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Glass ingots from the Uluburun shipwreck: Glass by the batch in the Late Bronze Age

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ABSTRACT

The glass ingots from the Late Bronze Age Uluburun shipwreck (ca. 1300 BCE) provide crucial insight into Late Bronze Age glass production and exchange in the Mediterranean. Almost all of the approximately 200 glass ingots on board the ship were sampled as well as five of the 30 Mycenaean glass relief beads. Here we report the full chemical compositional results for these samples, along with 49 strontium isotope analyses representing at least 48 separate glass ingots. We include as well 355 unpublished analyses of Late Bronze Age Egyptian, Mesopotamian, Levantine, and Aegean glasses; these represent a total of 222 different objects since many of the core-formed vessel fragments are polychrome and each color was analysed separately. The results suggest that the Uluburun glass ingots were produced in as few as 28 discrete production events or batches. The largest batch included at least 16 ingots representing 40 kg of glass with a chemical composition as perfectly uniform as could be determined by our LA-ICP-MS analyses. Cluster analysis and comparison of Ti/Cr and Li/Zr ratios indicate that all of the ingots are Egyptian glass. In addition, based on this new dataset we have identified the first Egyptian glasses to be found at Mesopotamian sites as well as several examples of Mesopotamian glass used to produce Egyptian objects. The Mycenaean glass relief beads on board the ship were also produced with Egyptian glass, although in this case more similar to glass from Amarna than to the Uluburun ingots. These results, coupled with our finding that glass almost identical to ingots found on the ship was used to produce several of the unprovenanced Mycenaean relief beads from museum collections, presents a picture of overall technological continuity combined with geographic flexibility at the end of the Amarna Period in Egypt.

1. Introduction

Late in the 14th century BCE (1320 ±15; Manning et al., 2009), a 15-meter sailing vessel sank along the southern coast of Turkey, just off the promontory known as Uluburun, or Grand Cape. Excavation by the Institute of Nautical Archaeology from 1984 to 1994 revealed a remarkable cargo of precious materials, primarily copper and tin ingots but also many finished and unfinished goods including an estimated 200 discoid glass ingots (Pulak, 2008). During the excavation, each complete or partial glass ingot was given a KW (Kaş Wreck) number if at least one-half of the ingot was preserved, and a Lot number if less than half of an ingot had survived. Where possible, an estimate of the ingot's original size and color was recorded in the excavation notebooks along with details of its locus, and its findspot plotted on the excavation site plan.

One-hundred and forty-six complete or nearly complete glass ingots were raised by the excavators and 28 were recovered in fragments, each representing one-half to slightly less than one-half of an ingot. At the time of excavation, it was clear that many other ingots were partially or completely lost due to severe degradation; in some cases, all that was left was an ingot-shaped void in marine encrustation. The precise number of ingots originally on the ship is impossible to determine with certainty although it had been estimated initially that, at a minimum, there would have been 175 glass ingots on board (Pulak, 1997; Pulak, 2001). Based on ongoing studies it now appears that this number is higher, perhaps around 200 glass ingots. With an average weight for well-preserved ingots of around 2.3 kg this would represent at least 460 kg of glass carried on board the ship. In addition, 30 Mycenaean glass relief beads of two different designs were recovered, consistent with the presence of

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two elite Mycenaeans aboard the ship (Pulak, 2005).

2. Archaeological questions

In this paper we examine the chemical compositions of the Uluburun glass ingots for insight into how they were produced, then compare these compositions to glass from known Late Bronze Age (LBA) glass production regions to determine whether the ingots were made in Egypt or Mesopotamia. In addition, we wondered how the Mycenaean relief beads found with the ship were related to the glass ingots and what their chemical compositions might tell us about glass exchange in the late 14th century BCE.

3. Materials

3.1. Glass ingots and Mycenaean glass relief beads from the Uluburun shipwreck

We analysed 201 different Uluburun glass ingot samples between 2 and 5 mm in size that were removed from complete or partial ingots on display or in storage at the Bodrum Museum of Underwater Archaeology in Bodrum, Turkey. Nine ingots were studied for color only because of severely weathered surfaces, leaving 192 samples for which we obtained excellent results for both major and trace elements (Table S1). Each sample was associated with a specific glass ingot KW number (130 samples) or glass ingot fragment Lot number (55 samples) with seven exceptions: two of these were impressions of nearly complete ingots with small amounts of glass preserved in the marine encrustation attached to one of the ship's stone anchors and five were ingot fragments for which the original excavation numbers could not be ascertained. Our 192 samples represent 114 glass ingots that were complete or more than

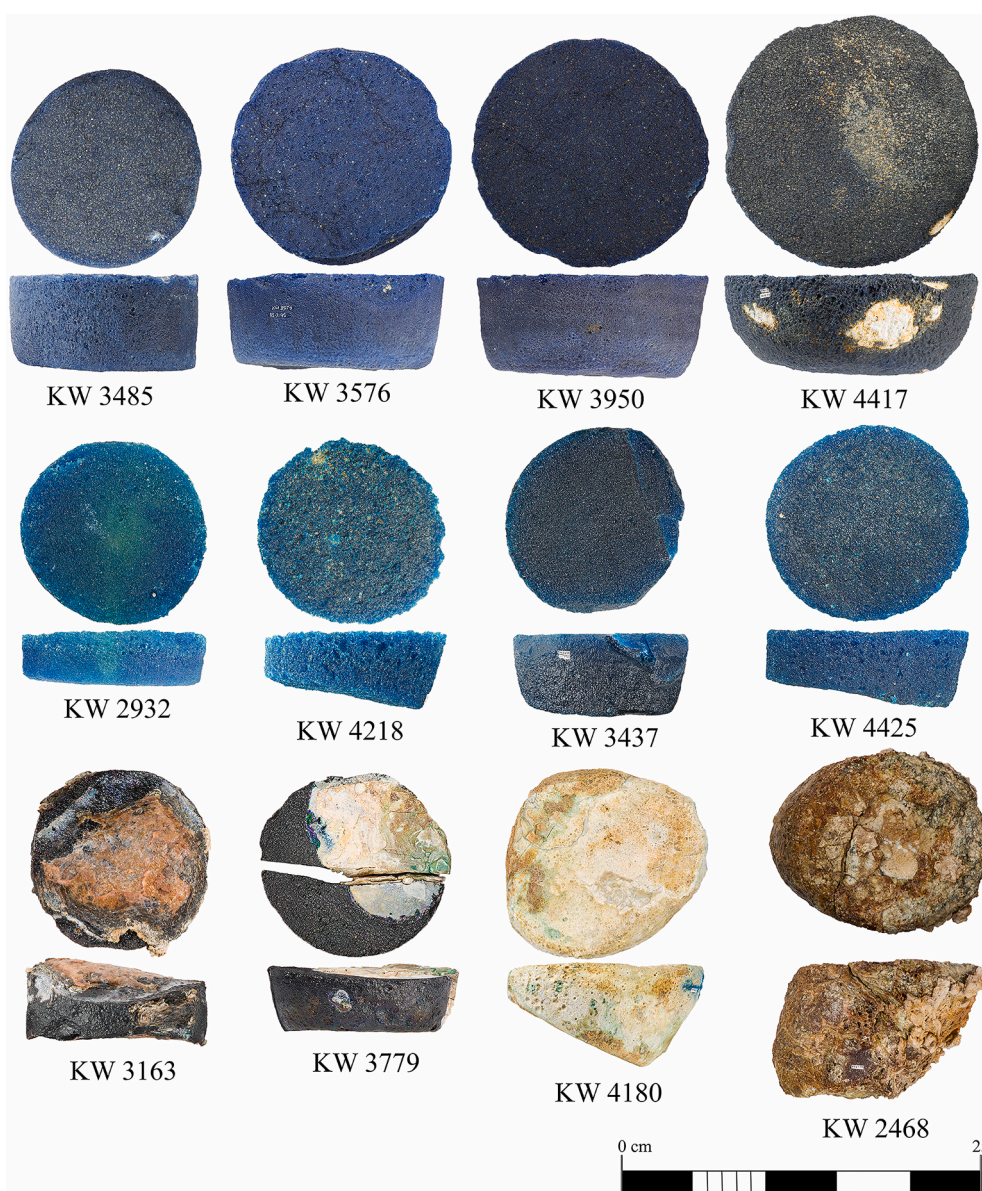


Fig. 1. KW 3485-KW 4417: cobalt-blue glass ingots; KW 2932-KW 4425: copper-blue glass ingots; KW 3163: purple glass ingot; KW 3779: amber glass ingot; KW 4180: copper-blue glass ingot cast in a tilted mold; KW 2468: cobalt-blue glass ingot cast in a deep irregular mold. Ingots are arranged to show the open surface on top and mold surface on bottom.

