Glass ingots from the Uluburun shipwreck: Addition of glass cullet during manufacture and evidence for the changing context of New Kingdom Egyptian glass production in the late 18th Dynasty

James Lankton, Cemal Pulak, Bernard Gratuze

Journal of Archaeological Science: Reports 45 (2022) 103596

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Glass ingots from the Uluburun shipwreck: Addition of glass cullet during manufacture and evidence for the changing context of New Kingdom Egyptian glass production in the late 18th Dynasty

James W. Lankton [a,*,1], Cemal Pulak [b,1], Bernard Gratuze [c]

[a] UCL Institute of Archaeology, London, United Kingdom
[b] Institute of Nautical Archaeology, Texas A&M University, College Station, TX, USA
[c] IRAMAT-CEB, UMR 7065, CNRS / Université d'Orléans, Orléans, France

ARTICLE INFO

Keywords:
Late Bronze Age
Uluburun
Glass ingots
LA-ICP-MS
Amarna
Glass cullet
Egypt 18th Dynasty

ABSTRACT

Our recent LA-ICP-MS analyses of glass ingots from the Uluburun shipwreck along with additional samples from Egyptian sites, primarily Amarna, encourage us to question how and where the ingots were produced. Because almost all the approximately 200 glass ingots are either greenish blue, colored by copper, or purplish blue, colored by cobalt, we focus here on these colorants and their attendant trace elements. Based on the evidence for copper and antimony in cobalt-blue glass, we conclude that the most likely explanation is the addition of glass cullet during ingot production, in accordance with suggested evidence from glassmaking texts thought to date to the Late Bronze Age. By comparing the Uluburun ingots with glass from Amarna using multivariate statistics and trace element ratios, we determine that while a few of the ingots might be consistent with Amarna manufacture, the great majority are not, but rather represent production from other workshops, probably following those at Amarna. The importance of these workshops is suggested by our finding that over half of the cobalt-blue Mycenaean relief beads for which trace element data is available were made with Egyptian glass closer to that of the Uluburun ingots than to glass found at Amarna.

1. Introduction

Intensive study of the chemical compositions of the glass ingots from the Uluburun shipwreck (ca. 1300 BCE) as well as other Late Bronze Age (LBA) glass has the potential to provide important insight into the use of raw materials and means of production, allowing us to question how and where the ingots were manufactured. We previously published full LA-ICP-MS (laser ablation-inductively coupled mass spectrometry) analyses of 192 Uluburun glass samples, representing almost all the approximately 200 glass ingots from the shipwreck (Lankton et al., 2022a; Supplement 1 here). In addition, we included LA-ICP-MS results for five Mycenaean relief beads from the shipwreck and 355 LBA glasses from Egypt, the Levant, and Mesopotamia. Based on multivariate statistical techniques of Principal Components Analysis (PCA) and Cluster Analysis (CA), we identified 28 discrete production events or batches for the glass ingots on board the ship. The ingots in each batch match at the same 97 % similarity index observed for repeat LA-ICP-MS analyses of samples from a single ingot, the limit of our ability to separate two glasses based on chemical composition. However, there were considerable differences from batch to batch that may reveal how the three main components of LBA glass (plant-ash flux, silica from crushed and ground quartz pebbles, and transition metals as colorants) resulted in the observed variation.

The importance of the Uluburun glass ingots cannot be overstated since they comprise the largest and most coherent group of Late Bronze Age glasses yet discovered. Although all the ingots were made from Egyptian glass (Lankton et al., 2022a), the actual production site is not certain. The New Kingdom 18th Dynasty capital at Amarna, long appreciated as a glassworking and, more recently, glassmaking (Smirniou and Rehren, 2011) center, has been a prime candidate. Evidence from Amarna includes the remains of glass workshops for making vessels and beads, as well as the recovery of glass ingots (Hodgkinson, 2015) similar in morphology to those from the Uluburun ship. The relatively short duration of Amarna as an Egyptian capital ended shortly after the

* Corresponding author at: 1003 Stern Lane, Foster City, CA, USA.
E-mail address: jamesyuri@gmail.com (J.W. Lankton).
[1] Co-First authors.

https://doi.org/10.1016/j.jasrep.2022.103596
Received 29 April 2022; Received in revised form 8 August 2022; Accepted 8 August 2022
Available online 13 August 2022

death of Akhenaten ca. 1336 BCE (Shaw, 2000), although some industrial activity could have continued at the site. The consensus radiocarbon date for materials on board the Uluburun ship place the wreck slightly later (1320 ± 15 BCE, 68 % probability; Manning et al., 2009), in the post-Amarna phase of the late 18th Dynasty, although overlap is still a possibility. Additional evidence that the shipwreck post-dates Amarna is the recovery in the wreck of a gold scarab with the name of Akhenaten's wife, Nefertiti. This has been interpreted to suggest that the ship sailed after the death of Akhenaten and Nefertiti, when the scarab's only value would have been the weight of the gold or for recycling into jewelry (Weinstein, 2008).

The research presented here focuses on understanding glass-coloring agents, particularly copper and cobalt, to uncover fundamental processes in LBA glass production. In addition, we will examine the relationship between the Uluburun glass ingots and glass from Amarna, as well as the connections with Mycenaean glass relief beads, including those recovered from the shipwreck. As a result of these studies, we interpret variations in antimony and copper content as strong evidence for the addition of glass cullet during production of the Uluburun glass ingots. Furthermore, it is likely that the addition of cullet with various compositions accounts for the variable levels of copper in 18th-Dynasty cobalt-blue glass. Next, based on multivariate statistical analyses and a new way of looking at the colorants used to produce cobalt-blue glass, we conclude that while some of the glass ingots on board the ship might have been made at Amarna, the majority were likely produced elsewhere, thereby representing the first evidence for new primary glass production workshops, probably following those at Amarna, that went on to produce the major proportion of late 18th-Dynasty Egyptian glass.

2. Materials

The data from this paper are from our published (Lankton et al., 2022a) dataset, consisting of 192 glass ingot fragments and five Mycenaean relief beads from the Uluburun shipwreck, as well as 373 LBA glass samples from Amarna and other sites including 18 that had been previously reported (Varberg et al., 2015; Varberg et al., 2016). Please see supplement Table S1 for brief sample descriptions and full compositional results, including cluster assignments from multivariate statistical analyses. For some figures (as noted), we included cobalt-blue Amarna glass analyzed by Hodgkinson and Frick (2020) using LA-ICP-MS, with results very similar to ours for the elements in question. In addition, we supplemented our analyses of cobalt-blue Mycenaean relief beads with those from Smirniou et al. (2012) and Walton et al. (2009). While the Smirniou et al. samples were analyzed in our laboratory using the same protocols and equipment, the Walton et al. beads were analyzed at the Getty Museum using LA-ICP-TOFMS.

3. Methods

3.1. LA-ICP-MS

All samples were analyzed using LA-ICP-MS by James Lankton or Bernard Gratuze at the Institut de recherche sur les archéomatériaux (IRAMAT), Orleans, France. See Lankton et al. (2022a) for full details regarding the technique and equipment as well as a discussion of trueness and precision based on repeat analyses of reference glasses NIST 612 and Corning A. In general, detection limits were in the range of 1 part per million (ppm) for most elements, although they were somewhat higher for elements with high background counts such as Al, P, K, Cr, Fe, Cu, Sb, and Pb. Precision and trueness ranged from 4 % or below for major (greater than 1 wt%) and minor (0.1 to 1 wt%) oxides or elements, and in the 5 % or lower range for most trace elements (less than 0.1 wt% or 1000 ppm). For elements with high background counts present at very low levels (less than 10 ppm), trueness may be excellent but with lower precision.

3.2. Multivariate statistical analysis

PCA and CA were used to determine the similarity between the glasses studied by LA-ICP-MS (Lankton et al., 2022a). Multiple iterations determined that the following oxides or elements, calculated as log-ratio values (Aitchison, 1999), gave the most consistent results: 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. CA was applied to the entire dataset, those from Uluburun as well as the other LBA samples from our laboratory, with groups or clusters formed using the average linkage model. A more comprehensive description of the statistical techniques is provided in Lankton et al. (2022a).

4. Results and discussion

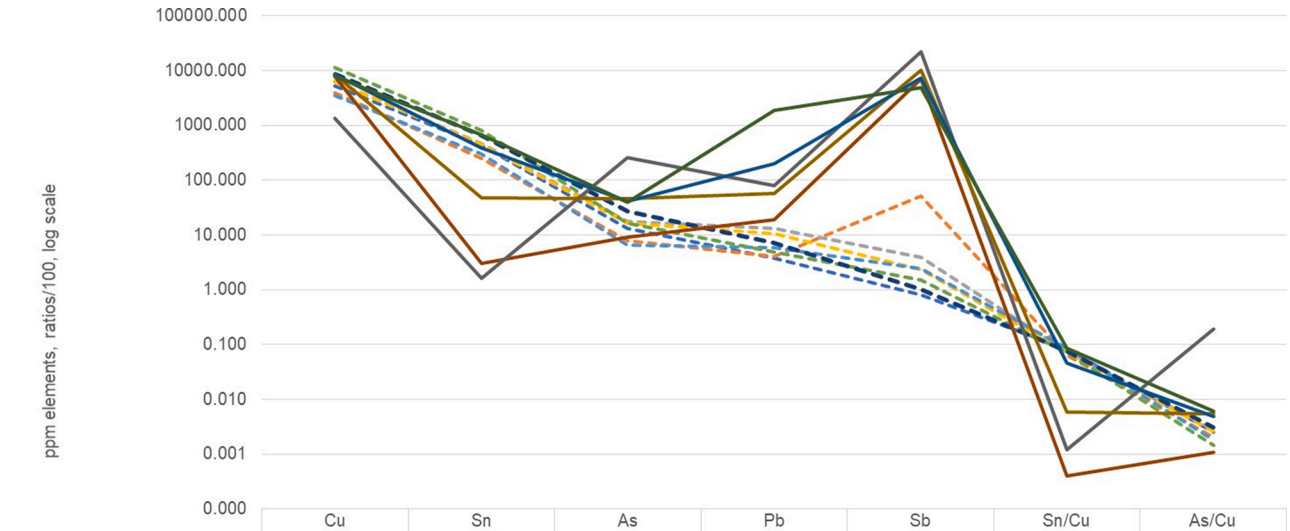
4.1. Colorants

While Cu, Co, Sb, Pb, and colorant-associated elements like Ni, Zn, and As were not included in the statistical analysis, color was consistent within each Uluburun ingot group and for almost all other LBA glass groups, with a few exceptions where both yellow and white glass were contained in the same cluster. For blue glass, constituting 98 % of the ingot samples, this consistency may be related at least in part to the inclusion of colorant-associated major elements like Al_2O_3 for Co-blue glass, or to trace-element patterns associated with individual colorants. Manganese (Mn) was also included in the analysis and was a distinguishing characteristic of the three purple Uluburun ingots. Iron levels for the single amber ingot (KW 3779) were relatively low, with measured Fe_2O_3 0.33 wt% versus an average of 0.49 wt% for all ingots, so this glass may be differentiated more by the lack of colorant-related elements. It is most likely colored by low levels of an iron-sulfide chromophore (Schreurs and Brill, 1984). For this paper, we will use the amber glass composition as our 'colorless' glass compared to those colored by copper or cobalt.

4.1.1. Copper

4.1.1.1. Uluburun copper-blue glass. Fifty Uluburun ingot samples (26 % of total) are transparent greenish-blue colored by dissolved copper oxide (CuO). Elemental values of Cu range from 1778 to 11,630 ppm (mean 5915 ppm), corresponding to CuO 0.22 wt% for a very pale greenish-blue ingot fragment to 1.46 wt%, with mean value 0.74 wt%. While metallic copper was probably added to Mesopotamian glass, the use of scrap bronze or bronze scale was more common in Egypt and contributed, in addition to copper, tin (Sn), arsenic (As), and lead (Pb). For the copper-blue ingots, copper correlates with Sn (R^2 0.77) and As (R^2 0.55), but not Pb. The average Sn/Cu ratio is 7.7 % and As/Cu is 0.2 %, corresponding to the addition of bronze with average composition 7.1 % Sn and 0.23 % As, with a range from 5.2 to 12.4 % Sn and 0.1 to 0.4 % As. Copper-blue compositional group mean values for Cu, Sn, As, Pb, and the Sn/Cu and As/Cu ratios are shown in Fig. 1.

