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*Review*

# Known Glass Compositions in Iron Age Europe—Current Synthesis and Emerging Questions

Valentina Lončarić and Mafalda Costa *

HERCULES Laboratory, University of Évora, Palácio do Vimoso, Largo Marquês de Marialva 8,
7000-809 Évora, Portugal; vloncaric@uevora.pt
* Correspondence: mcosta@uevora.pt

**Abstract:** Ancient glass has been extensively studied from a technological and raw material provenance perspective since the middle of the 20th century. With the rising applications of analytical techniques in the field of Heritage Studies, the last two decades saw an exponential increase in publications on ancient and historical glass technology from around the globe. Given the amount of works on glass chemical composition, it is surprising to note that the long-held production model for Iron Age glasses found in Europe has only recently been challenged by the publication of uncharacteristic glass compositions. Traditionally, LBA glass industries based on plant-ash fluxes/HMG (Egypt and the Levant) and mixed-alkali fluxes/LMHK (Italy) are thought to be supplanted by natron-fluxed/LMG production operating in Egypt and the Levant since around the 9th century BCE. Recently, however, arguments have been put forth for a more diversified network of glassmaking traditions, including small-scale autonomous European workshops. This article reviews the current state of research into Iron Age (1st millennium BCE) glasses in Europe by examining the available published data on glass compositions to critically assess some practical and theoretical issues stemming from this heterogenous field of research. Key questions are addressed, and future lanes of research are proposed.

**Keywords:** Iron Age; glass technology; glass chemical composition; trade and exchange networks; provenance studies

## 1. Introduction

The period of the Iron Age in Europe—lasting roughly throughout the 1st millennium BCE—is a dynamic period marked by cultural, political, and technological heterogeneity. In fact, because the archaeological convention for the local onset of the Iron Age is the replacement of bronze tools by their iron and steel equivalents, there are significant chronological differences between the north and the south and east of Europe [1]. Similarly, the period is conventionally thought to end with the advent of historiography or Roman conquests, which in some cases means that the Iron Age lasted until the migration period. In fact, continental and Mediterranean Europe are often considered separate areas of study because the social and technological changes following the collapse of the established power structures at the end of the Late Bronze Age [2] affected the two regions differently. The exact timing of the LBA/IA transition varies by geography and the associated cultural entities, but the start of the transition can be traced back to the multifactorial crisis of the 13th and 12th centuries BCE, which resulted in the decline of prospering Bronze Age palatial economies in the Eastern Mediterranean, for example, the Mycenaean civilisation in the Greek mainland, the Minoan civilisation on the Aegean islands, and the Hittite kingdom in Anatolia [2]. The power vacuum resulting from the decline of major Bronze Age political centres, coupled with emergent ironworking technology, and related technological innovations, allowed for the rise of new political entities, the reconfiguration of old trade and exchange networks, and colonial expansion [3,4]. In continental Europe, the Early Iron Age is largely equivalent

with the Hallstatt cultural complex and the associated chronological system, spanning most of central Europe. The system itself is not uniform, but generally, it encompasses the period from ca. 800 BCE to 450 BCE. In the Mediterranean, this encompasses the Iron Age and Classical Antiquity periods, which saw Greek and Phoenician colonial expansion to the west of the Mediterranean, Greco-Persian wars, the founding and rise of Carthage in the western Mediterranean, the expansion of the Etruscan civilisation in the Apennine peninsula, and the founding of the Roman Republic, to name the most prominent historical events [5]. The second half of the 1st millennium BCE in continental Europe was marked by a social restructuring around urban centres, which enabled firmer control of territory and economic activities such as trade or craft organization [1,6–8]. Locally produced forms of material culture, referred to as either "Celtic" or belonging to the La Tène cultural complex, appearing from the middle of the 5th c. BCE, can be considered a reaction to the aesthetic and artefact repertoire of the Mediterraneanised world, which had formed some centuries earlier [3,5] and which ultimately enabled the Roman empire to emerge as the political victor. Because of this chronological and cultural ambiguity, adopting a network perspective facilitates navigating the large-scale technological, political, and economic processes taking place in the Mediterranean and Europe during the 1st millennium BCE [5,9]. Iron Age glass is usually interpreted as evidence of ancient trade, and is, therefore, a suitable subject for study, especially given the possibility of discerning the raw material sources with the help of geochemical analyses such as trace element or radiogenic isotope analyses [10–12]. Artefacts such as beads, vessels, pendants, bracelets, and glass-decorated fibulae dated to the Iron Age are known across Europe, with differing chronological and geographical distributions. Morphologically simple objects, such as monochrome glass beads or polychrome eye-beads, are not highly informative in terms of chronology or the possible location of secondary workshops, since similar decorative schemes were in use throughout the whole 1st millennium BCE [13–15]. On the other hand, some regionally, chronologically, or contextually limited forms—for example, "black" glass beads from funerary sites in Southern Portugal [16], glass-bow fibulae (*Bügelfibeln*) from the Apennine peninsula [17], Hellenistic core-formed vessels [18–20], ram-head pendants in Slovenia [21], Celtic bracelets [22–25], and amphorae-shaped beads [26]—serve to show that glass was not merely an exotic import, but also a material that was actively manipulated to fulfil the needs of various populations during the 1st millennium BCE.