half-complete, 35 ingots that were one-quarter to one-half complete, and 24 with less than one-quarter ingot remaining. These counts, combined with the chemical compositional results discussed below, indicate good results for at least 142 different ingots. Additional ingots examined are the nine analyzed for color only, and nine other complete ingots that were not sampled because of their extremely weathered surfaces; attempting to do so would have severely damaged them. We also analyzed five of the Mycenaean glass relief beads from the shipwreck. Fig. 1 shows some of the representative Uluburun glass ingots examined, and Table S1 lists and describes the Uluburun samples used for this study. Some ingots were analysed more than once; these repeat analyses are included as mean values for the ingots shown in Table S1.

3.2. Other Late Bronze Age glasses

For comparative purposes, in addition to the 192 ingot and five relief bead samples from the Uluburun shipwreck, we present here 355 unpublished analyses of LBA glass from Egypt, the Levant, the Aegean world and Mesopotamia, including the first trace element analyses from the Elamite site Choga Zanbil. These new analyses represent a total of 222 different objects since many of the core-formed vessel fragments are polychrome and each color was analysed separately. We also include in the same format 18 previously published Amarna analyses (Varberg et al., 2015; Varberg et al., 2016) from the Orleans laboratory. A full list and description of the samples can be found in Table S2. Overall, 82 % of the new analyses are from Egypt, mainly Amarna (59 % of total samples), and 16 % from Mesopotamia, primarily Nuzi in Iraq or Tell Brak in Syria. The preponderance of Amarna material in this database reflects both the importance of Amarna as one of the few LBA glass primary production sites that have been identified (Smirniou and Rehren, 2011), and the dispersal of glass samples to western museums following the Amarna explorations of W.M. Flinders Petrie in the late-19th and early-20th centuries (Petrie, 1894). Photographs of representative vessel fragments and production debris are shown in Fig. 2.

4. Methods

4.1. La-ICP-MS

All chemical analyses were conducted by James Lankton and

Bernard Gratuze at the Institut de recherche sur les archéomatériaux (IRAMAT) in Orleans, France, using laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS). In this process, a laser beam, operating in the UV range, makes a cylindrical hole 30 to 100 μm in diameter and approximately 200 μm deep, in the glass surface. The minute quantities of glass released from the sample are then carried in an argon stream to a plasma torch (8,000 $^{\circ}\text{C}$), where the matter is atomized and ionized; these ions are focused at the entrance of a high-resolution magnetic sector mass spectrometer where they are separated according to their mass/charge ratios. The ionic beams (corresponding to pre-selected values of mass/charge ratios) are then collected by a channel electron multiplier or a faraday cup according to their intensities. Each sample was analyzed for 60 s, and the raw counts were converted to oxide weight percent (wt%) using the method of Gratuze (1999, 2016), based on reference glasses NIST 610, Corning B, C and D, and an in-house high-chlorine glass. During the study, we changed the sampling system from a VG Elemental Nd:YAG laser operating at 266 nm and using an argon gas flow to a Resonetics M50E excimer ArF laser operating at 193 nm and using an argon-helium gas flow. A Thermo Fisher Scientific Element XR mass spectrometer was used for almost all analyses, although the sampling time was reduced from 60 to 30 s for the last group of samples (see Van Strydonck et al., 2018 for further details). The only exceptions are 19 analyses from 14 of the 22 objects from Tell Brak made with a VG PQXS quadrupole mass spectrometer. LA-ICP-MS is the only current technique combining quantitative analysis of up to 60 different elements with minimally invasive sampling. A further advantage is that, unless the glass is severely weathered, the sample requires little or no preparation. Detection limits vary but are in the range of 1 ppm or below for most elements. Notable exceptions are some of the lighter elements, including sodium and potassium, with quantification limits in the 0.001 to 0.01 wt% (10 to 100 ppm) range because of high background counts. Trueness, commonly referred to as accuracy, was 4 % or below for major and minor elements and approximately 5 % or lower for most trace elements. Precision was in the same 4 to 5 % range for most of the elements but slightly higher for analyses from 2008. See Table S3 for repeat analyses of NIST 612 and Corning A performed on the same days as the LBA samples.



Fig. 2. Representative Late Bronze Age vessel fragments and production debris mounted in the LA-ICP-MS sampling cell. Most of these samples are from Amarna, along with copper-blue ingot fragments from Tell Brak. For sample identification see Table S2.

4.2. Strontium isotope analyses

Forty-nine samples representing at least 48 different glass ingots from the Uluburun shipwreck, as well as 11 Amarna samples, were selected for determination of $^{87}\text{Sr}/^{86}\text{Sr}$ strontium isotope ratios at the Isotope Geochemistry Laboratory at the University of North Carolina at Chapel Hill. The glass fragments were cleaned with distilled water, then approximately 10 mg of each sample was dissolved using ~ 4 mL of Teflon-distilled HF and 0.5 mL of quartz-distilled HNO_3 . Strontium was separated from solutions using Teflon columns containing about 60 μL of cleaned EiChrom Industries Sr-Spec resin. Each Sr sample was loaded onto a single rhenium filament. Isotopic ratios were measured using a VG (Micromass) Sector 54 thermal ionization mass spectrometer (TIMS) in quintuple-collector dynamic mode, using the internal ratio of $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ to correct for mass fractionation. The Sr carbonate standard SRM 987 was analyzed 99 times during the period in which the glass samples were analyzed. The average $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for SRM 987 was 0.710259 ± 0.000020 (2 standard deviations). Sample $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are reported relative to a value of 0.710250 for SRM 987 (i.e., if the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for the 5-to-9 standards analyzed with each group of samples averaged 0.710259, a value of 0.000009 was subtracted from the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for each of these samples). Internal precision for strontium runs is typically ± 0.000012 to 0.000018 % (2σ) standard error based on at least 120 dynamic data collection cycles.

4.3. Multivariate statistical analysis

We used PCA (Principal Components Analysis) and CA (Cluster Analysis) to determine the similarity between various glasses. In order to have interpretable results, two considerations are important. The first is the degree to which the analytical technique is accurate and precise at the compositional levels present in the glass. The second is whether some less-soluble elements are more likely to be unevenly distributed. Although repeat analyses (not shown) of samples from the same ingot suggested that the levels of Li, Cr and Zr tended to vary more, resulting in higher values for precision, we chose to include these elements in the statistical analysis because of their ability to distinguish Egyptian from Mesopotamian glass (Shortland et al., 2007). For the Uluburun ingots, including or omitting these three elements did not substantially affect the cluster assignments since all ingots have relatively low levels of Li and Cr and are relatively high in Zr. After several iterations, we decided that the following 35 oxides or elements, calculated as log-ratio values (Aitchison, 1999), were most useful: SiO_2 , Na_2O , K_2O , MgO , CaO , Al_2O_3 , Li, B, P, Ti, V, Cr, Mn, Fe, Rb, Sr, Y, Zr, Nb, Ba, La, Ce, Pr, Nd, Sm, Eu, Tb, Dy, Ho, Er, Tm, Yb, Lu, Th and U, but not including the colorants or colorant-associated elements Co, Cu, Ni or Zn. For the 19 Tell Brak VG PQXS quadrupole analyses (see above), the only REE values available were for La, Ce, Nd and Dy. In order to include these samples in the PCA and CA, we assigned temporary values for the missing elements based on those from similar samples analysed using the Thermo Fisher Scientific Element XR mass spectrometer. Because of this, the cluster assignments for these 19 Tell Brak analyses are less reliable.