4.1.1.2. Amarna copper-blue glass. By comparison, our 37 Amarna copper-blue samples (Lankton et al., 2022a) are similar in Sn (430 vs 450 ppm for the ingots) and higher in As (45 vs 15 ppm), with average Sn/Cu 5.3 % and As/Cu 0.6 %. However, the variation is much greater. The base glass for one of the Amarna Cu-blue samples (CMG AMN BC 02) has a Mesopotamian composition (Lankton et al., 2022a), and the copper colorant substantiates this with both Sn/Cu and As/Cu around 0.1 %. Sample CMG AMN BC 44b, previously identified as mixed Mesopotamian and Egyptian glass, is similar but with slightly higher As. An additional four samples suggest the use of relatively pure copper with Sn less than 1 % of Cu, while one Amarna sample, CMG 7190 blue Cu, a copper-blue trail on a cobalt-blue vessel, appears to have been colored by the addition of an arsenical copper alloy with Sn/Cu 0.1 % and As/Cu



	Cu	Sn	As	Pb	Sb	Sn/Cu	As/Cu
UBN Cu-1 A	5250	435	13	4	1	8.3%	0.2%
UBN Cu-1 B	3890	249	8	4	51	6.4%	0.2%
UBN Cu-2 A	6227	457	18	13	4	7.3%	0.3%
UBN Cu-2 B	6344	447	16	10	2	7.0%	0.3%
UBN Cu-3 A	3548	301	6	6	2	8.5%	0.2%
UBN Cu-3 B	11232	807	16	5	2	7.2%	0.1%
UBN Cu-4	8742	666	27	7	1	7.6%	0.3%
AMN Mesopotamian	7925	3.0	9	19	7154	0.04%	0.1%
AMN high As, Sb	1351	1.6	262	81	22032	0.1%	19.4%
AMN low-Sn, moderate As	8511	48	45	58	10105	0.6%	0.5%
AMN lower-Sn	8603	384	41.6	201	7391	4.5%	0.5%
AMN higher-Sn	7989	664	39	1843	4941	8.5%	0.6%

Fig. 1. Cu and related trace elements for copper-blue glass groups from the Uluburun shipwreck (UBN) and Amarna (AMN). Note the greater variability in Sn, As, Pb, and Sb for the Amarna copper-blue glasses. All samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

19 %. However, a closer look reveals Sn at the lower limit of detection and Sb at 22032 ppm or 2.2 wt%. The 1350 ppm Cu would not ordinarily produce a blue color, and we suspect that this particular analysis was mixed with white glass from an adjacent trail. The remaining 30 samples contain both Sn and As and are shown in Fig. 1 as 'AMN lower-Sn' and 'AMN higher-Sn.' The 15 samples with higher Sn correspond to copper alloys with Sn from 6.2 to 8.9 % and As 0.3 to 1.7 %. Antimony, an opacifier sometimes added during primary glass production in Amarna (Smirniou and Rehren, 2011), is relatively high for all Amarna groups. A possible Sb/As correlation (R^2 0.5) may indicate that not all of the As in the Amarna samples came from the copper source, since As may also be associated with Sb. The greater variety in Cu sources for the Amarna samples is striking and contrasts with the relative standardization of production for the Uluburun ingots, using less copper except for the three ingots in group Cu-3B, and more uniform raw materials. While Amarna was a likely glass primary production site (Smirniou and Rehren, 2011), there were also glassworking areas with what appear to be multiple workshops (Hodgkinson and Frick, 2020). It is clear from the chemical compositions that the copper-blue glass used to produce ornaments and vessels had a variety of sources including Mesopotamian but also Egyptian workshops that produced blue-green glass, perhaps using whatever copper source was available at the time.

4.1.1.3. Copper-blue glass lithophile element and REE profiles. Other than Cu and related elements (Sn, As, Pb), most of the lithophile and rare earth element (REE) concentrations in copper-blue glass, whether from the Uluburun shipwreck or Amarna, range from 5 to 10 % of those for the continental crust. The relative enrichment in all samples for Li, Rb, and Sr is most likely the result of additional contributions from the plant ash flux used in LBA glassmaking. While contents for the copper-blue ingots overlap those for the 'baseline' amber ingot, the mean values for most elements, including Al, are 20 % higher for the copper-blue ingots, as shown in Fig. 2, which includes the Uluburun Cu-blue ingot group mean values, the Amarna groups mentioned above, and the Uluburun amber ingot, with all elements normalized to the continental crust (Wedepohl, 1995). The Amarna samples with a Mesopotamian composition stand out for their high values for Li and Cr and generally low values for Ti and Zr, as discussed in Lankton et al. (2022a). In addition, Cs, as observed also by Reade (2021, 161), and Ta are high, raising the possibility of other elements that may help separate Mesopotamian from Egyptian samples. As expected, the amber glass is low for most trace elements, particularly REE, although for some lithophile elements Uluburun ingot groups Cu-1 A and B are also very low.

The REE patterns for both the amber and copper-blue ingots are flat with a slight negative Eu anomaly, consistent with a granitic source. The narrow range for most lithophile elements suggests a similar origin for the lithophile trace elements in the Amarna glasses and the Uluburun glass ingots. Rehren (2016) has suggested that the bulk of the lithophile trace elements came from grinding stones used to prepare the batch powder and presented experimental evidence showing that progressive grinding stages contributed increasing amounts of lithophile trace elements that are roughly proportional to the compositions of the granite or granodiorite blocks found at Qantir. We wondered whether the REE followed this pattern as well. Although REE data for Egyptian granite are limited, El-Taher (2007) published analyses of granites from four quarries near Aswan, the most likely source for granite in the New Kingdom. In Fig. 3 we compare El-Taher's neutron activation analysis (NAA) trace element data for an 'old' granite from Wadi El-Allaqi, southeast of the Aswan High Dam, to the data for Amarna and Uluburun glasses. The granite trace element curve, although higher in absolute values, is very similar in shape to those of the copper-blue and amber glasses from the Uluburun shipwreck and Amarna. When divided

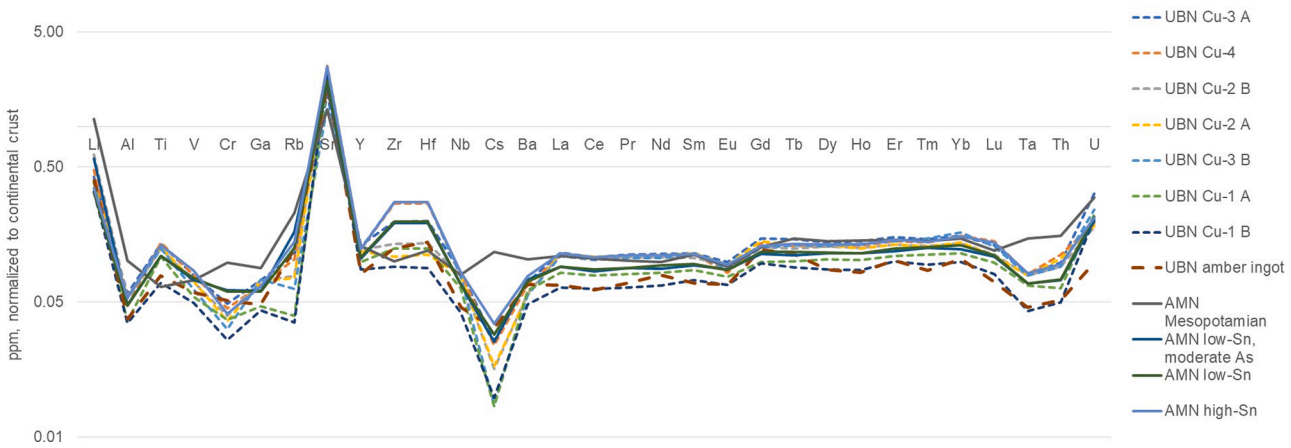


Fig. 2. Lithophile trace elements and rare earth elements (REE) for Uluburun and Amarna copper-blue glass groups. Elements (ppm) normalized to continental crust (Wedepohl, 1995). Log scale. All samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

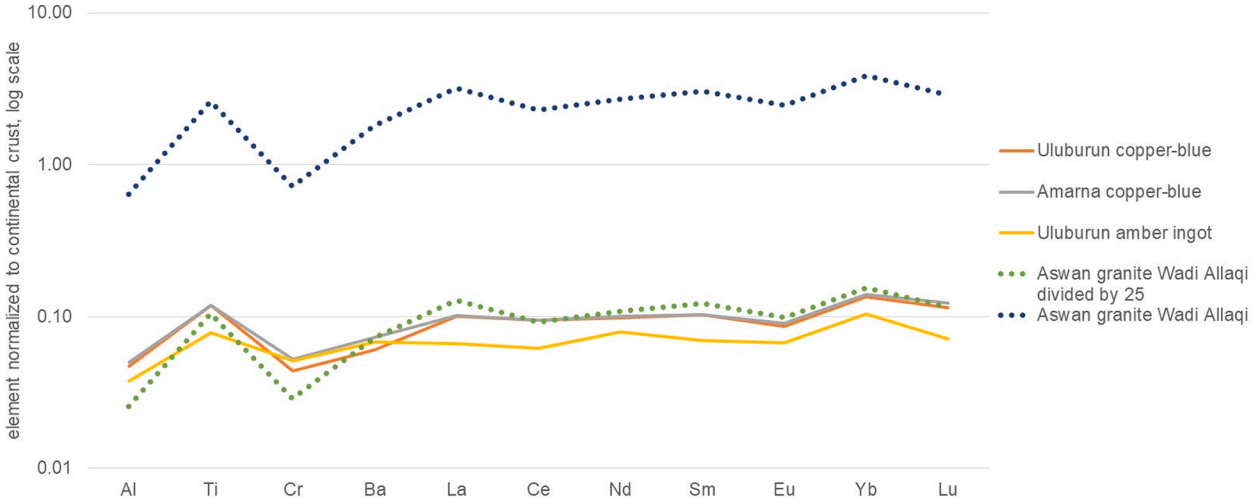


Fig. 3. Lithophile elements and REE for Wadi Alaqi Aswan granite, Uluburun and Amarna copper-blue and amber glass. Elements (ppm) normalized to continental crust. Granite data from El-Taher, 2017. Uluburun and Amarna samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

by 25, indicating a 4 % contribution to the final glass, the granite curve is almost identical to those of the copper-blue glasses. If most of the Al, lithophile trace elements, and REE did come from grinding stones, their procurement and usage appear to be quite similar at Amarna, the presumed production site for most of the glass found there, and wherever the Uluburun ingots were made.

In a different approach to the study of REE content in LBA glass, Reade (2021) compared the chondrite-normalized REE content of Near Eastern glass from Nuzi (Iraq) and Pella (Jordan) to that of chrondrite-normalized MUQ data (Kamber et al., 2005) representing the continental crust. For glass from both sites, there was a consistent ratio of 0.05 to 0.06 between the glass and crustal values. Reade used these ratios to define a 'sedimentary background signal' (SBS) to reflect the amount of sedimentary material incorporated into the glass, whether through plant ash contamination or grinding stones. This SBS ratio of 5 to 6 % is slightly lower than the 10 % we observed for Egyptian copper-blue glass (Figs. 2 and 3), although similar to that for the Uluburun amber ingot, and may indicate overall lower REE levels in Near Eastern glass, perhaps due to the use of different types of grinding stones. It is important to remember that a 5 % REE content compared to crustal values does not necessarily mean a 5 % grinding stone contribution since, as for Egyptian granite, the grinding stones may be relatively rich in REE.

4.1.2. Cobalt

4.1.2.1. Uluburun cobalt-blue glass. LBA cobalt-blue glass may have been an Egyptian specialty, where this lapis lazuli-colored glass was used to produce beads, vessels, and even the dark blue inlays on Tutankhamun's gold funeral mask (Brill, 1976). Purplish-blue glass colored by Egyptian cobalt shows a distinctive chemical signature. Cobalt (Co) may correlate with Al, Ni, Zn, Mn, and Fe, a pattern recognized in the cobaltiferous alums (Kaszmarszyk, 1986) found at the Dakhla and Kharga oases in the Western Desert, with the suggestion that the raw alum was refined before use by precipitation and heating. This resulted in a CoAl-spinel with high alumina and variable amounts of Zn, Ni, Mn, and Fe.

