coloured with copper rather than cobalt. Our isotope results suggest that glass was fused from raw materials in this area. This might suggest that glass makers in Mesopotamia specialised in the manufacture of copper-coloured glasses. Both in Mesopotamia and Egypt there is the possibility that colorant materials were added to raw glasses as a secondary process, perhaps on separate glass production sites, though this has not be demonstrated archaeologically. Indeed at Qantir both primary glass production and the additions of colorants occurred on the same site.

Our research demonstrates that Greek cobalt blue moulded glass plaques, similar in chemical composition to contemporary Egyptian glasses (Nikita and Henderson 2006), were made out of raw glass imported from Egypt. Given the range of context dates for the Greek samples (15th - 11th century BC) it is evident that the glass trade route between Egypt and Greece must have persisted for several centuries, even if some of the glass was stored in palace workshops for periods of time and/or used as heirlooms; glass continued in use after the collapse of the palaces. Our results show that as early as the 15th century BC one turquoise bead was exported from Mesopotamia to Mycenaean Greece; evidence for this trade has recently been augmented using trace element analysis (Walton et al 2009). On present isotopic evidence we can state that, during the late Bronze Age, glass was imported from both Egypt and Mesopotamia to Greece. This still does not exclude completely the possibility that glass was made independently in Bronze Age Greece.

All of the Egyptian samples tested here are of 14th century BC date. The scientific evidence for Egyptian primary manufacture discussed here extends its chronological range from the 13th century BC to the 14th century. None of the Egyptian glasses tested were proven to have been imported from Mesopotamia. However, the first glass vessels were made in 16th century BC Mesopotamia, and glass vessels have been found in 15th century BC Egyptian contexts (Lilyquist *et al*. 1993). Isotopic studies of the earliest dated Egyptian glass (vessels) would show whether they were made in Egypt and would provide evidence for when technology transfer (*primary glass making*) between Mesopotamia and Egypt occurred and when primary glass making first occurred in Egypt. This transfer possibly involved artisans (Lilyquist *et al*. 1993) who specialised in primary glass making and in vessel production. Technology transfer probably occurred between Mesopotamia and Greece (Barag 1970, 187-93). This would have involved moulding hot glass, a technique that originated in Mesopotamia leading, for example, to the manufacture of the first mosaic glass vessels (Van Saldern 1966, 1970, 207, 213-5). Technology transfer, probably via Minoan Crete to Greece (Nikita 2003, 21), led to the specialized production of intricately decorated glass pendants that became a hallmark of the Mycenaean glass industry.

Overall, the use of radiogenic isotopes has provided a *geological provenance* for glasses made in Egypt and in Mesopotamia. In turn, this has produced firm scientific evidence for the export of Late Bronze Age glass, from Egypt and Mesopotamia to Greece.

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## APPENDIX

### *Detailed information about the samples*

All samples are described in Table 1. Tell Brak samples, all of a 14th century BC date, were excavated by Dr D. Oates and Dr. J. Oates and their detailed descriptions are published (Oates and Oates 1997). The (unpublished) glass artefacts from Tell el Amarna used in this study were excavated by Sir Flinders Petrie and accessioned to the Department of Antiquities, Ashmolean Museum, Oxford in 1893 (AM403) and 1924 (AM3-AM9). The glass samples from Atalanti-Spartia, Athens, Elateia and Thebes, Greece, for which results are given in Table 1, are being prepared for full publication by Dr. Kalliopi Nikita. Detailed descriptions for the unpublished Amarna and Greek glass, together with specific find locations for the Greek glass are as follows:

### *Egyptian samples*

Tell el Amarna.

AM3 Translucent dark blue fragment of raw glass, probably part of an ingot (Ashmolean Museum no. AM403J).

AM4 Translucent dark blue flake, possibly part of an ingot (Ashmolean Museum no. AM403K).

AM5 Translucent pale purple chunk of raw glass with two flat faces, probably part of an ingot (Ashmolean Museum no. AM403L).

AM6 Translucent pale brown raw glass chunk with one flat face (Ashmolean Museum no. 403M).

AM7 Nearly colourless raw flake with a pale green tint (Ashmolean Museum no. AM403N).

AM9 Translucent deep purple vessel fragment with combed opaque yellow and opaque white decoration (Ashmolean Museum no. AM 1924.92).

***Greek samples***

Atalanti-Spartia

KN-18-39 Translucent dark blue double five-petal rosette plaque found in tomb 4-OM7. Late Helladic IIIB-IIIC. Museum of Atalanti AK/D 1584.

Athens

KN1 Translucent turquoise 'Nuzi' type bead found in Promachou and Parthenonos cemetery, tomb 5, no. 89. Late Helladic II.