Another critical variable for CA is the way the clusters are formed. We found the average linkage model, simultaneously analysing the Euclidean distances of all elements, to be most consistent, as opposed to emphasizing individual elements that are closest or farthest apart. All statistical comparisons were performed using Minitab version 18. The dataset used for PCA and CA included the 192 Uluburun glass ingot samples and 5 Mycenaean relief beads from the shipwreck (Table S1), plus the 373 analyses of LBA glass (Table S2) from our laboratory. We also included 35 analyses from different areas on the cross-section of an Uluburun ingot fragment (CMG 5975, KW unknown) and 133 analyses of reference glass NIST 612.

Cluster Analysis results in a percent similarity (or difference) between any two samples. By including the repeat analyses of the ingot section (CMG 5975) and NIST 612, we can estimate the percent

similarity expected for duplicate analyses of the same glass. For NIST 612, all 133 analyses were similar at 97 % and 111 analyses similar at 98 %; at 99 %, the analyses separated into ten major groups with several outliers. Most of this variability was due to the high background counts for Fe, with a mean value (48 ppm) close to detection limits for our LA-ICP-MS system. The 35 repeat analyses of the ingot section were less consistent, with a similarity of 95 % only after we discarded three analyses with very high Zr (200 ppm or greater). Removing five other measurements with the most apparent variation due to heterogeneity left 27 analyses (77 % of the original 35) with a similarity of 97 %, equal to the similarity of our NIST 612 analyses. The precision for these 27 analyses was excellent, with RD (relative deviation calculated as standard deviation divided by mean value) below 5 % for all major and most minor and trace elements, with the exceptions of Li (9.4 %), Cr (19 %), Zr (13 %) and a few other elements present at low levels in the glass. Thus, both the CA results and the RD values for groups of samples similar at the 97 % similarity level pointed to the same conclusion: with the CA settings mentioned above and the included samples, two or more glasses similar at the 97 % level were consistent with being from the same parent object or, as discussed below, from the same original glass batch. As we had to remove eight of the 35 measurements from the ingot section to reach 97 % similarity, we recognize that some samples similar at lower levels might also fit these groups; however, we felt it more important to demonstrate that compositional groups exist than to specify exactly how many ingots were in each group. It should be noted that percent similarity and the RD values that they imply relate only to the specific circumstances of this study and are not suggested as absolute criteria; values vary depending on the clustering method and the elements and samples included, and the conditions most useful for each archaeological context must be determined.

5. Results and discussion

5.1. Overview

In addition to sample descriptions, Tables S1 and S2 show the LA-ICP-MS results for 54 oxides and elements expressed as weight percent (wt%) for major and some minor oxides and as parts per million (ppm) for all other elements, most of which are present at levels well below 1.0 wt%. We have also listed 'reduced compositions' (Brill, 1999, Vol. 2), normalized to the sum of the six major oxides plus Fe_2O_3 , for SiO_2 , Na_2O , K_2O , MgO , CaO and Al_2O_3 and indicated by an asterisk after the chemical signature. Although these values vary little from the measured values, since Cu, Sb and Pb are low for all ingots, the reduced compositions are included here to facilitate comparison of the Uluburun samples with other glasses from Egypt or Mesopotamia, some of which do contain significant Sb and Pb. In addition, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are given for the 49 samples that we studied, along with strontium isotope results from the same laboratory reported by Brill and Stapleton (2012).

All Uluburun ingots were made from soda-lime-silica glass with soda (Na_2O) between 15.3 and 20.6 wt%, with a mean value 17.9 wt%, with the exception of one slightly weathered sample (KW 3276) at 14.3 wt%. Magnesia (MgO) is generally > 2 wt%, indicating plant-ash as the primary flux, although 7 % of the samples had MgO between 1.4 and 1.8 wt%. Potash (K_2O) levels are low, below 2.0 wt% for all cobalt-colored and most copper-colored ingots, and lime (CaO) varies from 4.3 to 10.7 wt%. Using PCA and CA based on SiO_2 , Na_2O , K_2O , MgO , CaO and B, P, Rb and Sr, the elements most likely to reflect the plant-ash source, as well as alumina (Al_2O_3), reflecting both the cobalt colorant and a lithophile contribution possibly from grinding stones used during various stages to prepare raw and semi-finished material for melting (Rehren, 2008), we divided the samples by chemical composition using PCA and CA. This resulted in 11 groups, each with a similarity index of 87 %.

5.2. Compositional groups based on plant-ash elements and alumina

5.2.1. Cobalt-colored ingots

Cobalt blue (Co-blue), whether containing cobalt alone or including small amounts of copper, was by far the most common color, comprising 138 (72 %) of the 192 ingot samples. Based on plant-ash elements and alumina, we identified five major compositional groups, shown graphically in Fig. 3. The number of samples in each group, color, minimum number of ingots represented, and mean chemical compositions with relative deviations are listed in Table 1. When trace elements were included these major groups formed 20 sub-groups, each matching at the 97 % level by cluster analysis, as shown in Table S4.

Group Co-1 is very low in potash, mostly below 0.6 wt%, as well as in magnesia, lime and phosphorus, but high in Cl and B. Although neither Co nor Cu were included in the PCA and CA used to define the groups, all samples in this group would fit into the category ‘CoCu-coloured’ as described by Smirniou and Rehren (2013), with the lowest Cu level of 785 ppm, slightly below the 850-ppm cutoff suggested. Group Co-2 is slightly higher in potash, magnesia, lime and phosphorus, and higher in alumina. The levels of cobalt are similar to those for Group Co-1; however, the Cu is lower, <500 ppm for almost all samples. Group Co-3 is higher in lime, magnesia and potash than most samples in Groups Co-1 and Co-2; in addition, all Co-3 samples are very low in Cu, generally below 200 ppm. Group Co-4 is less consistent but most of the samples are relatively high in potash, up to 1.6 wt%, and alumina and moderate in lime. Although just over half of the samples would be characterized as CoCu blue, both Cu and Co are variable, with the variability in cobalt reflected in the wide range of alumina values for group Co-4 evident in Fig. 3. Group Co-5 is much smaller and has the lowest potash, magnesia and lime values but high alumina and Co. In this group, Cu is very low, around 100 ppm.

It is striking that Groups Co-3 and Co-4 include a higher percentage of complete ingots. These two groups are also the highest in lime, with moderate levels of alumina. Although both elements may be associated with improved resistance to weathering, it appears that lime is the more important factor as the ingots in Group Co-5, the highest in alumina but low in lime, were not as well preserved.

5.2.2. Copper-colored, purple and amber ingots

The 50 samples colored greenish-blue by added copper can be divided into four groups, shown in Figure 3 and Table 1, based on plant-ash elements and alumina. With the inclusion of trace elements, these major groups separate into seven sub-groups, each at 97% similarity by CA, shown in Table S4. Major group Cu-1 is high in soda but low in potash, with values all below 1.0 wt%. The levels of magnesia, lime, alumina and phosphorus are also relatively low, while Cl and B are high. Copper (Cu) is around 5000 ppm for most samples, between 0.6 and 0.7 wt% CuO. Group Cu-2 is higher in potash, magnesia and, in particular, lime. Copper is slightly higher than for Group Cu-1, although still below CuO 1.0 wt%. KW 3156, an ingot transparent pale green streaked with pinkish-red, is an excellent example of the potential difficulty to achieve a strong copper-blue colour. The chemical composition and copper content is essentially identical to other members of Group Cu-2 but, most likely because of insufficient oxygen in the furnace, the copper remained reduced to its metallic state and, based on microscopic observation of a very thin chip from the ingot, appears only as streaks of sub-micron-sized particles of Cu in an otherwise colorless glass. This colorless glass when re-heated in an open furnace would most likely turn greenish-blue as the copper dissolves as CuO. Group Cu-3 is higher in potash, approximately 1.5 wt%, but very low in magnesia, below 2 wt%, and relatively low in lime. The samples in this group also have low Cu with CuO below 0.5 wt%. The seven samples in Group Cu-4, all from complete, well-preserved ingots, are much more similar in major-element composition to what is considered to be typical LBA glass (Rehren, 2008), relatively high in potash, between 1.90 and 1.98 wt%, magnesia and, not surprisingly, lime, about 10 wt%. Alumina is low, as is characteristic for all non-cobalt coloured glasses. The general correspondence between the degree of preservation and lime levels is just as apparent in the copper-colored ingots as in those colored by cobalt, supporting the idea that lime, not alumina, is the major factor in ingot preservation.