Cobalt blue is by far the most common color in the Uluburun ingots, comprising 70 % of both our ingot samples and the ingots stored in the Bodrum Museum of Underwater Archaeology (Lankton et al., 2022a). Co varies from 293 to 1041 ppm, corresponding to CoO from 0.037 to 0.132 wt%, with a mean Co 445 ppm. Co correlates strongly with Ni (Fig. 4a), with an overall correlation R^2 0.80. However, there appear to be at least two different correlation lines, one slightly higher and steeper

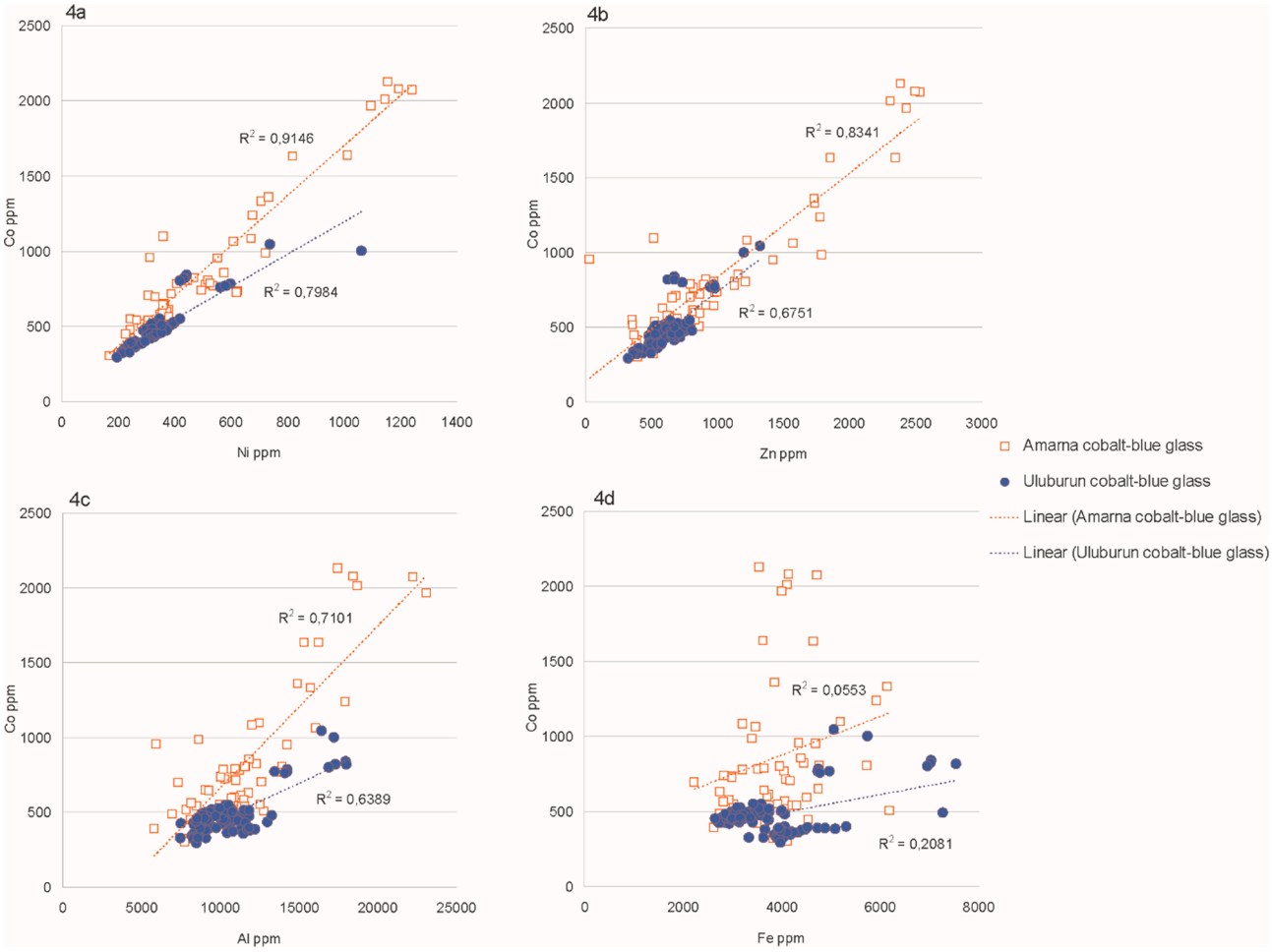


Fig. 4. a-d. Cobalt correlations with Ni, Zn, Al, and Fe for Amarna and Uluburun cobalt-blue glasses. See text for discussion. All samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

than the other, with R^2 0.96 and 0.93, respectively. The plot of Co/Zn (Fig. 4b) is similar although less distinct (overall R^2 0.67), and Co/Al is relatively strong overall (Fig. 4c, R^2 0.64); most of this correlation depends on a few samples with high Co and high Al, and without these samples there is no overall correlation. Again, there may be at least two groups with slightly better correlation. There is no overall Co/Mn (not shown) or Co/Fe (Fig. 4d) correlation, although in both cases there appear to be groups of ingots with stronger associations. We could find no regularity in the differences. For example, the members of groups showing better correlation for Zn were not the same as the members of groups showing better correlation for Mn or Fe. These differences in Co correlation for the various Uluburun ingot groups suggests variability in the chemical composition of the alum-derived colorants used for different ingot batches. Such variability could result from the use of alums from different locations, or, perhaps more likely, from random variations in the alum from a single source. Cobaltiferous alums were mixtures of efflorescent aluminum sulfate minerals with different chemical properties and binding capacity for not only Co, but also Ni, Zn, Mn, and Fe. In addition, variation in processing the alum through dissolution and precipitation could result in the concentration of different chemical species (see Shortland et al., 2006 for discussion), even from the same original batch of alum. Such chemical and technological variability makes it difficult to interpret both differences and similarities in Co/transition metal ratios.

4.1.2.2. Amarna cobalt-blue glass. The Co content of Amarna cobalt-blue samples is almost twice that of the Uluburun ingots. For the 55 Amarna cobalt-blue samples from our database (Lankton et al., 2022a), mean Co is 865 ppm, while for the 37 samples reported by Hodgkinson and Frick (2020) mean Co is even higher at 1142 ppm. As with the ingots, Amarna Co level correlates most strongly with Ni (Fig. 4a, R2 0.91). The Zn and Al correlations are also stronger (Fig. 4b and 4c), with less obvious sub-grouping, at least for Zn. Overall Co/Mn and Co/Fe correlation is weak, with two possible Co/Fe (Fig. 4d) correlation lines. The Amarna samples reported by Hodgkinson and Frick (2020) are similar to Amarna glasses in our database; however, the overall correlations are slightly lower, with R^2 0.7 for Ni, Zn, and Al, no correlation with Mn, and possible slight correlation with Fe (R^2 0.39).

All Uluburun and Amarna cobalt colorants fit the 'Type A' classification suggested by Abe et al. (2012). Although Abe et al. used NiO-CoO-ZnO and NiO-CoO-MnO ternary diagrams to define Type A (18th Dynasty) and Type R (Ramesside) cobalt, we found that a plot of Co/Ni by Co/Zn provides a similar separation, neglecting the low- and high-Mn subgroups in the Ramesside cobalts. Fig. 5 shows the mean values for the Uluburun Co-blue ingot batches along with Co-blue glasses from Amarna, Malkata, Gurob, Badari, Timna, four unprovenanced Egyptian vessels, and nine Mycenaean relief beads, five of these from the Uluburun shipwreck, from our dataset (Lankton et al., 2022a). Also included are Type A and Type R samples from Abe et al. (2012). All Amarna samples, whether from our laboratory or analyzed by Hodgkinson and Frick (2020), along with the Uluburun, Malkata, Mycenaean, and Egyptian-vessel samples show high-Ni, high-Zn cobalts typical for the 18th Dynasty. On the other hand, the Gurob beads and two of the Timna vessels match the lower-Zn Type R compositions from Dashur

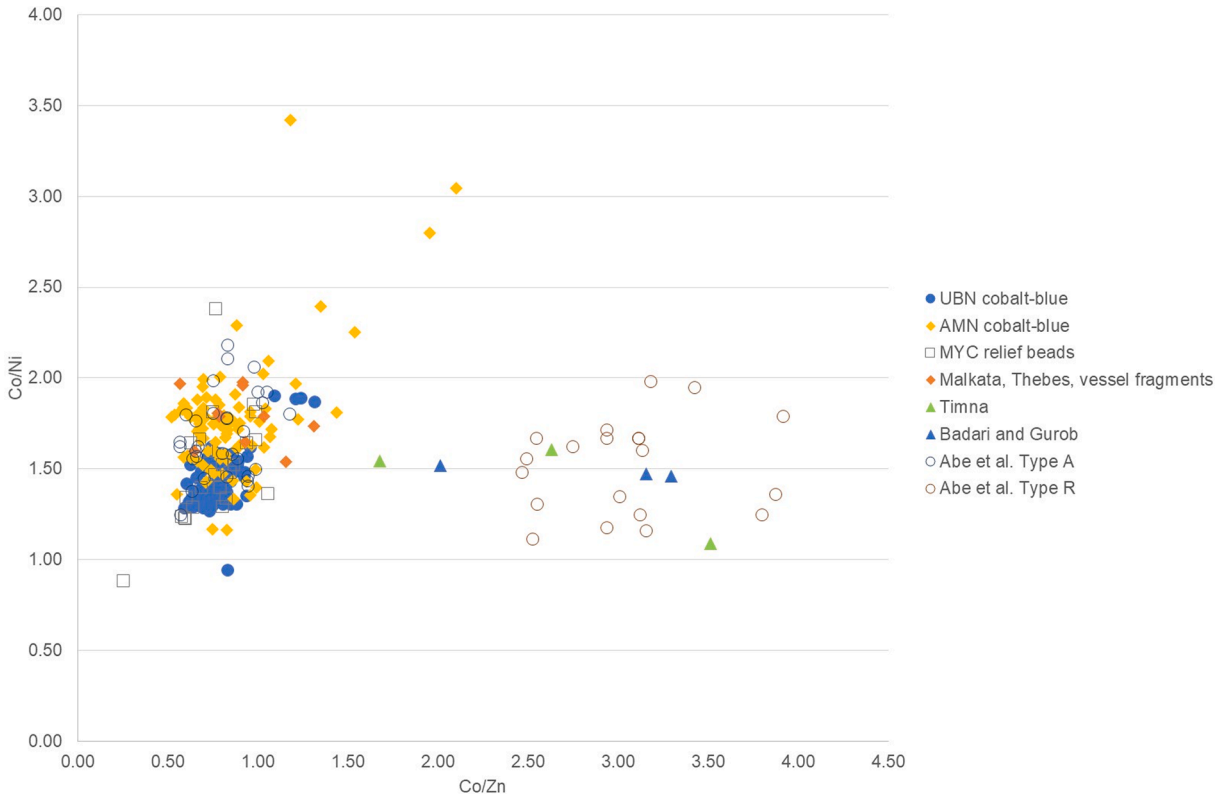


Fig. 5. Co/Ni by Co/Zn for Late Bronze Age Egyptian glass from Lankton et al. (2022a), Hodgkinson and Frick (2020), and Abe et al. (2012). All samples Lankton et al. (2022a) unless noted.

North and Northwest Saqqara (Abe et al., 2012). Two samples appear to be between the two groups: a cobalt-blue bead from Badari and a cobalt-blue vessel with yellow and white trail decoration from Timna. Both of these objects may have come from a production center where cobalt-blue glass with various trace-element signatures was available, possibly indicating later recycling of 18th-Dynasty glass. Neither the Gurob nor the Badari beads are well dated; for these samples, the cobalt typology is useful and could be applied to beads of other colors found in the same groups. It is also noteworthy that the similarities between the Uluburun cobalts and those used for glass from Amarna, Malkata, and the Mycenaean beads support the pre-19th Dynasty date for the shipwreck.