Elateia

KN-98-224 Translucent dark blue double four petal rosette on oval plaque in moulded relief found in tomb 62-F3-Pit H-OM36σζ. Late Helladic IIIC middle/ Sub-mycenaean. Museum of Atalanti 3596.

KN-96-216 Translucent dark blue Amygdaloidal bead found in tomb 24-Chamber-Pit 3. Late Helladic IIIA-B. Museum of Atalanti AK/D 3866.

KN-I-69-132 Translucent dark blue triple six-petal rosette plaque found in tomb 56-Chamber-Pit A. Late Helladic IIIB-IIIC. Museum of Atalanti.

KN-I-65-126 Translucent dark blue triple six-petal rosette plaque found in tomb 56-Chamber-Pit A. Late Helladic IIIB-IIIC. Museum of Atalanti.

Thebes

MK1966 Translucent dark blue fragment of spherical bead found in the Megalo Kastelli cemetery. Late Helladic IIIB. Museum of Thebes.

KN-P-134-251 Translucent dark blue discoid pendant with papyrus motif found at Megalo Kastelli, 66.XI.302b. Late Helladic IIIB. Museum of Thebes.

KN-138-121 Translucent turquoise reclining calf, moulded in relief found in arsenal, site 3, Kadmeia I-II. Late Helladic IIIB, Museum of Thebes, AM283.

KN-M-100-363 Translucent dark blue globular bead found at the Kordatzis plot. Late Helladic IIIB. Museum of Thebes.

## Results of electron microprobe analyses of glass samples

***Electron microprobe analyses of samples from Tell Brak, Syria***

(The electron microprobe analyses of BRAK-02, BRAK-08, BRAK-12 and BRAK-17 are published in Henderson 1997, Table 6b, p. 96 as Br1a, Br4, Br9 and Br5 respectively).

| | BRAK1 | BRAK9 | BRAK14 | BRAK16 |
|---|---|---|---|---|
| $Na_2O$ | 16.95 | 18.38 | 16.14 | 14.17 |
| MgO | 2.8 | 6.09 | 5.47 | 2.44 |
| $Al_2O_3$ | 0.18 | 0.27 | 0.76 | 0.20 |
| $SiO_2$ | 62.91 | 63.64 | 63.34 | 69.06 |
| $P_2O_5$ | 0.24 | 0.39 | 0.27 | 0.19 |
| $SO_3$ | 0.35 | 0.38 | 0.23 | 0.35 |
| Cl | 0.5 | 0.55 | 0.45 | 0.51 |
| $K_2O$ | 1.7 | 2.71 | 4.42 | 1.42 |
| CaO | 8.35 | 5.82 | 4.98 | 6.04 |
| $TiO_2$ | ND | ND | 0.03 | ND |
| $MnO_2$ | ND | 0.08 | 0.06 | ND |
| $Fe_2O_3$ | 0.14 | 0.13 | 0.79 | 0.11 |
| CoO | ND | 0.05 | 0.24 | ND |
| NiO | ND | ND | 0.06 | ND |
| CuO | 1.25 | ND | 0.32 | 1.7 |
| ZnO | 0.15 | 0.11 | 0.07 | ND |
| BaO | ND | 0.05 | 0.07 | 0.06 |
| $As_2O_3$ | ND | ND | ND | ND |
| $Sb_2O_5$ | 2.53 | 0.06 | 1.67 | 2.34 |
| SnO | ND | 0.05 | ND | ND |
| PbO | ND | 0.05 | 0.05 | 0.14 |
| Total | 98.05 | 98.81 | 99.42 | 98.73 |

Sought but not detected: $Cr_2O_3$, $V_2O_3$, $SrO_2$, $ZrO_2$; ND = Not Detected

**Table 4 Electron microprobe analyses of samples from Tell el Amarna, Egypt**

| | AM3 | AM4 | AM5 | AM6 | AM7 | AM9 |
|---|---|---|---|---|---|---|
| $Na_2O$ | 16.74 | 18.3 | 16.85 | 17.8 | 17.5 | 17.4 |
| MgO | 3.13 | 2.46 | 4.85 | 4.44 | 4.1 | 3.83 |
| $Al_2O_3$ | 1.63 | 1.38 | 0.6 | 0.6 | 0.49 | 0.56 |
| $SiO_2$ | 67 | 70.6 | 65.2 | 64.1 | 64.4 | 65.5 |
| $P_2O_5$ | 0.1 | 0.04 | 0.22 | 0.17 | 0.13 | 0.13 |
| $SO_3$ | 0.38 | 0.28 | 0.28 | 0.38 | 0.42 | 0.38 |
| Cl | 0.87 | 1.28 | 0.85 | 0.86 | 0.73 | 0.82 |
| $K_2O$ | 1 | 0.6 | 2 | 2.34 | 2.64 | 1.78 |
| CaO | 6.9 | 4.15 | 6.2 | 7.46 | 7.8 | 8.9 |
| $TiO_2$ | 0.05 | 0.03 | 0.06 | 0.02 | 0.03 | 0.05 |
| MnO | 0.13 | 0.1 | 0.4 | ND | ND | 0.76 |
| FeO | 0.33 | 0.28 | 0.6 | 0.32 | 0.27 | 0.32 |
| CoO | 0.07 | 0.08 | ND | ND | ND | ND |