Only four glass ingots were not colored by cobalt or copper. Three purple ingots—two complete or nearly complete and one fragmentary and heavily weathered—have major elements similar to the typical LBA composition referenced above; both potash and magnesia are above 2 wt %, alumina is low and lime is high at 10 wt%. Manganese was used as a colorant, with Mn 3380 to 6000 ppm, corresponding to MnO 0.4 to 0.8

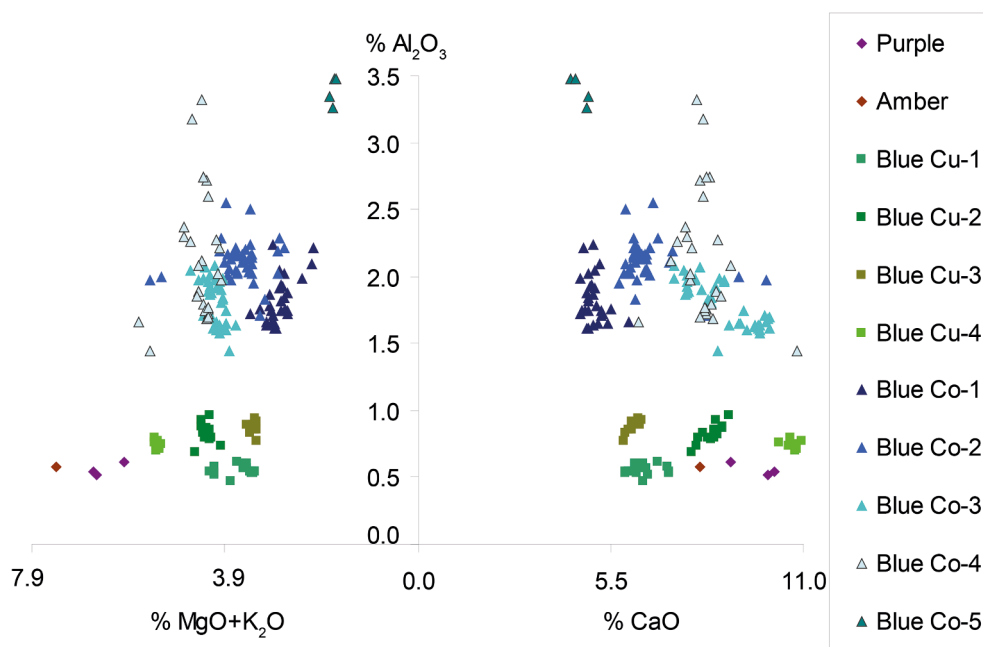


Fig. 3. Major compositional groups identified among Uluburun ingots (Co 1–5, Cu 1–4, purple and amber) based on plant-ash elements (MgO, K₂O), lime and alumina. Subgroups given in Tables S1 and S4 (A, B, C ...) are not represented.

Table 1
Mean compositional values and relative deviations (RD) for 11 Uluburun glass ingot groups based on plant-ash associated elements. Color, number of samples, and minimum number of ingots indicated for each group. The high RD values for some elements reflects relative heterogeneity within these large groups. For sub-groups corresponding to individual batches see [Table S4](#). RD calculated as standard deviation divided by mean value. Oxides and Cl in weight percent, elements in parts per million, bd below detection.

Group	Co-1		Co-2		Co-3		Co-4		Co-5		Cu-1		Cu-2		Cu-3		Cu-4		Purple		Amber
	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	mean of	min #	
	29	20	40	25	39	37	26	22	4	3	17	8	16	11	10	5	7	7	3		1
color	blue	RD	blue Co/	RD	blue Co	RD	blue Co/	RD	blue Co	RD	blue Cu	RD	blue Cu	RD	blue Cu	RD	blue Cu	RD	purple	RD	amber
	CoCu	(%)	blue CoCu	(%)		(%)	blue CoCu	(%)		(%)		(%)	blue CoCu	(%)		(%)		(%)		(%)	
SiO ₂	69.4	1	67.6	2	65.5	2	66.0	2	67.0	0	68.2	1	67.0	1	69.6	1	64.7	0	63.5	2	66.3
Na ₂ O	18.6	5	18.1	4	17.9	4	17.1	5	20.4	1	18.8	4	17.0	5	17.4	2	16.0	1	17.9	4	16.1
K ₂ O	0.53	15	0.79	10	1.11	19	1.28	19	0.31	15	0.78	8	1.32	7	1.35	29	1.94	1	2.37	10	2.82
MgO	2.22	10	2.76	17	2.93	6	3.08	15	1.38	2	2.76	10	2.87	3	1.90	22	3.21	1	3.89	15	4.44
CaO	4.91	6	6.43	13	8.59	10	7.98	9	4.54	5	6.29	7	8.20	3	6.01	3	10.40	2	9.53	7	7.93
Al ₂ O ₃	1.79	9	2.08	7	1.77	10	2.11	23	3.32	3	0.54	6	0.81	8	0.86	6	0.72	4	0.55	9	0.57
P ₂ O ₅	0.06	11	0.08	11	0.10	18	0.13	14	0.03	4	0.07	6	0.12	6	0.11	22	0.13	2	0.18	17	0.28
Cl	1.31	7	1.14	11	1.03	7	1.01	9	1.32	3	1.38	4	1.10	7	1.22	4	1.06	2	0.85	14	1.04
Fe ₂ O ₃	0.46	10	0.45	16	0.55	16	0.61	15	1.02	4	0.30	11	0.46	5	0.46	5	0.41	2	0.45	2	0.33
B	247	9	195	7	148	13	125	26	198	3	198	12	125	6	150	10	70	4	70	14	82
Ti	578	3	443	14	509	18	605	8	720	3	395	20	511	4	537	4	561	3	378	15	319
Mn	881	7	839	10	945	18	1310	35	1773	4	98	5	130	3	152	19	137	1	4265	36	249
Co	468	9	445	6	423	19	521	39	819	2	6	18	3	40	2	16	2	4	4	68	3
Ni	355	11	323	8	276	18	375	53	434	2	8	8	7	14	6	26	8	7	5	33	4
Cu	994	16	256	69	102	62	993	87	96	25	4890	16	5876	22	5813	64	8636	5	37	39	18
Zn	695	11	586	10	512	22	678	35	678	7	17	40	20	9	19	21	17	3	26	34	14
Rb	2.9	16	4	16	5.7	12	7	21	3.4	30	3.0	7	6.0	5	7.5	24	8.1	3	12.8	3	9.6
Sr	487	13	666	27	876	14	750	15	210	11	719	11	882	8	540	10	934	2	875	2	669
Y	13.2	9	13.1	17	6.5	28	14.0	26	17.6	3	2.3	8	2.8	5	3.1	5	2.9	4	4.4	22	1.9
Sb	1	31	4	23	10	212	142	111	8	13	1	20	2	64	1	63	51	4	19	168	bd

wt%. Two of the purple ingots are similar at the 97 % level and are included as group P-1 in Table S2. The one amber ingot, complete but in two pieces and relatively non-weathered, is high in potash and magnesia and moderate in lime, with low levels of Co and Cu, 2.8 and 18 ppm, respectively.