4.1.2.3. Lithophile trace elements and REE in cobalt-blue glass. Fig. 6 shows the mean trace element contents (normalized to the continental crust: Wedepohl, 1995) for the 138 Uluburun cobalt-blue samples and the 92 Amarna cobalt-blue samples, including 55 from our dataset and 37, with results very similar to ours, published by Hodgkinson and Frick (2020). Comparison lines include the mean values for the 50 Uluburun copper-blue ingot samples and 37 Amarna copper-blue glasses (our data), as well as the Uluburun amber ingot KW 3779. While there are considerable differences in the absolute values, the trace element patterns for all glasses are similar from Li to Ba except for higher Al and yttrium (Y) values in the cobalt-blue glasses. The two cobalt-blue glass means are not as close as those for copper-colored glass, where curves are nearly parallel for all elements although the Amarna Cu-blue

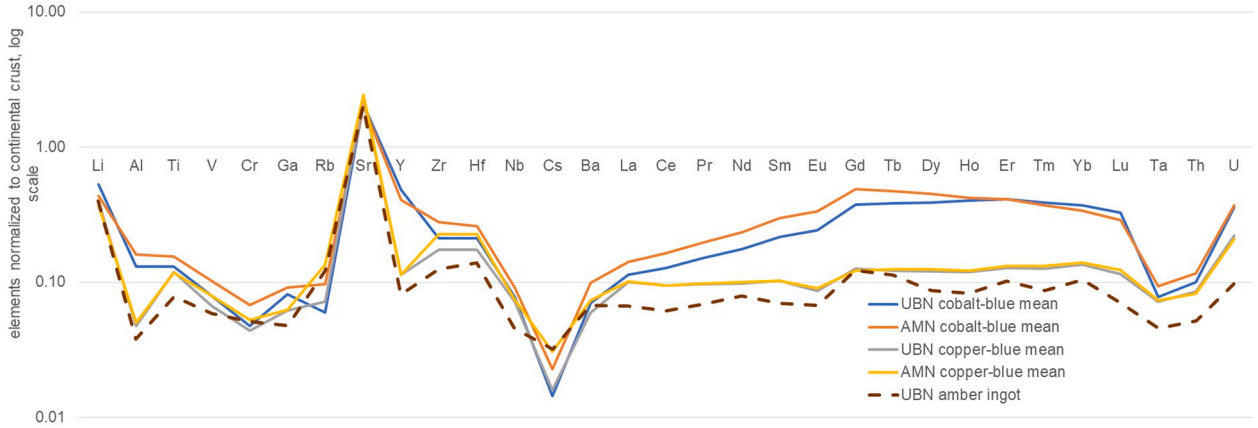


Fig. 6. Mean trace elements normalized to crustal values for cobalt- and copper-blue glass from Amarna and the Uluburun blue and amber-colored ingots. Log scale. Samples from Lankton et al. (2022a) and Hodgkinson and Frick (2020). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

samples are slightly richer in most lithophile traces. In addition to elevated Y, an element that substitutes for and is strongly correlated with the HREE, the cobalt-blue and copper-blue curves separate increasingly at REE above La and the cobalt curves remain elevated through the mid-weight (MREE) and heavy REE (HREE). While Shortland et al. (2007) noted the correlation between cobalt and Y, Walton et al. (2009) discussed and illustrated cobalt-blue glass enrichment in MREE and particularly HREE, demonstrating a similar convex REE pattern for Egyptian and Mycenaean cobalt-blue glasses as well as a glass made using a cobalt alum from the Kharga Oasis. There is considerable variability within our Amarna and Uluburun samples; for example, the cobalt-blue ingots containing the lowest amount of REE are only slightly above the copper-blue ingots. However, on average, the Amarna cobalt-blue samples appear to be higher in REE up to Tb and then lower in the heaviest REE. Our own study of a glass made by heating a Kharga Oasis alum sample (Maurice Picon, alum AL 56a) with added NaOH and SiO_2 showed the same REE pattern as the archaeological cobalt-blue glasses.

How the REE were incorporated and into which species of aluminum sulfate is not clear, but we would expect no mineralogical Co/REE association. While the distribution of REE in efflorescent sulfates from Egyptian desert sites has not been studied, recent work on the incorporation of REE into efflorescent sulfates resulting from the evaporation of acid mine drainage may be relevant (Ayora et al., 2021). Using PCA, the authors sought to link trace elements, particularly REE, to the major sulfate-forming cations (Ca, Mg, Fe, Al) in the sulfate precipitate, a mixture of gypsum, alunogen, Mg sulfates and Fe (II) sulfates similar to those found in analyses of the Egyptian alums (Shortland et al., 2006). PC1 linked LREE and some MREE with Ca, possibly from the gypsum component of the sulfate mixture. PC2 grouped HREE, including Y, together, but not with a particular sulfate cation, while PC3 was formed by Mg, Fe, Co, Cu, Mn, Ni, and Zn, corresponding to Mg-Fe (II) sulfates with substitution of the Mg and Fe by trace elements with similar charge and ionic radius, but not including REE. Based on these results, the authors conclude that the HREE must have formed an independent mineral, not detected by their X-ray diffraction analyses.

These results by Ayora et al. provide insight into the trace element patterns of Egyptian LBA glass, particularly in explaining the association between Co and Ni, Zn, Mn, and Fe. They also demonstrate that REE, including HREE, may be incorporated into evaporitic sulfates, although different species than would bring the Co, Ni, and Zn found in the Western Desert cobaltiferous alums. Because the HREE were not contained in the same minerals incorporating Co, there should be no correlation between Co and HREE, and that is what we found in the Amarna cobalt-blue glasses for Co/Y correlation, using Y to represent the HREE. On the other hand, for the cobalt-blue Uluburun ingots, there is a possible overall Co/Y correlation (R^2 0.37) for the individual samples, and R^2 0.59 for the batch mean values. However, most of this correlation is due to the three batches with highest Co, and if these are removed, the R^2 value drops to 0.20 for the remaining 13 batches. One possible reason for the apparent correlation is the use of different amounts of the same cobalt colorant, since once the cobalt alum was precipitated from the dissolved alums, the ratio of cobalt to REE for that particular colorant would be fixed and adding more or less of the same cobalt colorant to different batches of glass would provide a false Co/HREE correlation.

4.2. What does the CoCu glass reveal about glass production in LBA Egypt?

4.2.1. Mind the gap: Cobalt and copper in cobalt-blue glass

Many Egyptian LBA cobalt-blue glasses contain both cobalt and copper, as observed by Shortland and Eremin (2006) and Smirniou and Rehren (2013); this is true for the Uluburun cobalt-blue ingots as well. While the highest Cu levels for the ingots are below 3000 ppm or 0.375 wt% CuO, the Amarna cobalt-blue glasses are generally richer in Cu, up to Cu 14,814 ppm or 1.85 wt% CuO, with values overlapping those for both Amarna and Uluburun copper-blue samples. However, because CoO is a much stronger colorant than CuO, the glass color remains the typical cobalt purplish blue, even if the mean Co levels are slightly lower in the high-Cu cobalt glasses, with, for the Amarna samples in our database, 834 vs 936 ppm, and, for the Uluburun ingots, 444 vs 481 ppm.

Both Shortland and Eremin (2006) and Smirniou and Rehren (2013) observed gaps in the Cu content of their cobalt-blue glasses, below about 0.1 wt% or 800 ppm, and between 750 and 1000 ppm, respectively. Smirniou and Rehren (2013) suggested Cu 850 ppm as the cutoff between low-Cu or Co-blue glass, and high-Cu or CoCu-blue glass. In addition, they found elevated Sn content in the CoCu glasses as well as slightly lower (by 7 to 27 %) levels of the transition metals such as Fe, Mn, Ni, and Zr usually associated with Co. They considered four possible explanations for CoCu glass, including the mixing of cobalt-blue and copper-blue glass, adding copper to cobalt-blue glass, cobalt-copper glass as a type of cobalt-blue glass resulting from the use of a copper-rich variety of the cobaltiferous alum colorant used in LBA Egypt, and contamination of the cobalt alum by copper and tin during processing in bronze vessels. They concluded that the available data best supported the use of a copper-rich source of cobalt alum, leaving open the possibility that further analyses may change this picture. One scenario that seemed unlikely was the intentional or accidental mixing, perhaps at secondary glass production sites, of cobalt-blue and copper-blue glass since neither base glass compositions nor colorant-associated trace elements appeared compatible with a 3:1 mix of Egyptian Co-blue and Cu-blue glass.

The quantity and quality of our LBA glass data (Lankton et al., 2022a) provide an opportunity to revisit the question of CoCu-blue glass. For the Uluburun cobalt-blue glass ingots, we also observed a gap in Cu values between 690 and 785 ppm and labeled the samples on the low side as Co-blue and on the high side CoCu-blue. This may be a convenient way to sort the samples, but does it make sense in technological or archaeological terms, and will the gap disappear when more samples are examined? Fig. 7a shows the full range of Cu and Sn values for cobalt- and copper-blue samples from Amarna and the Uluburun ingots, with the omission of KW 4400, a cobalt-blue ingot with moderate Cu (452 ppm) and anomalously high Sn (1741 ppm). In spite of considerable variation in all four groups, Cu/Sn correlation is good for the Co-blue and Cu-blue ingots (R^2 0.62 and 0.77) and for the Amarna Co-blue glass (R^2 0.85). The correlation curves for the Uluburun samples are similar in both slope and intercept.

Fig. 7b focuses on Cu between 0 and 2000 ppm. When we include the Uluburun ingots, the Amarna samples in our dataset, and 24 cobalt-blue Mycenaean relief beads, including four from Walton et al. (2009) and 11 from Smirniou et al. (2012), the gap between 690 and 785 ppm Cu disappears. However, there are still no samples between 615 and 690 ppm, nor in the larger gap from 1332 to 1438 ppm. We suspect that these gaps are random, without technological or archaeological meaning. The much more interesting question is why is there Cu at all in the cobalt-blue glass, and what could account for such a wide, almost continuous distribution starting from below 50 ppm, the 'geological background' suggested by Smirniou and Rehren (2013), to, at least for glass from Amarna, levels equal to those for copper-blue glass?

4.2.2. Antimony and the case for adding glass cullet during ingot production

Before attempting to answer this question, it may be useful to look at antimony content in blue glass from the Uluburun ingots and Amarna samples in our database. Antimony (Sb) was a common additive to LBA Egyptian glass, and at higher levels, could opacify blue glass or produce opaque white glass or, in combination with lead, yellow glass. Sb could be added along with other colorants and was sometimes included in the original colorless batch as suggested by evidence from Amarna (Smirniou and Rehren, 2011). Fig. 8 is a plot of Sb by the ratio of Co to Cu for Amarna and Uluburun cobalt-blue and copper-blue glasses. Antimony levels vary from near zero to above 1 wt%, with no clear division to

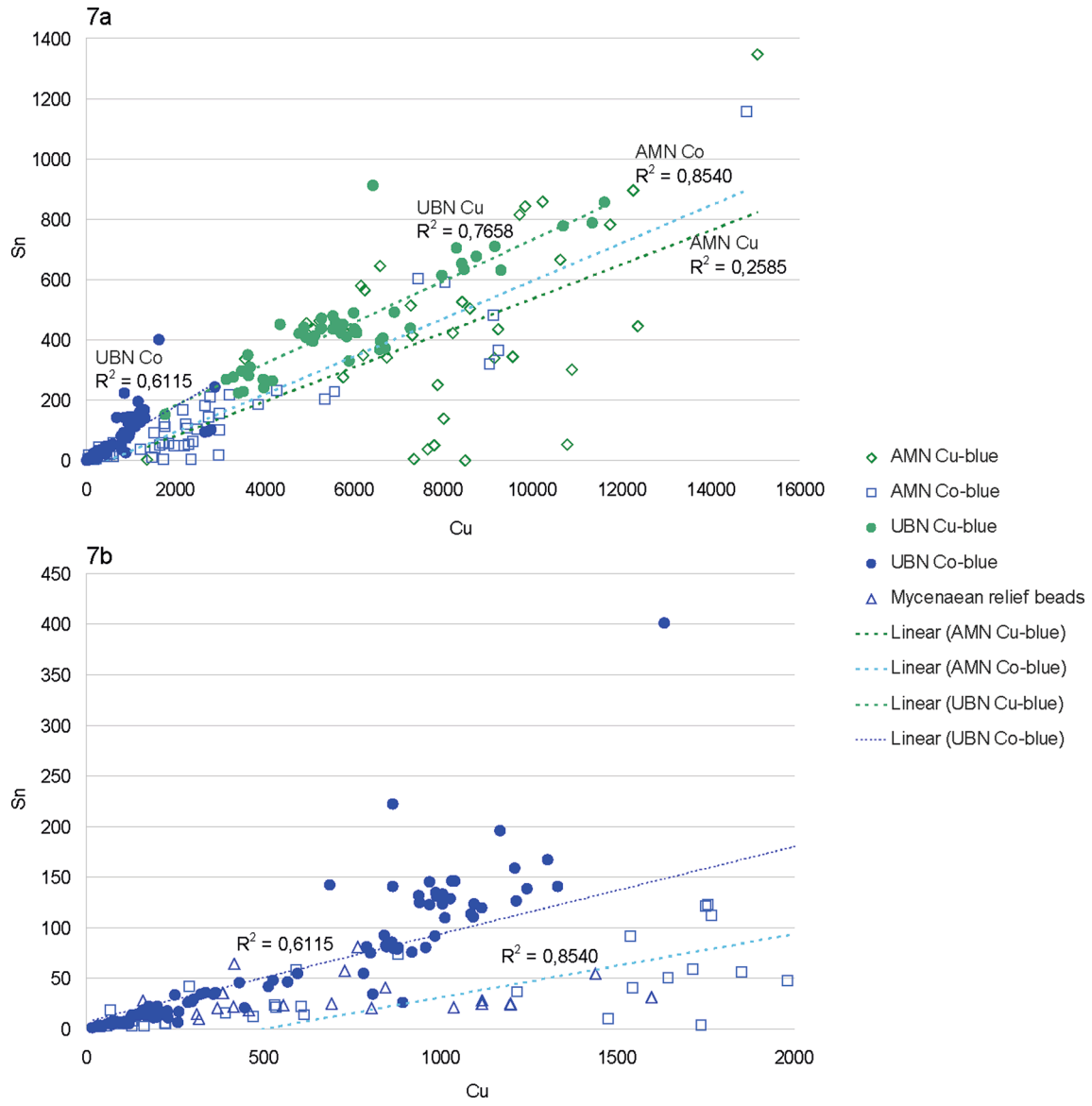


Fig. 7. a and b. Sn vs Cu for cobalt- and copper-blue Uluburun ingots and glass from Amarna for the full range of values (a) and for Cu up to 2000 ppm (b). Uluburun and Amarna samples Lankton et al. (2022a). Mycenaean relief beads from Lankton et al. (2022a), Walton et al. (2009), Smirniou et al. (2012). All samples LA-ICP-MS in our laboratory except for the LA-ICP-TOFMS results of Walton et al. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