The purpose of this review is to inform researchers of the sciences and humanities alike about the current state of research into Iron Age glass compositions, and to encourage stronger interdisciplinary dialogue. The article is structured around a geochemical understanding of Iron Age glass, because approaching glass (or any other artefact category) from a material perspective can allow for the more ephemeral phenomena, which dictated the changes in glassmaking practices, glass trade networks, artefact manufacture, and consumption practices, to be explored from an objective viewpoint. In other words, Iron Age Europe was a culturally heterogenous area and period, the current knowledge of which is heavily influenced by historically different research approaches across European countries. Having the basic knowledge of a certain type of material—in this case, glass—provides a solid basis for addressing external and internal factors that shape cultural processes, even including highly complex ones, such as identity formation or skill transfer [7,16,27]. This is because glassmaking implies that certain technical requirements, such as procuring suitable raw materials or adequate mastery of pyrotechnology, need to be met regardless of the period or cultural ascription. Glassmaking sites, primary production sites, and primary workshops are workshops equipped to produce glass from raw materials, the geochemical characteristics of which are reflected in the glass. Scientific analyses of glass composition aim to understand glassmaking recipes and raw material provenance, and are, therefore, more focused on primary workshop activity. Secondary (tertiary, etc.) production sites/workshops or glassworking sites are places of artefact manufacturing and/or raw glass modification, e.g., colouring. A single workshop could, in theory, be both a glassmaking and a glassworking site, which sometimes complicates the distinction, and this

cannot always be inferred from glass composition alone. Interpretations based on typo-chronological classification systems or find contexts allow the results of scientific analyses to be placed within the scope of knowledge of specific archaeological populations. As such, a humanities/archaeology-based approach can help to differentiate between different glassworking sites on the basis of artefact morphology or distribution. Integrating the two approaches enables researchers to understand communities of Iron Age Europe as active participants on the economic stage of the 1st millennium BCE.

This review is structured around the results of the last two decades of research into the chemical compositions of Iron Age glass artefacts found in Europe. The most commonly used techniques for glass analysis in the last two decades include X-ray fluorescence spectrometry (XRF), scanning electron microscopy with energy/wavelength dispersive X-ray spectrometry (SEM-EDS/WDS), electron microprobe analysis (EMPA), and inductively coupled plasma mass spectrometry (ICP-MS) (Table 1). Each of these techniques has advantages and disadvantages depending on the exact configuration, including spatial resolution and sample preparation requirements. Detailed explanations of the physical principles behind each of the techniques are outside the scope of this review; however, some specifics that should be kept in mind when assessing glass chemical compositions must be pointed out. It is important to keep in mind that the mentioned techniques differ greatly in their detection limits (LOD), meaning that some, such as SEM-EDS or EMPA, will not provide insight into the trace element compositions used for raw material provenance studies. Furthermore, SEM-EDS and EMPA usually rely on destructive sample preparation techniques, including polishing and coating the samples. Variable pressure (VP) SEM-EDS setups allow for completely non-destructive analysis, but the obtained results can be greatly influenced by glass degradation processes such as alkali leaching or environmental contamination of the glass surface. Studies using handheld or portable XRF instruments, on the other hand, have raised concerns regarding variations in their accuracy and precision, and the influence of matrix effects on measured elements is known to complicate the calibration procedures required for element quantification. Moreover, in the last few decades, ICP-MS has increasingly become a popular technique for major and trace element analysis. The addition of laser ablation systems (LA) for solid sample introduction enables minimally invasive artefact sampling. Compared to traditional ICP-MS techniques relying on wet (digested) samples, LA-IC-MS has somewhat lower measurement reproducibility, but this disadvantage is offset by the fact that LA-ICP-MS results only in microscopic damage to the sample, making it a preferred technique for archaeological objects.