5.3. Defining compositional groups and production events

The most striking result of the PCA and CA described above was not the overall similarity between the samples. It was the fact that within the major-element groups, there were subgroups of samples that were so similar to one another that they seem to have been made from identical glass. Early in our study, we had wondered whether we could determine if two partial ingots were originally from the same parent ingot, thus attempting to make a ‘chemical join’ where a physical join was not possible because of extensive surface degradation. While we had minimal success because of incompatible fragment sizes and shapes, it became clear that something much more fundamental was at play. As described above, we knew that under certain circumstances and with relatively few exceptions, repeat analyses of the same glass should have a 97 % similarity index by CA. We looked for ingot samples that fit these criteria using the full suite of 35 elements previously listed and found that over 90 % of the samples were a match to at least one other sample and, in one case, to almost 30 other samples. The chemical analyses had uncovered ingots that could not be distinguished from one another using LA-ICP-MS. We interpret this to mean that the ingots were produced in groups or ‘batches’ using the same glass and under very similar furnace conditions. In Table S1 these sub-groups or batches at 97 % similarity are indicated as Co-1 A to E, Co-2 A to D, etc., with other possible members of each sub-group indicated in parentheses. As mentioned above, the minimum number of ingots, mean compositions, and relative deviations for these sub-groups are given in Table S4.

At a similarity index of 97 %, there would have been no more than 20 production events for 124 of the 138 cobalt-blue samples and seven production events for 39 of the 50 copper-blue samples. Although most of the remaining cobalt- or copper-colored samples were matches at a lower similarity index, we could not be certain whether these samples represented heterogeneous parts of the same batches or slightly different production circumstances. Two of the three purple ingots matched at the 97 % level, with the third only at 90 %. The one amber ingot was notably different from all the other samples, even though specific coloring agents had not been included in the statistical analysis.

For the samples similar at the 97 % level, which corresponds to our ‘batches’, the variation in chemical composition matched that of repeat analyses of the ingot section (CMG 5975); relative deviation (RD) for most major and minor elements was between 2 and 7 %, with higher deviations for Li, Cr and Zr. Relative deviation for REEs in cobalt-colored glass was usually 5 % or lower while slightly higher for copper-colored glass because of REE levels closer to the background counts for our Thermo Fisher Scientific Element XR mass spectrometer. Both Sb and Pb are very low in the Uluburun glasses, with higher RD values, although in the one group of cobalt-blue ingots with Sb 250 ppm the RD was 12 %.

Although it may seem counterproductive to use the word batch in the general sense of a group of objects produced at one time when ‘glass batch’ has a specific meaning in glass technology to indicate the various raw materials to be smelted together, we feel that ‘batch’ better captures the workshop conditions in which the ingots were manufactured than alternative terms like ‘production event’. Here we follow the lead of Freestone et al. (2009) in their description of vessels blown from a single batch of glass.

5.4. Sr isotope results

The Uluburun strontium isotope ratios vary from 0.70781 to 0.70799. This is a relatively restrained range compared to published values from Amarna and Malkata (Degryse et al., 2010; Henderson et al.,

2010; Degryse et al., 2015), but large enough to make us wonder how the isotope values align with the defined chemical groups. Therefore, we examined the values for samples between 93 and 97 % similarity index; here, plus or minus 2σ is the equivalent of plus or minus one digit at the 5th decimal so that the values 0.70784 and 0.70786 could be considered as equal, while 0.70784 and 0.70787 would be off by 0.00001. For the batches defined at 97 %, only one of 52 strontium isotope values (including the results from Brill and Stapleton (2012)) was off by 0.00001, and none were off by more than one digit at the 5th decimal. The similarity between the strontium isotope ratios decreased with percent similarity, with a possible inflection point at 95 % similarity by CA, with four samples falling out of range, two by one digit and two by four digits. On the other hand, groups quite different chemically, such as Cu-4, Cu-2 A and B, Co-3C and Co-4, may have the same strontium isotope ratios, suggesting that the same raw materials were processed in different ways or that raw materials from different geographic locations might share the same geological background. Without more detailed knowledge of plant-ash isotope ratio characterization in Egypt, it is difficult to further interpret these results in terms of geographic origin. In any case, the similarity of strontium-isotope values is useful as an independent typology that provides strong confirmation for our group identities based on chemical composition alone.

5.5. Steps in glass ingot manufacture

How did the glassmakers achieve the high degree of uniformity found in the production groups that were revealed through chemical analysis? Late Bronze Age glass ingot production began with sourcing, selecting and preparing the plant ash and quartz pebbles that provide the fluxes and silica. These ingredients were first ground into a flour-like consistency using grinding stones similar to those identified by Rehren (2016). Afterward, they were heated together to form glass ‘frit’, a foamy, partially-fused glass, examples of which have been found at both Amarna and Qantir (Smirniou and Rehren, 2011; Smirniou et al., 2018). This frit was then re-ground, perhaps with plant ash or silica added as needed by the workers, placed in ceramic crucibles and put in a glass-making furnace capable of reaching temperatures close to 1000° C or higher. It is unlikely that the resulting glass was uniformly ‘smelted’, and physical separation may have been required to obtain fully fused glass. This imperfect glass was probably ground and remixed, then re-melted in crucibles to produce homogeneous glass. At some stage, before or after this step, colorants were added to the mixture, followed by further grinding and mixing before final heating to form colored glass ingots. The largest batch included at least 16 ingots totaling 40 kg of finished glass. This would mean that the glassworkers would have processed well over this weight in ingredients or intermediate materials, particularly during the early grinding and mixing stages. Based on the relatively small grinding stones found at Qantir, it would not have been possible to grind this much material uniformly. Therefore, thorough mixing would have been required to achieve the observed compositional uniformity for not only the major glass ingredients but also the lithophile trace elements from the grinding stones. The copper and cobalt colorants were also identical across the ingots in a certain batch, and this could result only from thorough mixing, either of already colored glass or of glass powder with ground colorants prior to the final melting stage.

For LBA glasses, the chemical compositions reflect not only the original ingredients but also furnace conditions. Major oxides like Na₂O, K₂O, MgO and CaO were likely controlled through such mechanisms as partial batch melting, temperature control of alkali earth oxides, and possible salt melts (Rehren, 2000; Shugar and Rehren, 2001; Tanimoto and Rehren, 2008). Therefore, furnace conditions must have been very similar for all ingots in a single batch, particularly during the early ‘smelting’ stages when the chemical composition of the glass was determined.

Although we think of the Uluburun ingots as ‘raw glass’, they were, in fact, a high quality product manufactured to an exacting standard. It

is not known whether LBA glass furnaces were large enough to hold as many as sixteen crucibles. If they were not, the glassworkers replicated conditions for sequential firings with great precision, followed by thorough mixing to even out any small differences in chemical composition. The ingots resulted from several manufacturing stages, each requiring technological choices embedded in social and political contexts. This mode of production would have been both well established and well organized to provide unworked glass that could be stored as wealth or distributed to elite secondary production centers manufacturing beads, vessels and decorative components. Workshop practices may have been driven by tradition, efficiency or customer requirements. However, regardless of the reason, the marked chemical compositional uniformity of Uluburun ingots within production groups strongly contrasts with the variability of glass from later Byzantine and early Islamic period primary tank furnaces (Brems et al., 2018).