indicate whether Sb was an intentional additive or a contaminant. In any case, Sb levels are considerably higher in the Amarna samples, with only a few values below 100 ppm, while few Uluburun ingots have values above that amount. Of these, batch Co-4B, a group of at least eight ingots, stands out with mean Sb 296 ppm and Cu 1007 ppm. In addition, there is one Co-4 outlier with Sb 573 ppm and four other ingot groups with slightly elevated Sb levels ranging from 4 to 90 ppm, still low compared to the Amarna samples but higher than half of the ingot groups with Sb 1 ppm or lower. The Sb content within the Uluburun production groups is very consistent, with relative deviations around 10 % for most of the higher-Sb samples even though Sb was not included in the statistical analyses by which the production batches were defined. It is unlikely that this relatively modest amount of Sb would have been an intentional addition, since there was no visual sign of the opacification for which Sb was usually added to blue glass. As approximately the same amount of Sb is found in all ingots in a given batch, the introduction of Sb must have occurred prior to the final grinding and mixing stages before melting the glass powder in ingot molds.

The Uluburun ingots in group Co-4B, and some but not all of those with higher Sb, are also higher in Pb (ca. 130 ppm), suggesting that Sb may have been introduced into the glass with or without Pb, providing further evidence that Sb was not added as an intentional pigment. The most likely source of the added Sb and Pb would be other glass. The Sb/Pb profiles of Amarna copper- and cobalt-blue, white, opaque green, red, and yellow glasses from our database (Fig. 9) provide candidates that would account for all observed quantities and ratios of Sb and Pb in the Uluburun ingots. For example, if yellow or opaque green glass were added, we would expect to see elevations in both Sb and Pb, in the former without Cu and in the latter with Cu; if white glass were added, only Sb would be elevated, as seen in many of the cobalt-blue ingots. We interpret the Sb and Pb values in the Uluburun ingots as evidence that glass scrap or cullet, with variable amounts of Sb and Pb, was added during ingot production. For the ingots with low Sb and Pb, the cullet must also have been low in Sb and Pb, and many of the Amarna blue glasses, as well as almost all the ingot glass, could have been used. However, this addition would be difficult to detect unless some other

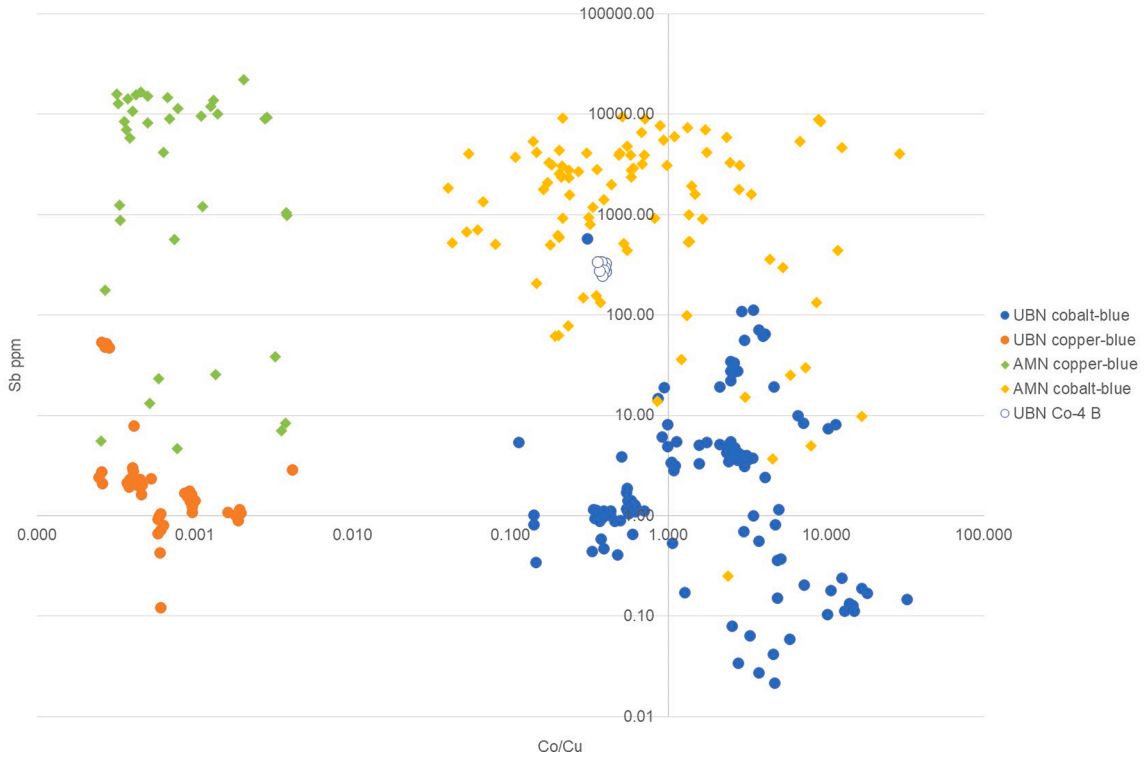


Fig. 8. Sb vs Co/Cu for Uluburun and Amarna cobalt- and copper-blue glass. Amarna samples from Lankton et al. (2022a) and Hodgkinson and Frick (2020). Log scales. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

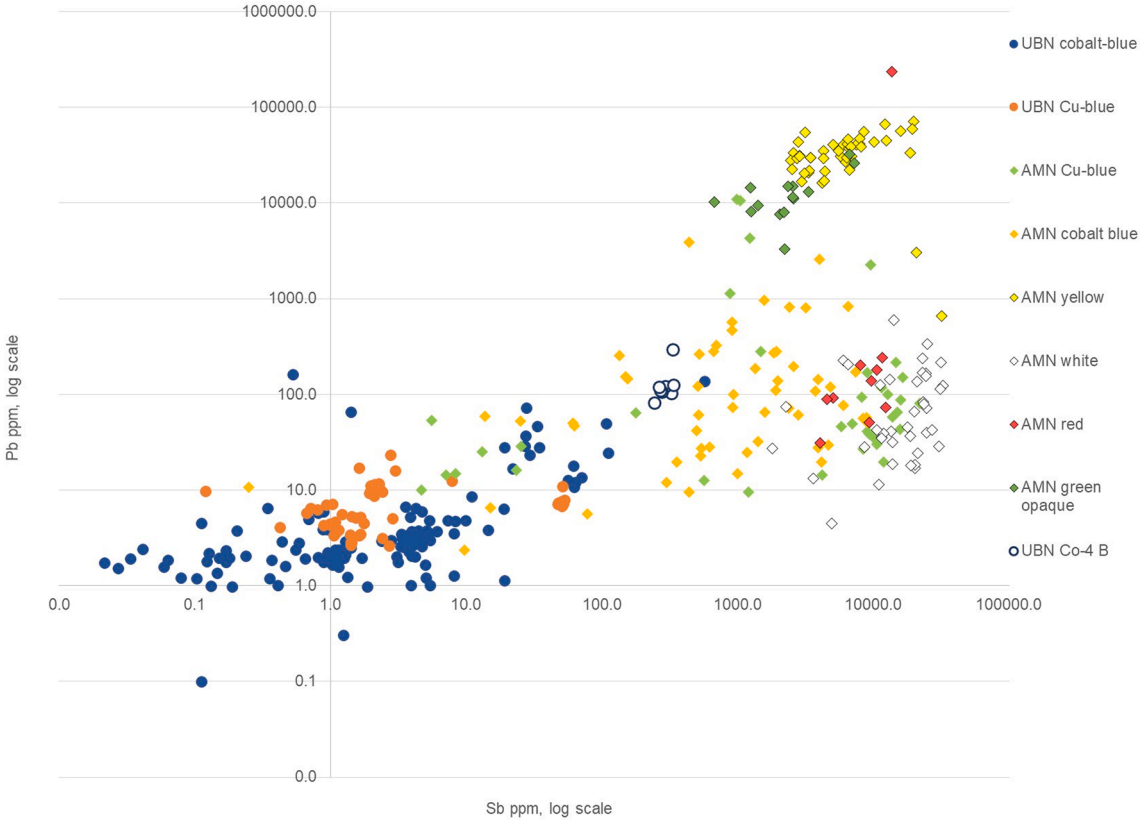


Fig. 9. Pb vs Sb Uluburun and Amarna glasses highlighting the intermediate position of Uluburun ingot batch Co-4B between most of the cobalt-blue ingots and copper-blue, yellow, and opaque green glasses from Amarna. All samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

elements or colorant were high in the cullet but not in the final glass. Based on the Sb and Pb values alone it is difficult to estimate how much cullet was added since it is possible or even likely that the added glass was heterogeneous. If we assume that only one color of cullet was added in the production of the Uluburun high-Sb ingots in group Co-4B, which also contain Cu, a 1:10 addition of green glass with Sb and Cu at the mean values of our 15 Amarna opaque-green glasses would provide the right amounts of Sb and Cu, although in this case the final Pb values would be too high. On the other hand, there are several green Amarna glasses for which a 1:20 ratio would satisfy the values for Sb, Pb, and Cu levels. Alternatively, many of the Amarna copper-blue glasses would be a good fit at 1:10 ratio for one or two of the trace elements, but not all three. However, there is no reason to expect that our 37 copper-blue Amarna samples represent all possible chemical compositions, so copper-blue glasses with the appropriate compositions may well have been available.

4.2.3. Textual clues

Mesopotamian glassmaking texts discovered at Nineveh in northern Iraq are thought to reflect LBA technologies (Oppenheim et al., 1970) and provide possible evidence for the use of cullet during glassmaking operations. Several cuneiform tablets describe combining two raw materials, probably quartzite pebbles and plant ash, along with 'White Plant,' possibly a lime compound, to produce *zukû-glass* (Oppenheim et al., 1970, Tablet A, p. 35). This colorless product was then mixed with 'slow' copper to form *tersītu-preparation*, an intermediate glass that was not used on its own. In the final steps, the *tersītu-preparation* was combined with up to two additional types of glass, *būşu-* and *anzahhu-glass* before re-melting to make finished *zagindurû*-colored glass (probably similar in appearance to a greenish type of lapis lazuli).

The identity of *būşu* and *anzahhu* is not known, but the *anzahhu-glass* could also be used on its own to produce beads and vessels. *Anzahhu-glass* is also the only ingredient that is described as 'washed' in the instructions, and the Akkadian name suggests that it was made by other craftsmen and brought from the outside into the primary glassmaking workshop. Whether *būşu* or *anzahhu* were glass cullet or not is not clear, although the treatment of *anzahhu*, washing before use, parallels the preparation of waste glass or cullet in both ethnographic and commercial glass furnaces. In any case, the texts demonstrate that LBA glassmakers were familiar with adding finished glass at various steps prior to final glass production. The addition of glass cullet would have had definite advantages in decreasing the required melting temperature and increasing the efficiency of the glass melt, thus lowering the overall energy required (Deng et al., 2019).