**Table 1.** Summary of practical considerations for some analytical techniques usually employed for glass studies.

| Technique | Element Range | LOD | Main Disadvantages | Advantages |
|---|---|---|---|---|
| handheld/portable XRF | Z > 12 (Mg) | element-specific, down to trace concentrations | complicated calibration procedures, frequent element over/underrepresentation | fast analysis times, non-destructive |
| LA-ICP-MS | All (except H, He, N, O, Ne and Ar) | trace concentrations, down to ppm, ppb, or ppt, depending on configuration | lower measurement reproducibility than wet sample ICP-MS | minimally invasive, fast, high spatial resolution |
| SEM-EDS | Be-U | 0.1 wt. % | surface-sensitive, influenced by surface leaching and contamination for non-polished samples | high spatial resolution, non-destructive depending on sample preparation |
| EMPA | Z < 3 (Li) | down to 100 ppm | surface-sensitive, requires sample polishing and coating | high spatial resolution |

Firstly, a short summary of glassmaking components and their indicative elements is given. Then, a broad overview of the key differences between Bronze Age and Iron Age glassmaking traditions is presented to provide context to the main section, which places the growing body of evidence for the pluricentric organisation of glassmaking in the

Early Iron Age and the subsequent increasing homogeneity of glassmaking recipes into historical perspective. The closing sections are dedicated to addressing the outstanding issues regarding research on Iron Age glass.

## 2. Contributions of Glassmaking Components to the Final Chemical Composition of Archaeological Glass

The role of various glassmaking components on the chemical and physical properties of glass have been discussed in detail in the previous literature [11,12,20]. Herein, we only provide a general summary of the most common raw materials used to make ancient silicate glass, as well as their elemental contributions to the bulk glass composition.

Raw materials used to make ancient silicate glass can be divided into four main components: 1. the silica source, or the network former; 2. the fluxing agents, or network modifiers used to lower the melting temperature of the glass batch; 3. network stabilisers, which offset the damaging effect of network modifiers, either naturally present in one of the previous components or specifically added to the batch; and 4. (de)colouring and opacifying agents which may be either naturally present in one or more glassmaking components or intentionally added.

### 2.1. Silica Sources

In nature, silica is most readily available either as high-purity quartz or quartz-bearing sand. Both are commonly encountered, and it is generally accepted that they are not transported over great distances. Therefore, glass provenance studies rely on the assumption that the quartz source used as the network former was extracted relatively close to the glassmaking site, as opposed to more geographically restricted components, such as natron or (de)colourants, which would have had to be transported to the glassmaking site, sometimes from rather great distances. The first man-made glasses were fused using quartz pebbles and plant ash as the two main ingredients. In its pure form, quartz ($SiO_2$) does not contain many accessory elements. For glasses made from fairly pure quartz sources, the trace elements Ti, Zr, Cr, and La, as well as combined Sr/Nd isotope analyses, have been used as discriminants between those of Egyptian and Mesopotamian origin [28–33]. In the case of glasses made from pure quartz sources and plant ash, Shortland et al. [29] pointed out that non-colourant-related elements such as Ti, La, and Cr could potentially have been introduced as contaminants, for example, as clays introduced into the glass batch with plant ashes, or as environmental contaminations during quartz processing. On the other hand, higher Zr values in Egyptian glass derive from the quartz source itself, which commonly contains zircon as an accessory mineral [29].