5.6. Egypt or Mesopotamia?

5.6.1. Cluster analysis

The complete Cluster Analysis (CA) may be reconstructed for Table S2 alone or for the entire dataset by lining up the columns for clustering results in Tables S1 and S2. While the large groups at 85 % similarity contain samples from both Egyptian and Mesopotamian sites, at 90 % similarity almost all the overlap is resolved, suggesting that for most samples 90 % similarity in this particular cluster analysis was adequate to differentiate Egyptian from Mesopotamian glass. In particular, this is true for all Uluburun samples, grouped only with other Egyptian glasses at the 90 % level. The only exceptions are the ten samples from Egyptian and Mesopotamian contexts in cluster 16 at the 90 % level. Based on major and many trace elements these samples are quite similar. However, at a cluster level of 93 % they separate into four groups of one to three samples each. Two of the groups, working debris from Amarna or decoration on Egyptian vessel fragments, are consistent with an Egyptian composition, although Cr levels are high for two of the samples, raising the possibility that there could be some mixing with glass with a more Mesopotamian composition (see below for further discussion). The other five samples in cluster 16 at 90 % are from Amarna, Tell Brak, and Alalakh. Based on CA alone it is difficult to say whether these samples are Egyptian or Mesopotamian, although when trace element ratios are considered four of the samples are clearly Mesopotamian and the fifth, an opaque green rod from Amarna, sample 217 in Table S2, most likely Mesopotamian glass with some Egyptian admixture. We have indicated geographic origin based on CA in Table S2, Column K, as E (Egyptian, 304 samples) or M (Mesopotamian, 59 samples), with additional categories 'E mix' (8 samples) and 'M mix' (2 samples) for glasses E or M by CA but with inconsistent trace element values. Origin is not included in Table S1 since all of the samples are clearly Egyptian. We would like to emphasize that while CA worked well in this study, uniform data with high accuracy and precision over multiple elements are necessary for consistent and interpretable results. This is unlikely to occur when combining data from multiple studies, and we wondered if it might be possible to achieve the same separation using fewer elements.

5.6.2. Trace element ratios

Shortland et al. (2007) demonstrated that trace element ratios are useful for distinguishing Mesopotamian from Egyptian glass. This was a watershed moment, not only for addressing questions of origin but also for showing how valuable trace element data can be for understanding glass production. Shortland et al. used a plot of the ratios $1000\text{Zr}/\text{Ti}$ by Cr/La to differentiate their group of 54 samples from Amarna and Malkata in Egypt, Tell Brak in northern Syria and Nuzi in northern Iraq into two groups that corresponded to geographic origin. However, based on their Fig. 6, there is one Tell Brak sample close to the Egyptian group and another hiding behind two Amarna samples. The possible ambiguities, both because of low Cr values in Tell Brak samples and

considerable overlap on the $1000\text{Zr}/\text{Ti}$ axis, raise the question of whether there might be a more precise way to differentiate the two groups of glass. The dataset compiled and presented here has two advantages to the one used by Shortland et al.. First, we were able to work with a greater number and broader range of samples. Second, we had a good idea of what to expect since CA on the same samples, discussed above, showed a clear separation between Egyptian and Mesopotamian origin for all but a few samples.

To answer the specific question of whether a given glass sample was consistent with an Egyptian or Mesopotamian origin, we found that four elements were most useful for a shorthand method. Three of these, Ti, Cr and Zr, were used by Shortland et al. and the fourth is Li. Although Shortland et al. had not found a regular variation in Li, based on our larger dataset it appears that Li, like Cr, is generally higher in Mesopotamian glass, while both Zr and Ti are higher in Egyptian glass. Of these, Ti is probably the most reliable in terms of LA-ICP-MS measurement.

In our samples, as mentioned above, Cr, Li, and Zr have more variability than other elements. For Cr, background counts are high since the argon-carbide complex sometimes formed during atomization and ionization has the same mass/charge ratio as Cr. For this reason, higher values of Cr are likely to be more precise. For Li, an important factor may be the high volatility of this element. However, although its final concentration in the glass depends strongly on the melting temperature and the duration of the heating, the Li contents measured for Egyptian glasses are statistically systematically lower than those measured for Mesopotamian glasses. This difference could be due either to a higher concentration of lithium in the raw materials used by Mesopotamian glassmakers or to different working conditions such as furnace geometry, working temperature and the heating time used by these glassmakers. Zr variability is also a problem, particularly in Egyptian glass containing residual zircon grains that will produce anomalously high Zr counts as the laser hits or passes nearby, not reflecting the average content in the glass. However, since most Egyptian glass is already high in Zr, an artificially elevated value would simply make the sample appear 'more Egyptian'; we did not encounter zircon grains in any of the samples from Mesopotamia. In any case, we determined that it would be more useful to have two elements high in Mesopotamian glass and two high in Egyptian glass and calculated the Li/Zr and Ti/Cr ratios, one designed to be low and the other high for Egyptian glass, and vice versa for Mesopotamian glass. For ease of comparison, these calculated ratios are included in Tables S1 and S2.

We compared the results from both sets of ratios to those from CA. While the graphic display is similar for both ratio sets, the overlap zone appears larger on the $1000\text{Zr}/\text{Ti}$ by Cr/La plot but still present for Li/Zr by Ti/Cr , as shown in Fig. 4. While all of the Uluburun ingots were clearly in the Egyptian zone, some of our Egyptian samples, mostly from Amarna and representing workshop debris or parts of finished objects, appear to be half-way between the Egyptian and Mesopotamian groups.

There are several reasons, including possible measurement error for one of the four elements considered, that element ratios may misrepresent an object; we suspect that the most common reason may be the actual mixing of Egyptian and Mesopotamian glass, such as could have occurred in active glass workshops like those at Amarna.

For a more detailed analysis, we divided the samples sequentially into groups based on ratio values, at each stage comparing these results to those from CA. In this way, we were able to 'correctly' identify 96 % of the samples using $1000\text{Zr}/\text{Ti}$ by Cr/La and 99.5 % of the samples using Li/Zr and Ti/Cr . The steps for $1000\text{Zr}/\text{Ti}$ by Cr/La were as follows:

1. $1000\text{Zr}/\text{Ti} \geq 27.0$ and $\text{Cr}/\text{La} < 4.0$: 481 samples, 98.3 % Egyptian but including eight lower-Cr Nuzi and Tell Brak samples that were Mesopotamian by cluster analysis.
2. $1000\text{Zr}/\text{Ti} > 50.0$ and $\text{Cr}/\text{La} > 4.0$ and < 6.0 : 34 samples, 82 % Egyptian, with six Tell Brak samples that should be Mesopotamian.