For the Uluburun ingots, the addition of glass cullet with different amounts of Sb and Pb seems to be the best and perhaps the only explanation for the Sb and Pb levels that vary by production group, since different cullet would have been added to different groups depending on what might be available on a given day. When the glass for Uluburun group Co-4B was made, the cullet happened to be high in both Sb and Pb, possibly including yellow or opaque green glass or both. The many Uluburun compositional groups with low Sb levels could be made by adding cullet with a low-Sb Amarna composition or leftover fragments from almost any previous ingot batch. For the cobalt-blue ingots, the glassmakers must have known that adding small amounts of glass of other colors would not have a noticeable effect on the final color. The technical requirements were different for copper-blue ingots, colored, as mentioned above, by relatively low levels of CuO. None of the Uluburun copper-blue glasses contains significant cobalt, since even small amounts could affect the final color; thus, only non-cobalt-blue cullet could be used. To our knowledge, the Sb/Pb variation in the Uluburun glass ingots provides the first and best evidence for the addition of cullet in LBA glassmaking.

4.2.4. The case for CoCu glass

If added cullet could explain the Sb/Pb levels in the Uluburun glass ingots, what about Cu in glass colored by Co? Fig. 7a plots Sn and Cu for cobalt-blue and copper-blue glass from Amarna and the Uluburun ingots. The Uluburun copper- and cobalt-blue ingots show good Sn/Cu correlation, with R^2 0.77 and 0.61, respectively. As expected from the variety of copper sources at Amarna, overall correlation between Sn and Cu for the Amarna copper-blue glasses is poor, with R^2 0.23, although the correlation is higher for the Amarna cobalt-blue glasses at 0.85, in part because of one sample that is very high in both Sn and Cu. The plots for the Amarna and Uluburun samples have slightly different slopes. Notably, the slopes for copper- and cobalt-blue glasses from the Uluburun samples are almost continuous, suggesting that the copper source is similar regardless of the absolute amount present. Fig. 10 takes this one step further to the very lowest Cu levels. At least for the Uluburun ingots, even the smallest amount of copper is accompanied by Sn. This correlation between Cu and Sn at all levels rules out the possibility that Cu was introduced as part of the cobalt colorant. If that were the case, there would be no Cu/Sn correlation. There is still a chance that small amounts of Cu and Sn could have resulted from contamination by bronze glassmaking tools or vessels, but it is difficult to attribute the higher levels of Cu and Sn, in some cases almost equal to those in the Cu-blue glass, to contamination alone. While the Uluburun ingots show a consistent Sn/Cu correlation, even for very low Cu, this is not true for cobalt-blue glass from Amarna, where there is no apparent Sn/Cu correlation at the lowest values of Cu, despite an overall correlation (R^2 0.85) when all Amarna cobalt-blue glasses are considered. The Amarna data are still consistent with the addition of cullet but in this case, cullet with variable amounts of Cu and Sn, as seen in the copper-blue glass from Amarna. In addition, it is useful to keep in mind the important differences between a primary workshop producing ingots from freshly-made glass, and a secondary workshop making vessels and ornaments by reheating and likely mixing together glass from different sources.

It may seem intuitive that if the Cu in cobalt-blue glass ingots resulted from the addition of high-Cu cullet during glass production, then the ingots with high Cu should have lower values for Co and its associated elements Al, Ni, Zn, Mn, and Fe. Indeed, Smirniou and Rehren (2013) observed such a change, as did we for our cobalt-blue Amarna samples. However, this relationship is less clear for the Uluburun ingots for two important reasons. First, although we studied almost 200 ingots, almost all were members of production groups or batches, each internally identical. Thus, for the 20 batches of cobalt-blue ingots there are essentially only 20 different glass compositions. This limited sample size compounds the second reason: significant compositional heterogeneity between groups with relative deviations (RD calculated as standard deviation divided by mean value) between 30 and 40 % for the elements in question, which is sufficient to obscure 10 to 20 % changes that would result from cullet addition.

4.3. The Uluburun glass ingots in the context of late 18th-Dynasty Egyptian glass

This brings us to our last question: how do the Uluburun ingots fit into the context of Egyptian glass production in the late 18th Dynasty? While the ingots were found together as cargo on the ship, this does not necessarily indicate that they were made in the same place or even at the same time. Evidence from pottery and utilitarian objects on board suggests that the ship sailed from a Levantine port, possibly somewhere in the Bay of Haifa (Pulak, 2008). Therefore, the Egyptian materials, including the glass ingots, would have been transported and stored there until they were loaded on the ship along with the other cargo.

As mentioned in the Introduction, radiocarbon dates for materials on board the Uluburun ship suggest that the wreck followed the main phase of Amarna occupation that ended with the death of Akhenaten and ascension of Tutankhamun. During this time, Tutankhamun and his successor Ay may have reoccupied the Malkata palace of Tutankhamun's grandfather, Amenhotep III, 250 miles south of Amarna, near Luxor. Although Amarna was no longer the capital and was eventually

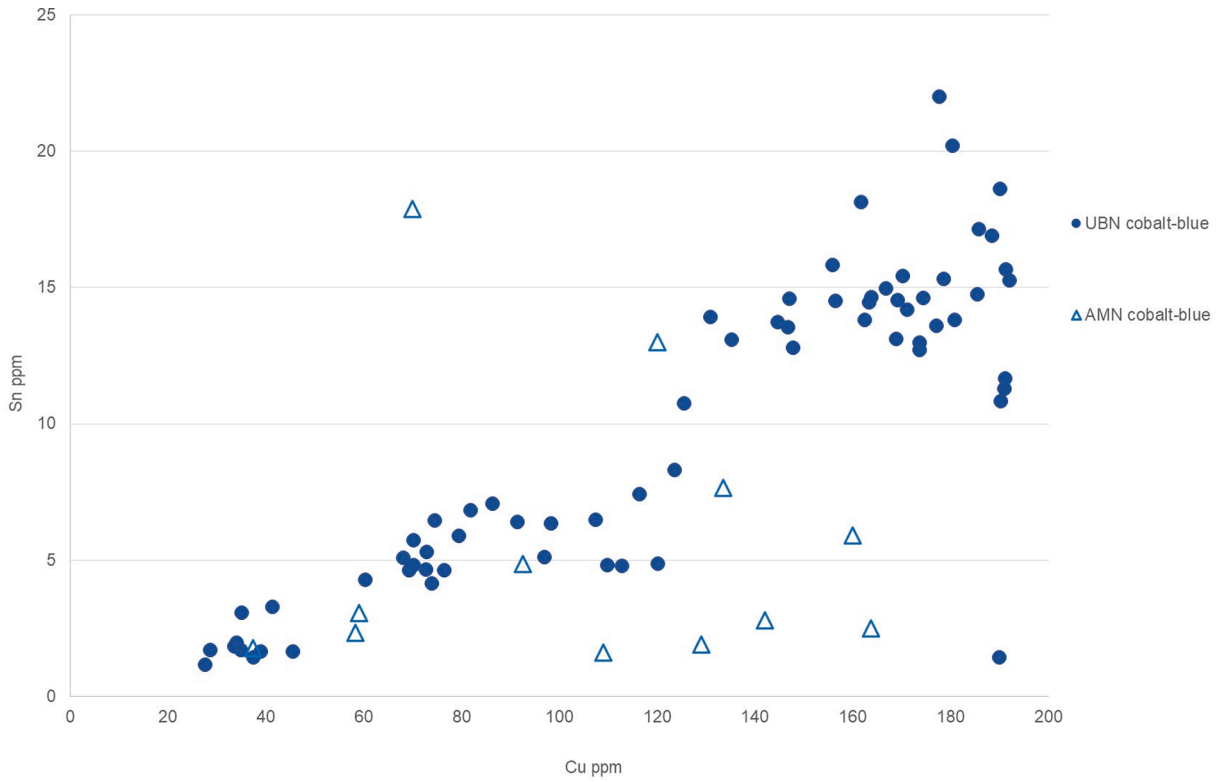


Fig. 10. Sn vs Cu for Uluburun and Amarna cobalt-blue glass for Cu between 20 and 200 ppm. All samples Lankton et al. (2022a). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

destroyed, it is not known whether some industrial production, including for glass, was continued there. Furthermore, glass made at Amarna may have been collected and stored for some time prior to its eventual distribution and use. Because Amarna is the only known Egyptian glass primary production center during the second half of the 14th century BCE, it is particularly important to understand the relationship between the Uluburun glass ingots and glass found, and presumably made, at Amarna. We will approach this question in three ways; first, by taking a closer look at the chemical compositions filtered through cluster analysis; second, by examining the types of cullet that may have been added to the ingots; and third, by studying the REE patterns associated with the cobalt colorants used at Amarna, for the Uluburun ingots, and for Mycenaean relief beads made from Egyptian cobalt-blue glass.

4.3.1. Results from statistical analysis

Principal Components Analysis (PCA) and Cluster Analysis (CA) of 192 Uluburun glass ingot samples combined with 378 other LBA glasses from our laboratory, including five Mycenaean relief beads from the Uluburun shipwreck, provide a chemical compositional similarity index to evaluate the closeness of one sample to another (the full cluster analysis available in Table S1). As mentioned in the Introduction, by including repeat measurements from the same large ingot section we concluded that at a similarity index of 97 % the closeness of two samples was within the error of our LA-ICP-MS determinations, thus enabling us to define ingot batches produced at the same time and with the same ingredients and technological processes. In some cases, there were other ingots similar at 96 % that may have been group members as well, since even some of the duplicate measurements differed by this amount, most likely because of heterogeneity in the glass. Our Sr isotope results were identical for samples matching at 96 or 97 % similarity index, with one exception, but at 95 % statistical similarity, the isotope results began to diverge (Lankton et al., 2022a). Our interpretation was that a similarity index of 97 % was strong evidence that two samples were made from the same glass, and at 96 %, very closely related and possibly the same, but at 95 % perhaps related but not matching. Applying these criteria to the Uluburun glass ingots and the other LBA samples allows us to determine how closely related were the Uluburun ingots to other LBA Egyptian glasses.

The majority of our non-Uluburun Egyptian samples are from Amarna and were recovered during the late 19th- and early 20th-century explorations of W. M. Flinders Petrie. Eventually, these samples were dispersed among museums in Europe and the US. Despite their varied trajectories, 68 % of our 225 Amarna samples were similar at the 95 % level or higher to at least one other sample, mostly from Amarna but also from other sources, including Malkata. Even more surprising, 20 % of the Amarna samples matched another sample at a level of 97 % or greater. Often this was a case of trailed decorations with the same composition on different vessels, indicating that the vessels were most likely produced or at least decorated in the same workshop. Another 26 % of the Amarna samples had a match at 96 % with a further 23 % matching one or more other samples at 95 %. We conclude from these figures that our statistical analysis could identify similar glasses for LBA samples in general, and not just for the Uluburun ingots.

Thus, it is striking that there are very few possible matches between the Amarna samples and the Uluburun ingots, with no matches at 97 %, two at 96 %, and five at 95 %. The most convincing link is a 96 % match between four ingots from Uluburun group Co-4 E, three Mycenaean relief beads from the Corning Museum of Glass, and one Amarna sample. While the match with the three relief beads may be strong evidence for a connection, the single Amarna match is difficult to interpret even though one other Amarna glass joins the group at a similarity index of 94 %. The other match at 96 % is between an outlier from Uluburun group Cu-4 and one sample from Amarna, with four other Amarna samples joining at 95 %. The four other members of Cu-4 are not part of this cluster but they do show similarity at 95 % level to three other Amarna samples. The single match for the Cu-4 outlier may be coincidental, but in general, it seems that there is some similarity between

group Cu-4 and the glass from Amarna. The other matches at 95 % are between one Amarna sample and the glasses from Uluburun groups Co-3C and Co-3 D; one Amarna sample and ingots from group Cu-3B; one Amarna sample and an outlier from Uluburun group Cu-2B; and one Malkata sample and the Uluburun amber ingot. An additional three Amarna samples and one from Malkata that join the groups at 94 % similarity perhaps strengthen the possible link between Amarna and Co-3C and Co-3 D ingots. The amber ingot is also similar at the 94 % level to two additional Amarna samples and one unprovenanced vessel fragment from a museum collection, but not to any of the other Uluburun ingots. Of the three purple ingots, two were made from identical glass and are unlike other Uluburun or Amarna samples, while the third (Lot 10067) is similar to one Amarna sample at 94 %.