Quartz sand can contain various accessory minerals that reflect its geological history, such as feldspars, clay minerals, iron and titanium oxides, carbonates, chromite, and zircon, among others, which are reflected in the minor-to-trace-element composition of glass [11,12,28]. The relationship between sand minerals and glass composition is generally well-understood for glass fluxed with natron [11,12,28]: feldspars and clays contribute significant amounts of Fe, Ti, and Al; trace elements such as Sc, V, and Cr are related to the presence of iron oxides in the sand; Zr and Hf reflect the presence of zircon; Ti is characteristic of rutile (along with Nb and Ta), ilmenite, and titanite; and Th and rare earth elements can be found in monazite and other phosphates, which also contribute P to the final glass. The most common way of differentiating sand sources is by using the $Al_2O_3$, $TiO_2$, $Fe_2O_3$, and $SiO_2$ contents of analysed glass as indicators of sand purity. Differences in sand purity are generally thought to reflect the use of different sands, although it bears pointing out that this can include both different source locations and different processing methods of the same source. Furthermore, aluminium and iron contents are also known to be influenced by the colouring technology. For example, the addition of cobaltiferous alum to produce blue glass results in elevated $Al_2O_3$ values [34–37]. This case is well-illustrated in 8th c. BCE blue beads from Sarno and Capua (Italy) [37], which contain high amounts of $Al_2O_3$ (avg. 6.3 wt. %), MgO (avg. 3.8 wt. %), and Co (avg. 803 ppm) and exhibit no Ti-Al

correlation typical of glasses made by fusing quartz sand. Here, the contribution of $Al_2O_3$ in the final composition was interpreted as coming from a glassmaking component other than sand, most likely the colourant [37]. Similarly, iron in various concentrations has been linked to a range of colours, spanning from amber and green to brown and "black" [37–40], because it is naturally present in sand and can be heavily concentrated in, for example Egyptian black sands [38]. When discussing Iron Age "black" glass from the territories of Italy, Slovakia, and Poland, both the use of iron-rich sands and the addition of iron slags have been offered as explanations for the elevated iron content [38,39]. The mentioned overlap between sand and colourant contributions to the final glass illustrates how a minor oxide concentration cannot always be taken as a definitive indicator of primary production sites where glass is fused. Rather, CaO, $Al_2O_3$, and FeO should be taken to reflect different glassmaking recipes, possibly those used in the same glassmaking region. Overall, Zr values, often plotted against Sr values, have shown to be a robust marker of Egyptian vs. Levantine primary production sites, for glass made with both sand and pure quartz [29–31,33,37,38,40–42]. Of the two, Zr is more directly related to the silica source, while Sr can reflect the Ca-bearing fraction of the glassmaking sand, purposefully added Ca stabilisers, or—in the case of glass made with pure quartz and plant ash—the Sr content of the ashed plant matter (see Section 2.2). Coastal sands in the Mediterranean contain <50 ppm Zr and >300 ppm Sr [43], and these criteria are taken as markers of a Levantine origin for natron glass, while the reverse relationship is usually attributed to Egyptian sand.

## 2.2. Fluxing Agents

For the Bronze and Iron Ages, compounds containing $Na^+$ ions, such as plant ash and natron, were the main fluxing agents. The main difference between these fluxes is the number of accessory elements they introduce into the final glass composition. Plant ash obtained from burning halophytic plants found in semi-desert or saline maritime environments was the earliest fluxing agent used in glassmaking, until it was gradually replaced by natron during the early 1st millennium BCE [12,20]. Plant ash contributes Na, Ca, Mg, K, and P to the final glass composition [12]. As a result, plant ash glasses contain notably higher amounts of Mg and/or K than natron-fluxed glass (see Section 3). Because Ca is naturally present in plant ash, these glasses would not have required an additional stabiliser-bearing ingredient. A trace element associated with Ca in plant ash is Sr. Experimental studies on modern-day plant ashes have shown that ash composition depends on the species of the plant used, the environment in which it was grown, the ashing procedure, and the plant section which was ashed [20,32,44–46]. The Sr isotopic ratio ($^{87}Sr/^{86}Sr$) of plant ash has been found to depend on local geology and hydrology, making it impossible to obtain a high spatial resolution for plant sourcing without a direct comparison or by relying on Sr isotopes alone [30,32,47]. A major shift to the production of natron-fluxed glass occurred at the start of the 1st millennium BCE. Natron is a mixture of minerals found in evaporitic lake deposits rich in sodium carbonates and bicarbonates, the most notable of which is the mineral trona ($Na_2CO_3 \cdot NaHCO_3 \cdot 2H_2O$) [20,48]. Natron is a more predictable and efficient fluxing agent than plant ashes; however, it does not contain stabilising impurities, making it unsuitable for glass made from high-purity quartz sand or quartz pebbles [20,49]. Natron is geographically limited to deposits in the eastern Mediterranean, the best-known being Wadi Natrun in Egypt, but other natural deposits are known in Anatolia, Syria, Macedonia, and North Africa, and may have been exploited in the past [20,50]. Because natron is a highly pure sodium source, its geochemical provenance is difficult to distinguish. Boron isotope ratios ($^{11}B/^{10}B$) have been successfully applied as a proxy for natron sourcing in Roman glass [48,51,52], but this methodology is not routinely implemented. Instead, combined isotope ratios for silica ($^{144}Nd/^{143}Nd$) and stabiliser-related elements ($^{87}Sr/^{86}Sr$) are preferred in ancient glass provenance studies [11,12,38,53].