| | | | | | | |
|---|---|---|---|---|---|---|
| NiO | 0.05 | 0.05 | ND | ND | ND | ND |
| CuO | 0.03 | 0.8 | ND | ND | 0.03 | ND |
| ZnO | 0.1 | 0.08 | ND | ND | ND | ND |
| BaO | ND | ND | ND | ND | ND | 0.07 |
| $As_2O_5$ | ND | ND | 0.05 | ND | ND | 0.04 |
| $SnO_2$ | ND | ND | ND | ND | ND | ND |
| $Sb_2O_5$ | 0.07 | 0.44 | 0.12 | 0.1 | 0.15 | 0.11 |
| Total | 98.58 | 100.95 | 98.28 | 98.59 | 98.69 | 100.65 |

Sought but not detected: $Cr_2O_3$, $V_2O_3$, $SrO_2$, $ZrO_2$, PbO; ND= Not Detected

**Table 5: Electron microprobe analyses of Greek samples**

(The electron microprobe analyses of KN-98-224 and KN-138-121 are published in Nikita and Henderson 2006: Table 4, as Ela/98 and The/138 respectively.)

| | KN1 | MK-1966 | KN-P-134 | KN-96-216 | KN-I-69 | KN-I-65 | KN-18-39 | KN-M-100-363 |
|---|---|---|---|---|---|---|---|---|
| $Na_2O$ | 17.36 | 19.88 | 18.09 | 19.8 | 17.42 | 17.36 | 19.29 | 20.9 |
| MgO | 4.06 | 2.26 | 4.21 | 2.72 | 2.5 | 2.48 | 2.9 | 5.45 |
| $Al_2O_3$ | 0.55 | 1.5 | 3.02 | 2.07 | 2.19 | 2.15 | 1.91 | 4.07 |
| $SiO_2$ | 68.75 | 66.21 | 63.24 | 64.98 | 69.54 | 69.49 | 64.78 | 60.65 |
| $P_2O_5$ | 0.17 | ND | 0.1 | 0.2 | 0.08 | 0.09 | 0.1 | 0.14 |
| $SO_3$ | 0.25 | 0.08 | 0.46 | 0.28 | 0.32 | 0.26 | 0.3 | 0.33 |
| Cl | 1.09 | 1.28 | 0.74 | 0.98 | 0.99 | 1.01 | 0.72 | 0.99 |
| $K_2O$ | 2.94 | 0.52 | 0.95 | 0.89 | 0.82 | 0.76 | 1.43 | 1.22 |
| CaO | 4.52 | 4.65 | 7.25 | 6.54 | 5.14 | 5.11 | 7.25 | 5.25 |
| $TiO_2$ | ND | 0.08 | 0.09 | 0.24 | 0.08 | 0.07 | 0.08 | 0.14 |
| MnO | ND | 0.28 | 0.21 | 0.34 | 0.21 | 0.2 | 0.18 | 0.27 |
| $Fe_2O_3$ | 0.34 | 0.38 | 0.6 | 0.6 | 0.62 | 0.6 | 0.73 | 0.89 |
| CoO | ND | 0.09 | 0.24 | 0.02 | 0.07 | 0.06 | 0.04 | 0.16 |
| NiO | ND | 0.11 | 0.13 | ND | 0.04 | 0.05 | 0.04 | 0.12 |
| CuO | 0.98 | 0.11 | 0.1 | ND | 0.16 | 0.13 | 1.4 | 0.16 |
| ZnO | ND | 0.14 | 0.26 | 0.05 | 0.08 | 0.08 | 0.05 | 0.21 |
| BaO | ND | 0.59 | ND | ND | ND | 0.06 | ND | 0.06 |
| $As_2O_3$ | 0.07 | 0.11 | 0.07 | ND | ND | ND | ND | ND |
| $Sb_2O_3$ | 0.14 | 0.96 | 1.11 | 0.08 | 0.07 | 0.1 | 0.21 | 0.17 |
| PbO | ND | 0.21 | ND | 0.14 | ND | ND | ND | 0.01 |
| Total | 101.22 | 99.44 | 100.87 | 99.93 | 100.33 | 100.06 | 101.41 | 101.19 |

Sought but not detected: $Cr_2O_3$, $V_2O_3$, $SrO_2$, $ZrO_2$; ND=Not Detected