In summary, less than 1 % of Amarna samples show a close (96 %) statistical match to the Uluburun ingots, with only 8 % having even a passing similarity (94 % similarity index). Viewed from the Amarna perspective, 33 of 192 ingot samples (17 %) are similar to at least one Amarna or Malkata glass at the 95 or 96 % index and most of the similarities are to just a few Amarna samples. All these similar samples are from ingot batches Co-4 E, Cu-4, Co-3C and D, Cu-2B (one outlier) and the amber ingot. While the similarity between the ingots of group Co-4 E and the three Mycenaean beads seems convincing, the evidence for Uluburun/Amarna connections is weak because so few Amarna glasses are involved. There are many strong links among Amarna samples, as noted above, but not to the Uluburun ingots. If anything, Uluburun group Cu-4 is the closest to Amarna among the copper-blue ingots while groups Co-3C and D are closest to Amarna among the cobalt-blue ingots. The amber and one of the purple ingots are more like Amarna than Uluburun glass, although the similarities are not particularly strong. The five Mycenaean relief beads from the Uluburun shipwreck are another group of interest as all were made from Egyptian glass. One of these is not similar to any other samples in the database, while the other four are similar to each other at 96 %, with two made from glass identical at 98 %, even though the designs on the two beads are different. These samples have no strong links to the other Uluburun glasses, but match one Amarna sample at 94 %, perhaps close enough to provide a hint as to where the original glass was made.

4.3.2. *Cullet compositions*

4.3.2.1. Copper-blue glass. If we accept that glass cullet was added during the production of cobalt-blue glass, as suggested by the Sb and CoCu evidence presented above, it seems likely that cullet was added during the production of other colors as well. While much of this cullet could have come from recycled or waste ingots from the same workshop (s) that produced most of the ingots, in some cases the compositions of the Uluburun glasses are compatible only with the use of cullet having a composition not found among the ingots. The most obvious case is ingot batch Cu-4. Here, the Sb content is about 50 ppm and Co level is negligible, suggesting that a relatively high-Sb, low-Co cullet was used. There are no Uluburun Cu-blue glasses that fit the compositional criteria, but many (71 %) of the Amarna Cu-blue samples provide the necessary levels if added in some proportion less than 10 %. Thus, the seven ingots that comprise group Cu-4 were made where and when Amarna-type glass was available. Remembering that Cu-4 is also the group that most closely matches an Amarna chemical composition, the cullet evidence strengthens the possibility that the ingots of group Cu-4 may have been made at Amarna.

4.3.2.2. Cobalt-blue glass. CA analysis indicates that Uluburun groups Co-3C and D seem more likely to have a connection with Amarna, even if the connection is not as strong as for group Cu-4. Both Co-3C and Co-3 D are very low in Sb, with Cu at about 100 ppm and small amounts of Sn (7 ppm), As (1 ppm), and Sb (1 ppm). There are very few Amarna samples, whether Cu-blue or Co-blue, that would provide the needed composition, since in most cases the resulting Sb would be too high. On the other hand, many of the Uluburun cobalt-blue glasses, with Cu between 1000 and 2000 ppm, would work well in a proportion of 5 to 10 %. In this case, the cullet mismatch with Amarna glass weakens the statistical evidence.

4.3.2.3. Amber glass. If cullet were added during the production of the amber-colored Uluburun ingot, it must have been low in all colorants since Sb, Co, and Cu are all very low in the resulting ingot. While it is possible that the amber ingot was made in the same workshop as the blue ingots, we wondered how well glasses found at Amarna would have worked as cullet to produce the amber ingot. Of the 15 colorless or amber-colored Amarna samples, one has a Mesopotamian composition and half are too high in Sb for even a 5 % contribution. On the other hand, the remaining half have compatible compositions suggesting that amber-colored Amarna cullet may have been used to produce the amber-colored Uluburun ingot. While this cullet evidence does not prove an Amarna origin for the Uluburun amber ingot, taken with the statistical similarity to Malkata and Amarna samples, the Uluburun amber ingot is more consistent with an Amarna origin than are the great majority of the cobalt- and copper-blue ingots.

4.3.3. *REE patterns in Uluburun and Amarna cobalt-blue glass and Mycenaean relief beads*

We mentioned earlier in this report that the Uluburun and Amarna cobalt-blue samples appeared to differ in cobalt-associated REE, with the Amarna glasses high in REE up to Tb and then decreasing in the heaviest REE, while the Uluburun ingots are relatively high in HREE (Fig. 6). This pattern is more obvious in Fig. 11, showing Y and REE, normalized to the continental crust, for representative Amarna and Uluburun cobalt-blue glass groups. We wondered if it were possible to quantify the shapes of the REE curves and selected four elements: Y, representing the HREE in general; La, as an index point for the other values; Yb, to represent HREE; and Dy, as an index of MREE. Fig. 12 is a plot of two ratios based on these values, Y/La and Yb/Dy, for the Uluburun and Amarna cobalt-blue glasses plus those from Malkata and four unprovenanced vessels in our database, five Mycenaean cobalt-blue relief beads from the Uluburun shipwreck, and four relief beads from the Corning Museum of Glass. Two additional groups are 11 cobalt-blue relief beads excavated from a Greek tholos tomb at Volos dated to the 14th/13th century BCE (Smirniou et al., 2012), and four relief beads from the J. Paul Getty Museum (Walton et al., 2009).

Almost all Uluburun and Amarna samples separate into two fields. Those from Uluburun tend to be higher in Y/La and Yb/Dy, with one group of samples, the low-REE ingots from groups Co-3C and D (Fig. 11), correspondingly low in Y/La but high in Yb/Dy. The Amarna glasses, along with those from Malkata, are mostly lower in Y/La and lower in Yb/Dy. Two of the three unprovenanced Egyptian vessels are also in the Amarna range. The Mycenaean relief beads plot in both regions of the graph. Three of the beads from the Corning Museum of Glass are in the Uluburun zone, confirming the CA results, while the fourth may be intermediate between Uluburun and Amarna. The five beads from the shipwreck are either closer to Amarna or intermediate. The Volos samples include six that overlap Uluburun and three that overlap Amarna fields, along with two that appear to be between the two groups. Three of the Getty Museum samples are in the Uluburun zone, and one is closer to Amarna. It is not surprising to find relief beads with intermediate compositions since the mixing of two morphologically similar but chemically different cobalt-blue glasses could have been a common practice at secondary workshops. While the elements on which Fig. 12 is based were included in the statistical analysis, using only REE ratios appears to mirror the result of the full CA while allowing the inclusion of additional cobalt-blue samples for which REE measurements are available.

Both CA and cobalt-associated REE patterns suggest that the

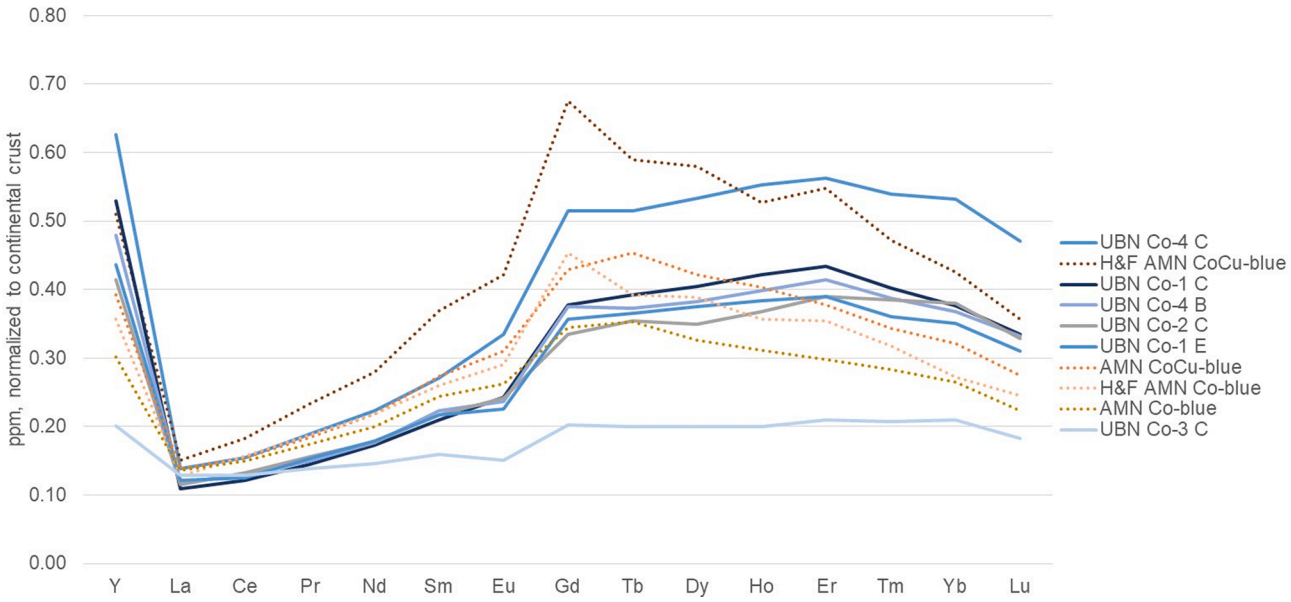


Fig. 11. Representative values for Y and REE normalized to continental crust for Amarna and Uluburun cobalt-blue glass groups. Amarna samples from Lankton et al. (2022a), and Hodgkinson and Frick (2020). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

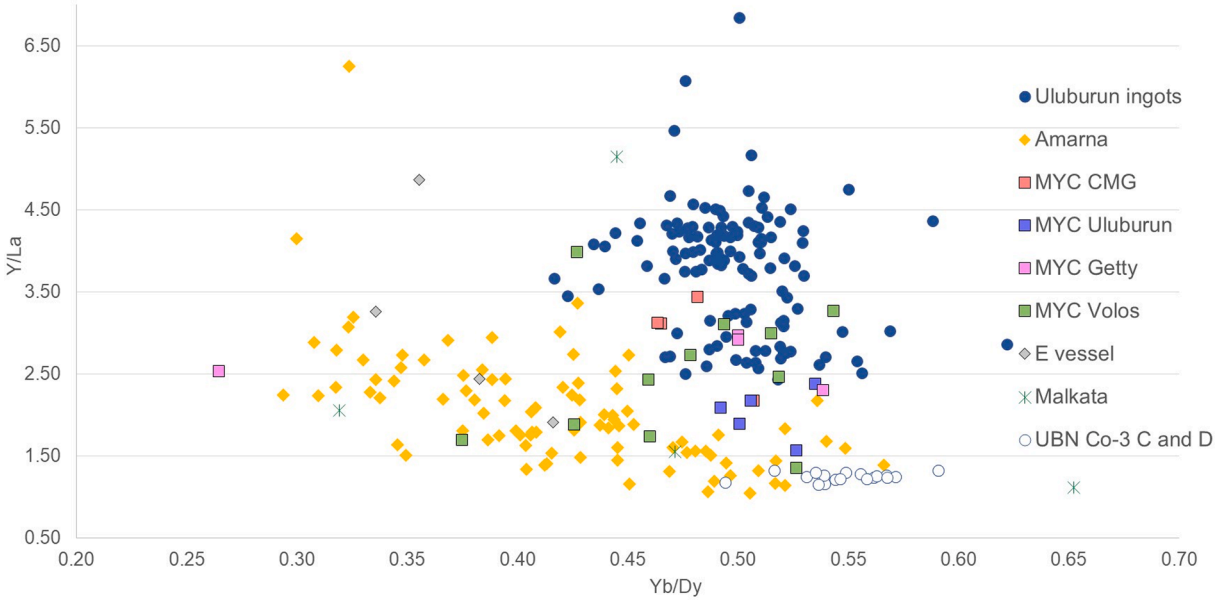


Fig. 12. Y/La vs Yb/Dy for cobalt-blue glass from the Uluburun shipwreck, including batches Co-3C and D, Amarna, Malkata, four Egyptian vessel fragments in museum collections, and Mycenaean (MYC) relief beads from the Corning Museum of Glass, the Uluburun shipwreck, a Greek tholos tomb at Volos (Smirniou et al., 2012), and the Getty Museum (Walton et al., 2009). Amarna samples Lankton et al. (2022a) or Hodgkinson and Frick (2020); all Mycenaean samples analyzed in our laboratory except those from Walton et al. (2009). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Mycenaean relief beads on the Uluburun ship were made from glass either intermediate or closer to that from Amarna. If these Mycenaean beads had been made in the same center as the ship's intended destination, then previous shipments would have included glass from Amarna. At the same time, the three Corning Museum of Glass relief beads that were a close match to the Uluburun glass ingots indicate that the Uluburun ship was not the only one to carry glass from this Egyptian production site(s) to Mycenaean glass relief bead production centers. The comparisons in Fig. 12 confirm this scenario with the surprising additional finding that over half of the Mycenaean relief beads for which we could find REE analyses were closer to the Uluburun ingots than to glass from Amarna. This evidence suggests a change in cobalt supply that may reflect post-Amarna production at a new location using very similar raw materials and technologies and possibly the same workers, or perhaps continued production at Amarna but in a location of the site not yet explored.