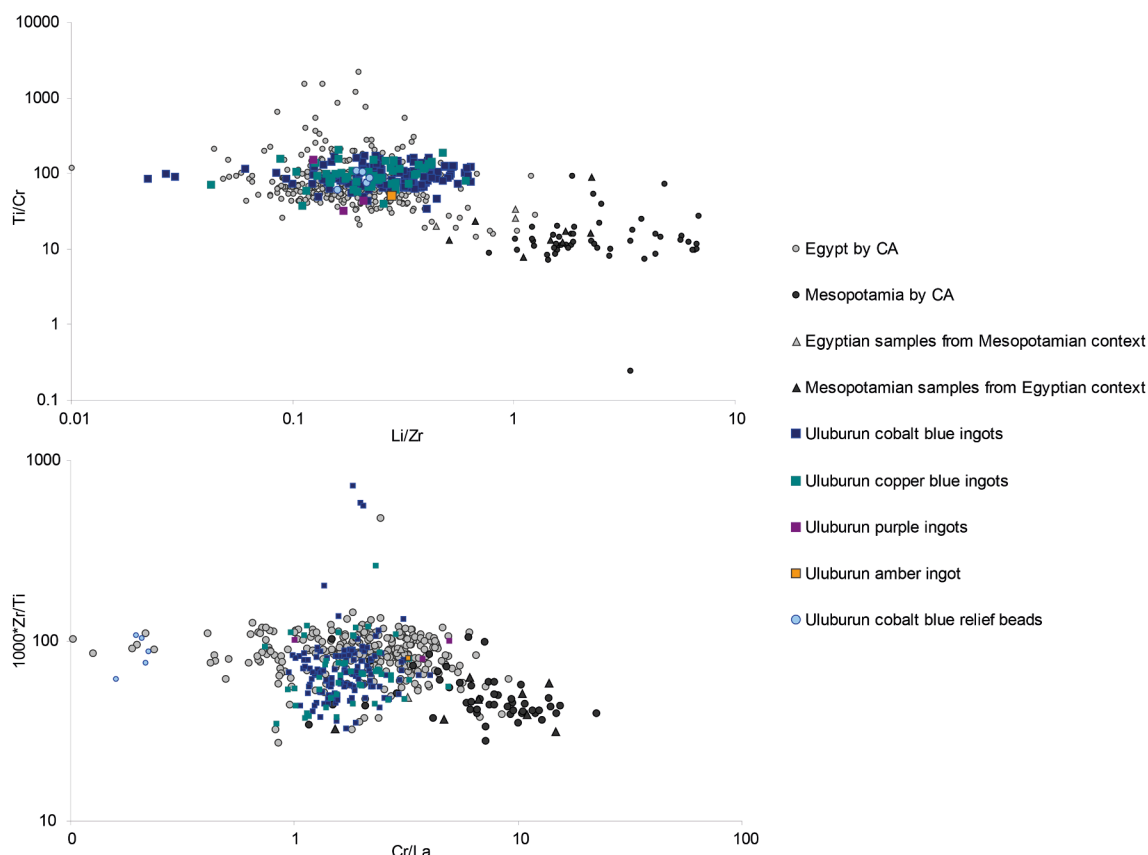


Fig. 4. Egyptian or Mesopotamian origin of glass samples based on PCA/CA classification (Table S2) and element ratios (Ti/Cr and Li/Zr on top, 1000*Zr/Ti and Cr/La below). According to their Ti/Cr, Li/Zr, 1000Zr/Ti and Cr/La ratios, all the Uluburun glass ingots and cobalt-blue glass relief beads have an Egyptian origin. While both plots include samples, mostly Egyptian by CA, that are between the Egyptian and Mesopotamian zones, the overall separation between Egyptian and Mesopotamian glasses appears greater when plotting Ti/Cr by Li/Zr.

- 1000Zr/Ti < 50.0 and Cr/La > 4.0 and < 6.0: three samples, all Mesopotamian.
- 1000Zr/Ti ≤ 60.5 and Cr/La ≥ 6.0: 50 samples, 84 % Mesopotamian and the others either Egyptian or mixed from Amarna and Alalakh.

Comparing the results with those from CA, this stepwise method using the 1000Zr/Ti and Cr/La ratios provided assignments that matched the CA results for all but 22 samples from Egypt and Mesopotamia.

For Li/Zr by Ti/Cr:

- Li/Zr < 1.00 and Ti/Cr not specified (although 98 % of the samples were > 20): 505 samples, 99.6 % Egyptian, with two exceptions of possibly mixed glasses from Amarna and Choga Zanbil.
- Li/Zr ≥ 1.00 and < 1.52, with Ti/Cr > 15.0: 7 samples, six Egyptian samples and one low-Cr white glass from Choga Zanbil that should be Mesopotamian.
- Li/Zr ≥ 1.00 and < 1.52 with Ti/Cr < 15.0: nine samples, all Mesopotamian.
- Li/Zr ≥ 1.52 with Ti/Cr variable (but with 80 % (20): 47 samples, all Mesopotamian.

Egyptian glasses typically had Li/Zr ratios < 1.00 and Ti/Cr ratios almost always > 20. For the relatively few Egyptian glasses with Li/Zr between 1.00 and 1.52, the Ti/Cr ratios were helpful since Ti/Cr is mostly higher in Egyptian glass. All samples with Li/Zr ≥ 1.52 were Mesopotamian regardless of the Ti/Cr ratio. This second method worked well for the Uluburun samples while the 1000Zr/Ti and Cr/La ratios were ambiguous for ingot groups Cu-2 A and Co-1B because of relatively

low Zr. Our conclusion is that while the 1000 Zr/Ti and Cr/La ratios are effective for most samples, if Li values are available, it is worthwhile to try the Li/Zr and Ti/Cr ratios as well, then to look carefully at the samples differently identified by the two techniques.

In summary, whether by CA or by Li/Zr and Ti/Cr ratios, all the Uluburun ingots are unquestionably Egyptian, consistent with an Egyptian identification for the three samples studied by Jackson and Nicholson (2010). While this result is important, it does not answer the question of where in Egypt the ingots were made, which will be the subject of subsequent study.

5.6.3. Sr isotope ratios

As a result of overlap between Egyptian and Mesopotamian samples, strontium isotopes alone cannot be used to distinguish between the two. A plot of Sr by $^{87}\text{Sr}/^{86}\text{Sr}$ (not shown) is not much better since there is considerable overlap for Sr as well. In general, Sr isotope ratios are lower in Egyptian than in Mesopotamian glass; 80 % of our 81 Egyptian analyses were ≤ 0.70794 while the lowest of 11 Mesopotamian values was 0.70798, a difference > 2σ. Only 4 % of the Egyptian samples but 70 % of Mesopotamian glasses were above 0.70820, another index that might be useful when combined with trace element ratios. There was no magic number, however, to separate glasses from the two regions based on Sr isotopes alone. Even so, isotope ratios for Sr and Nd have been useful for distinguishing Egyptian from Mesopotamian glass (Degryse et al., 2010). Egyptian glass is usually low in both $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{144}\text{Nd}/^{143}\text{Nd}$, while the Mesopotamian glasses, at least those studied from Tell Brak and Nuzi, are generally higher in Sr ratios and high (Tell Brak) or low (Nuzi) in Nd ratios.

5.7. Evidence for glass exchange between Egypt and Mesopotamia

As indicated on Fig. 4 there are six glasses, samples 69, 92, 178, 197, 224, and 242 in Table S2, with Mesopotamian compositions but found in Egyptian contexts. In addition, two other Amarna samples, numbers 88 and 217 in Table S2, seem to be Mesopotamian by CA but have inconsistent trace element ratios. Seven of these samples are working debris from Amarna, including a half-round strip (CMG AMN BC 02), while the eighth is the copper-blue body of an unprovenanced Egyptian-style vessel for which the white, yellow and cobalt-blue decorations are Egyptian. In particular, CMG AMN BC 02 is similar at 93 % to two vessels from Nuzi and at 90 % to a copper-blue ingot from Tell Brak.

In addition, there are three glasses that are Egyptian by CA and element ratios but found in Mesopotamian contexts. These include sample 355, HH 224 2B deep, dated ca. 1275 BCE, a possible fragment from a cobalt-blue ingot excavated at Tell Brak, Syria. The glass is colored by Egyptian cobalt with high Ni, Zn, Mn, Y and REEs, supporting an Egyptian origin. Sample 340, CMG 721b Cu, a copper-blue tile from Tell al-Rimah, Iraq, from a context dated to 1400 BCE, is also Egyptian glass. CMG 721b is similar at 94 % to a red glass cane from Amarna, raising a question about its proposed dating. The third example of Egyptian glass in a Mesopotamian context is sample 342, the amber glass from CMG 406, a piriform vessel in Mesopotamian style. While the Tell Brak ingot fragment plots clearly in the Egyptian zone, the other two Egyptian glasses may be at least partially mixed since they are on the borderline between the Egyptian and Mesopotamian samples. The cobalt-blue Tell Brak 'ingot' fragment was not very similar to any of the Egyptian glasses in the dataset, so it is difficult to interpret in more detail, but to our knowledge these Tell Brak and Tell al-Rimah samples provide the first clear demonstration of Egyptian glass from Mesopotamian archaeological contexts. While without archaeological context, the Mesopotamian vessel CMG 406 provides further support for the use of Egyptian glass in Mesopotamia.