## 2.3. Stabilisers

Calcium is the usual stabilising agent added to man-made silica glasses to make them more chemically stable. Because plant ashes naturally contain high quantities of both Mg and Ca, which act as stabilisers [12,44], glasses made with a high-purity silica source and plant ash are thought to have been made without the addition of other stabilising components, although the addition of another Ca-bearing component cannot always be excluded [44]. Nonetheless, in plant ash glass, the Ca and Sr contents are usually taken to reflect the ash composition, which, in turn, depends on factors such as the plant species, the part of the plant ashed, and the local geology and hydrology [30,32,44–46,54,55]. On the other hand, in natron-fluxed glass, stabilisers need to be introduced either with the silica source or as another glassmaking component. Limestone and shells can be used as calcium stabiliser sources in sand-fused glass, and could either be naturally present in the sand or intentionally added. The distinction between limestone and shell Ca sources is reflected in the Sr content of glass, since this element is more easily incorporated into aragonite than calcite and is, therefore, expected to be found in higher concentrations if shells and shell-rich coastal sand, rather than limestone, were used for glassmaking [11,12]. A low CaO/SrO ratio (<200) is taken as an indicator of an increased amount of shell fragments in the glassmaking batch, whereas high ratios (>600) indicate the use of limestone [28,54]. Here, care should be taken to differentiate between natron- and plant ash-fluxed glasses beforehand, since, as mentioned previously, the ash flux can contain varying amounts of both Ca and Sr [30,32,44–46,54,55]. For natron-fluxed glass, low (<200 ppm) and high (300–600 ppm) Sr contents in the final glass are often used as indicators of geological limestone or beach shell fragments used as glass stabilisers, respectively [12]. Many researchers have used Sr concentrations to hypothesise the nature of glass stabilisers used in the Early Iron Age [38,40,56] and in Hellenistic [42,56,57] and Celtic glass [7,58,59]. In particular, the relationship between Sr and Zr has proven to be indicative of the use of shell-rich coastal sands as opposed to zircon-rich inland sands/silica sources for glasses from the Bronze Age to the Roman period, and is often used as a discriminating factor between glass produced in the Levant (possibly with Belus river sands) and Egypt [7,29,40–42,58–62]. Some other elements, such as Al and Fe, can also act as stabilisers, and are, in fact, responsible for the preservation of some early natron glasses found in Europe that contain amounts of Ca insufficient to stabilise glass [35,37,38,49]. Iron and alumina are usually naturally present in sand as impurities, but they could also have been added as part of colouring agents, for example in Fe-rich black-appearing glass or blue glass coloured with cobaltiferous alums (see Section 3). However, as evidenced by the later optimisation of the Ca content in natron glass during the 1st millennium BCE, Al and Fe were probably not recognised as stabilisers by ancient glassmakers, but, rather, were introduced with the glassmaking sand or as colourants.

## 2.4. Colourants and Opacifiers

Ancient glass is usually brightly coloured, owing to the presence of transition metal ions, or, for some colours, metallic nanoparticles (Table 2). When present in high concentrations, some colours, such as blue, purple, green, or amber/brown, can appear as black. The addition of crystalline phases—opacifiers—to the glass matrix renders glass opaque while also acting as colourants [63] (Table 2). Commonly used opacifiers during the 1st millennium BCE include calcium and lead antimonates for white and yellow colours, respectively. Generally, colourants are only needed in trace to minor quantities in order to achieve the colouring effect in glass [64,65], but opacifiers and iron can also be present in major quantities [37,38,66] and complicate the understanding of glassmaking recipes.