5. Conclusions

In order to better understand some of the technological processes in the production of the Uluburun glass ingots we examined the Cu and Co used to color the glass ingots. In doing so, we found the first compositional evidence for the addition of waste glass or cullet during Egyptian LBA glass manufacture, as suggested by glass recipes in Mesopotamian

texts thought to derive from the LBA. Based on variations in the levels of Sb and Cu, along with their associated trace elements, we conclude that the use of cullet is the only possible explanation for the observed Sb levels in the Uluburun copper- and cobalt-blue ingots and the most satisfactory explanation for the variable Cu levels found in cobalt-blue glass. While the categories of Co– or CoCu-blue may continue to be useful in some cases, we suggest that the Cu distribution is more likely to be continuous and results from the addition of copper-blue glass imparting not only Cu but also variable levels of Sn and As.

Based on radiocarbon dating, it is likely that the Uluburun ship sailed at the very end of or just after the abandonment of the royal capital at Amarna, the only known primary production site for Egyptian glass prior to the Ramesside period. Thus, it is important to understand the relationship between the Uluburun glass ingots and glass thought to have been made at Amarna. Based on multivariate statistical evidence (Lankton et al., 2022a) further explored here, it seems possible that some of the glass from the Uluburun shipwreck was made at Amarna, particularly the amber ingot, the copper-blue ingots from batch Cu-4, and possibly the cobalt-blue ingots in batches Co-3C and D. However, most of the ingots have little compositional similarity to glass from Amarna, making production there, at least in any part of the site so far explored, unlikely. On the other hand, our study of the possible compositions of the cullet added during the production of the ingots suggest that, at least for some of the ingot groups, even if the ingots were made elsewhere, Amarna-type glass was still available for recycling purposes.

Finally, the molded glass relief beads made at Mycenaean centers using Egyptian glass, most likely imported as ingots similar to those on the Uluburun shipwreck, provide unique insight into changes in glass supply during the late 18th Dynasty. Of the 10 relief beads included in our multivariate statistical analysis, one bead, of copper-blue color, was made with Mesopotamian glass and the other nine, all cobalt blue, with Egyptian glass. While the compositions of the relief beads from the Uluburun shipwreck are more similar to glass from Amarna or to a mixture of Amarna and Uluburun-type glass, those from the Corning Museum of Glass are closer to Uluburun ingot glass. This indicates that other shipments with glass ingots similar to those onboard the ship had reached Mycenaean centers either before or after the Uluburun ship sank.

Further, we observed that the cobalt colorant used at Amarna differed in REE patterns from the cobalt used to color the ingots and achieved good separation using two ratios based on the proportions of light and heavy REE. Applying the same criteria to 15 previously published cobalt-blue Mycenaean relief beads for which REE measurements are available, we found that, while some are consistent with an Amarna origin, more than half, including examples from excavation and museum collections, were colored with cobalt not found at Amarna but more like that used for the Uluburun ingots. We interpret these results to suggest that the Uluburun glass ingots were not the result of a one-off event but rather represent the products of a different glassmaking center that would have followed the main occupation at Amarna, becoming the major source for Egyptian glass shipments to the Aegean. These two Egyptian glassmaking sites were closely related in terms of raw materials and technologies. For example, both used the same types of grinding stones to process the glass powder and similar molds to shape the finished ingots. However, subtle changes in such things as the type of cobalt colorant available may allow us to distinguish separate productions.

For now, the absolute number of Mycenaean glass analyses may be too low to reach firm conclusions, but if further studies support the pattern observed here, this may be the first evidence for an important shift of glass production away from Amarna. Where the new site might be is not yet known, but preliminary accounts of the recent discovery of 'the Dazzling Aten,' a city adjacent to the Malkata palace founded by Amenhotep III but thought to have been used later by both Tutankhamun and Ay, mention not only residential neighborhoods but also those for metal and glass production (Blakemore, 2021). While there is evidence from the Malkata palace for secondary glassworking to produce beads, inlays, and vessels (Mass et al., 2002), primary production has not been identified, although based on our analyses of 14 samples, the Malkata glass seems compositionally similar to that found at Amarna. If evidence for primary glass production is found in the new excavations at Aten, it will be very interesting to see if the glass made there is more like that from Amarna and the Malkata palace workshops, or like that of the glass ingots from the Uluburun shipwreck.

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.

Data availability

All data for this research included in Table S1

Acknowledgements

We would like to thank Yaşar Yıldız, former director, and the staff of the Bodrum Museum of Underwater Archaeology for their original assistance in Bodrum. Karol Wight, Stephen Koob, and Katherine Larson of the Corning Museum of Glass; Geralda Jurriaans-Helle and René van Beek of the Allard Pierson Museum of the University of Amsterdam; and Joan Oates, University of Cambridge, were instrumental in granting access to most of the non-Uluburun LBA samples referred to in this study. Special thanks also to the two anonymous reviewers whose helpful suggestions improved the quality of our manuscript. Without the support of the Institute of Nautical Archaeology (INA) at Texas A&M University for post-excavation conservation, scientific analysis, and study of the Uluburun shipwreck artifacts this work would not have been possible. Additional major financial support was generously provided by multiple grants awarded by the Institute for Aegean Prehistory (INSTAP).

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jasrep.2022.103596.

References

Abe, Y., Harimoto, R., Kikugawa, T., Yazawa, K., Nishisaka, A., Kawai, N., Yoshimura, S., Nakai, I., 2012. Transition in the use of cobalt-blue colorant in the New Kingdom of Egypt. J. Archaeol. Sci. 39, 1793–1808.

Aitchison, J., 1999. Logratios and natural laws in compositional data analysis. Math. Geol. 31, 563–580.

Ayora, C., Carrero, S., Bellés, J., Basallote, M.-D., Cánovas, C.R., Macías, F., 2021. Partition of rare earth elements between sulfate salts formed by the evaporation of acid mine drainage. Mine Water Environ. https://doi.org/10.1007/s10230-021-00803-0.

Blakemore, E., 2021. 'Lost golden city of Luxor' discovered by archaeologists in Egypt. National Geographic, April 8, 2021. https://www.nationalgeographic.com/history/article/lost-golden-city-luxor-discovered-archaeologists-egypt.

Brill, R.H., 1976. Notebook on the Tutankhamun treasures. Corning Museum of Glass, https://blog.cmog.org/2012/04/03/glass-of-king-tut-dr-brills-research-on-the-ancient-egyptian-glass-from-tutankhamuns-tomb/ (accessed 5 March 2022).

Deng, W., Wright, R., Boden-Hook, C., Bingham, P.A., 2019. Melting behavior of waste glass cullet briquettes in soda-lime-silica container glass batch. Int. J. Appl. Glass Sci. 10, 125–137.
El-Taher, A., 2007. Rare-earth elements in Egyptian granite by instrumental neutron activation analysis. Appl. Radiat. Isot. 65 (4), 458–464.
Hodgkinson, A.K., 2015. Archaeological excavation of a bead workshop in the main city at Tell el-Amarna. J. Glass Stud. 57, 279–284.
Hodgkinson, A.K., Frick, D.A., 2020. Identification of cobalt-coloured Egyptian glass objects by LA-ICP-MS: A case study from the 18th Dynasty workshops at Amarna, Egypt. Mediterranean Archaeol. Archaeometry 20 (1), 45–57.
Kamber, B.S., Greig, A., Collerson, K.D., 2005. A new estimate for the composition of weathered young upper continental crust from alluvial sediments, Queensland, Australia. Geochim. Cosmochim. Acta 69, 1041–1058.
Kaszmarszyk, A., 1986. The source of cobalt in ancient Egyptian pigments. In: Olin, J., Blackman, M. (Eds.), Proceedings of the 24th International Archaeometry Symposium. Smithsonian Institution, Washington, D.C., pp. 369–376.
Lankton, J.W., Pulak, C., Gratuze, B., 2022a. Glass ingots from the Uluburun shipwreck: Glass by the batch in the Late Bronze Age. J. Archaeol. Sci.: Rep. 42, April 2022, 103354.
Manning, S.W., Pulak, C., Kromer, B., Talamo, S., Bronk Ramsey, C., Dee, M., 2009. Absolute age of the Uluburun shipwreck: A key Late Bronze Age time-capsule for the East Mediterranean. In: Manning, S.W., Bruce, M.J. (Eds.), Tree-rings, Kings, and Old World Archaeology and Environment: Papers Presented in Honor of Peter Ian Kuniholm. Oxbow Books, Oxford, pp. 163–187.
Mass, J.L., Wypyski, M.T., Stone, R.E., 2002. Malkata and Lisht glassmaking technologies: Towards a specific link between second millennium BC metallurgists and glassmakers. Archaeometry 44 (1), 67–82.
Oppenheim, A.L., Brill, R.H., Barag, D., von Saldern, A., 1970. Glass and Glassmaking in Ancient Mesopotamia. Corning Museum of Glass, Corning, New York.
Pulak, C., 2008. The Uluburun shipwreck and Late Bronze Age trade. In: Aruz, J., Benzel, K., Evans, J.M. (Eds.), Beyond Babylon. Art, Trade and Diplomacy in the Second Millennium B.C. The Metropolitan Museum of Art, New York, and Yale University Press, New Haven and London, pp. 289-385.
Reade, W., 2021. The First Thousand Years of Glass-Making in the Ancient Near East: Compositional Analyses of Late Bronze and Iron Age Glasses. Archaeopress, Oxford.
Rehren, T., 2016. Another order for glass - or: how much glass does Pharaoh need? In: Franzmeier, H., Rehren, T., Schulz, R. (Eds.), Mit archäologischen Schichten Geschichte schreiben, Festschrift für Edgar B. Pusch zum 70. Geburtstag. Gerstenberg-Verlag, Hildesheim, pp. 257-267.
Schreurs, J.W.H., Brill, R.H., 1984. Iron and sulfur related colors in ancient glasses. Archaeometry 26 (2), 199–209.
Shaw, I. (Ed.), 2000. The Oxford History of Ancient Egypt. Oxford Univ. Press, Oxford.
Shortland, A.J., Eremin, K., 2006. The analysis of second millennium glass from Egypt and Mesopotamia, part 1: New WDS analyses. Archaeometry 48 (4), 581–603.
Shortland, A., Rogers, N., Eremin, K., 2007. Trace element discriminants between Egyptian and Mesopotamian Late Bronze Age glasses. J. Archaeol. Sci. 34, 781–789.
Shortland, A.J., Tite, M.S., Ewart, I., 2006. Ancient exploitation and use of cobalt alums from the Western Oases of Egypt. Archaeometry 48 (1), 153–168.
Smirniou, M., Rehren, T., 2011. Direct evidence of primary glass production in Late Bronze Age Amarna, Egypt. Archaeometry 53, 58–80.
Smirniou, M., Rehren, T., Adrymi-Sismani, V., Asderaki, E., Gratuze, B., 2012. Mycenaean beads from Kazanaki, Volos: A further node in the LBA glass network. In: Ignatiadou, D., Antonaras, A. (Eds.), Annales du 18e Congrès de l'Association Internationale pour l'Histoire du Verre. AIHV, Thessaloniki, pp. 11–18.
Smirniou, M., Rehren, T., 2013. Shades of blue — cobalt-copper coloured blue glass from New Kingdom Egypt and the Mycenaean World: a matter of production or colorant source? J. Archaeol. Sci. 40 (12), 4731–4743.
Varberg, J., Gratuze, B., Kaul, F., 2015. Between Egypt, Mesopotamia and Scandinavia: Late Bronze Age glass beads found in Denmark. J. Archaeol. Sci. 64, 168–181.
Varberg, J., Gratuze, B., Kaul, F., Hansen, A.H., Rotea, M., Wittenberger, M., 2016. Mesopotamian glass from Late Bronze Age Egypt, Romania, Germany and Denmark. J. Archaeol. Sci. 74, 184–194.
Walton, M.S., Shortland, A., Kirk, S., Degryse, P., 2009. Evidence for the trade of Mesopotamian and Egyptian glass to Mycenaean Greece. J. Archaeol. Sci. 36, 1496–1503.
Wedepohl, K.H., 1995. The composition of the continental crust. Geochim. Cosmochim. Acta 59, 1217–1232.
Weinstein, J.M., 2008. Nefertiti scarab. In: Aruz, J., Benzel, K., Evans, J.M. (Eds.), Beyond Babylon. Art, Trade and Diplomacy in the Second Millennium B.C. The Metropolitan Museum of Art, New York, and Yale University Press, New Haven and London, 358.