How significant was glass exchange between Mesopotamian and Egypt? While Egyptian requests for glass are known from the Amarna letters, and temple and tomb paintings show gifts of glass ingots that may have come from abroad (see Shortland 2012 for discussion), actual Mesopotamian glass has been less evident, limited until now to the two Amarna glass rods with Mesopotamian compositions reported by Varberg et al. (2016). The new samples reported here, combined with 18 glasses previously studied in our laboratory by Varberg et al. (2015), Varberg et al. (2016), provide an opportunity to reconsider the impact of the glass being exchanged. Focusing on Amarna, the source of 70 % of our Egyptian glasses, seven samples, discussed above, are Mesopotamian by CA (M or M mix). In addition, there are eight other Amarna samples, numbers 93, 124–125, 141–144, and 178 in Table S2, that are Egyptian by CA but with atypical trace element ratios (E mix) because of their high values for Li, Cr, or both; such compositions could easily result from admixture with Mesopotamian glass during the working process. Only one of these samples is cobalt-blue, raising the possibility that non-cobalt-blue glass was more likely to be mixed with Mesopotamian glass, perhaps because the cobalt color was degraded by mixing with other glass or simply that there was already adequate cobalt-blue glass from Egyptian sources. For the 169 non-cobalt colored Amarna glasses in Table S2, combining the seven high-Li or high-Cr Egyptian samples with the seven samples that are Mesopotamian by CA suggests that as many as 14 out of 169, or 8 %, of the non-cobalt-blue Amarna glasses are either Mesopotamian or show mixing with Mesopotamian glass. Although the absolute numbers may differ for other sample sets based on how the samples are selected, favoring one color or another, this new evidence not only multiplies the number of samples, but suggests a substantial presence for Mesopotamian glass in Amarna workshops, providing strong physical evidence for glass exchange from Mesopotamia or the Levant to Egypt as mentioned in the Amarna letters.

In contrast, our Egyptian glass in Mesopotamia, while striking, is more difficult to interpret, particularly because of the relatively small

number of well provenanced Mesopotamian samples with complete chemical analyses. While such shipments as the Egyptian Uluburun glass ingots, most likely staged near the ship's homeport in the Bay of Haifa, certainly provide a potential mechanism for the transfer of Egyptian glass to the Levant, the evidence for such glass at Levantine or Mesopotamian sites remains quite limited. On the other hand, it is possible that further study of these and other Mesopotamian samples will reveal evidence for mixing of local or regional glass with imported Egyptian glass. For example, about half of the 28 Tell Brak glasses in Table S2 have low values for Cr and an equal number are low in Li. Is this the result of natural variability in raw materials or could this be evidence for glass mixing at the site? More complete characterization of the Tell Brak and other Mesopotamian glass may help to answer this question.

5.8. The Mycenaean glass relief beads onboard the Uluburun ship

The sample descriptions and chemical analyses for the five Mycenaean relief beads are given in Table S1. There are two striking findings. First, four of the five beads are similar to one another at the 96 % level by CA, with two of these, KW 5834 and KW 5835, similar at 98 % even though one of the beads had a figure-eight design and the other a spiral. The second finding is that none of the analyzed beads are very similar to the ingots onboard the ship, but instead are much more similar to some of the Amarna cobalt-blue samples, matching at 94 % vs only 90 % to the ingots. This would suggest that if these Mycenaean beads had been made in the same center as the intended destination for the ship, previous shipments would have included glass from Amarna.

The six non-Uluburun Mycenaean beads listed in Table S2 tell a different story. While one of these, copper-blue, was made from Mesopotamian glass, the glass for the five others was Egyptian, with three of the five similar to one another at the 97 % level, as close as our Uluburun batches. What's more, the glass itself was similar to Uluburun batch Co-4 E at 96 %, not quite close enough to say they were from the same batch but close enough to indicate manufacture under very similar circumstances and probably in the same workshop. Because these Mycenaean beads are not accurately dated it is not possible to know if they were made before or after the shipwreck, but they do indicate that the Uluburun shipment was not the only one with glass from this particular production center and that similar glass was used to produce typically Mycenaean objects. While these three beads are all from the Corning Museum of Glass, they were from different sources, with one accessioned in 1959 and the other two in 1976. In addition, the designs of the beads are different, so it is not a case of three samples from the same original object. The currently accepted dates for Amarna, ca. 1348–1320 BCE, overlap much of the radiocarbon dating range for the Uluburun shipwreck, dated 1320 ± 15 BCE. The presence of Amarna-type glass objects on the ship carrying Uluburun-type ingots, along with evidence for the use of Uluburun-type glass for the production of Mycenaean objects, suggests continuity in glass production and exchange, using the same general glass production technology but with a possible change to a new location. Close analysis, including trace elements, of well dated Mycenaean beads will help fill in or perhaps refute this model.

6. Conclusions

The chemical analysis by LA-ICP-MS for almost all Uluburun ingots as well as 49 strontium isotope determinations reported here have provided the basis for new insight into the production and exchange of LBA glass. We interpret these results to suggest that 90 % of the ingots were produced in batches of up to at least 16 ingots. The members of these batches, as defined by CA, have the same chemical similarity as the repeat analyses of Uluburun ingot section CMG 5975 and glass standard NIST 612. Batch production with this high degree of uniformity would have required well-organized workshops and standardized technologies with carefully controlled mixing and firing to produce over 40 kg of homogeneous raw material at one time. Remarkably, at least some of the

ingots from the same batches were kept together from their point of production to staging at a Levantine port prior to final boarding.

We also have identified the production zone for the Uluburun shipwreck glass ingots using multivariate statistical analysis incorporating 35 oxides or elements determined by LA-ICP-MS and by comparing the trace element ratios Li/Zr and Ti/Cr. Both methods indicate that all glass ingots, regardless of color, are Egyptian; strontium isotope studies confirm this as well.

While the Uluburun shipwreck is, by itself, the best evidence for the long-distance nature of glass exchange in the Late Bronze Age, we were able to further explore the impact of ‘foreign’ glass at Egyptian and Mesopotamian archaeological sites. The case for Mesopotamian glass in Egypt, particularly in the workshops at Amarna, is much stronger, with a substantial number, as many as 8 % in the data presented here, of the non-cobalt samples indicating either Mesopotamian glass or possible admixture with Mesopotamian glass. The new evidence presented here for Egyptian glass at Mesopotamian sites, while suggestive, remains limited, with Egyptian samples found at Tell Brak in Syria and Tell al-Rimah in Iraq, along with the use of Egyptian amber-colored glass for the body of a Mesopotamian vessel.

The Mycenaean relief beads found with the ship, plus those included with the supplemental Late Bronze Age analyses, provide intriguing insight into glass trade and Mycenaean bead production. While the relief beads found with the ship had been made with glass much more similar to that found at Amarna, three of the five cobalt-blue beads from the Corning Museum of Glass were actually made with glass almost identical to some of the shipwreck ingots. These results show continuity in Late Bronze Age *trans*-Mediterranean glass exchange but with possible shifts in the Egyptian production workshops.

Subsequent reports will explore more details regarding ingot manufacture, including probable raw materials and workshop practices and how these affected the final chemical compositions. In addition, by comparing the ingots with the other glasses in our dataset, it may be possible to further specify the relationship of the Uluburun glasses with those found at Amarna and other Egyptian sites. We hope that the present publication of the full compositional results for this large group of samples will encourage others to investigate these and other questions important to our understanding of the production, exchange, and use of glass in the Late Bronze Age.

7. Dedication and Acknowledgements

We dedicate this article to the memory of George F. Bass (1932–2021) and Robert H. Brill (1929–2021). Both men were visionaries, playing pivotal roles in the creation and development of their respective specialties of nautical archaeology and glass study as scientific disciplines. We will miss their energy, warmth, and wisdom, and this study would not have been possible without their pioneering work.

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CRediT authorship contribution statement

James W. Lankton: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Cemal Pulak:** Conceptualization, Data curation, Funding acquisition, Visualization, Formal analysis, Investigation, Writing – review & editing. **Bernard Gratuze:** Conceptualization, Methodology, Data curation, Funding acquisition, Investigation, Writing – review & editing.

Declaration of Competing Interest

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

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jasrep.2022.103354>.

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