**Table 2.** Common colourants and opacifiers encountered in 1st millennium BCE glass [20,63–65].

| Colouring Ion/Phase | Hue |
| --- | --- |
| $Mn^{2+}$ | light violet |
| $Mn^{3+}$ | deep purple |
| $Fe^{2+}$ | blue, green in combination with ($Fe^{3+}$) |
| $Fe^{3+}$ | yellow, green in combination with ($Fe^{2+}$) |
| $Co^{2+}$ | blue |
| $Cu^{2+}$ | green/turquoise |
| cuprite ($Cu_2O$) | opaque reds and oranges |
| calcium antimonate ($CaSb_2O_6$/$Ca_2Sb_2O_7$) | white, opacifier for other colours |
| lead antimonate ($Pb_2Sb_2O_7$) | opaque yellow |
| tin-based opacifiers ($SnO_2$) | white, opacifier for other colourants |
| lead-tin-based opacifiers ($PbSnO_3$, $PbSn_{1-x}Si_xO_3$, $Pb_2SnO_4$) | opaque yellow |

On the other hand, tracing the provenance of colourants provides additional depth to the understanding of organisational aspects of glassmaking through the investigation of raw material procurement strategies. Blue cobalt-coloured glass from different periods has received much attention because of the possibility of tracing cobalt sources and understanding the role of cobalt trade in early glassmaking. Cobalt in blue-coloured glass is usually associated with elements such as nickel, zinc, copper, arsenic, manganese, and sulphur, and their relationship with the cobalt content of glass can be used to discern which cobalt sources were used in glass colouring. One of the known Co sources used for glass colouring since the Bronze Age is Egyptian cobaltiferous alum from the Western Oases [34,36], which imparts a characteristic Al-Co correlation to blue glasses coloured with Co from this particular source. Other Co sources are known to have been exploited and can be differentiated by examining the correlations of Co with Ni, Mn, and Zn oxides [20,40,57,67].

Determining the provenance of antimony ores used in glassmaking has also been a research avenue explored in the last decade. A method using Sb isotopes for antimony ore sourcing was developed on Roman glass samples [68] and successfully applied on Late Bronze Age Egyptian and Mesopotamian glass, identifying Caucasus ore deposits as a likely source of stibnite [69,70]. Additionally, the crystal morphology and distribution of opacifiers can be used to differentiate between in situ—characterised by a homogenous distribution of small, euhedral crystals—and ex situ—characterised by the presence of rosary-shaped crystal aggregates—opacification techniques in the glassmaking chaine operatoire [63,71], while the presence of different forms of calcium antimonate ($CaSb_2O_6$ and $Ca_2Sb_2O_7$) was experimentally linked to specific firing regimes. A study of laboratory-produced glass [72] showed that the synthesis of $CaSb_2O_6$ is kinetically favoured, whereas $Ca_2Sb_2O_7$ is more thermodynamically stable, meaning that the exclusive presence of $CaSb_2O_6$ in opaque glasses implies firing temperatures above 1100 °C and firing times shorter than 1 day.

## 3. Key Innovations in Glassmaking between the Bronze and Iron Age

The LBA glass industry in the Eastern Mediterranean and the Levant before the 1st millennium BCE relied on the use of pure quartz and sodium-rich plant ashes, resulting in glass with a high magnesium content (HMG) [20,73,74]. Further west, an independent LBA European glassmaking tradition is considered to have developed from the Early Bronze Age (c. 2100–1700/1650 BC) production of faience in northern Italy [75–77]. Compared to LBA plant ash glass produced in the Eastern Mediterranean, European LBA glasses exhibit a different composition, with low magnesium and increased potash content, which has led them to be labelled as low magnesium–high potassium (LMHK) glass or mixed alkali glass [75]. Investigations of radiogenic isotopes ($^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$) of 11th c. BCE raw and waste glass from the Italian site of Frattesina, coupled with investigations into the composition of north Italian sand and the local plant ash, confirmed local Bronze Age production of LMHK glass in the Po valley, but also suggested the existence of other primary

production sites in Europe [46]. The onset of a new millennium and the start of the Iron Age saw the beginnings of large-scale production of glass fluxed with a mineral source of sodium—natron—which will remain the main type of glass produced in the Mediterranean until the 9th century CE [20,35,37,38,78,79]. When glass artefacts are considered, the LBA/IA transition is seen both in the intensity of glass production and use, and in the changes in glass chemical composition. Considered a prestigious commodity in the LBA, glass objects became increasingly rare during the LBA/IA transition, only to become more common sometime around the 10th–9th century BCE. They appeared first in the Near East and the Eastern Mediterranean, and then spread north and west through Europe [12,20]. Unfortunately, not much glassmaking evidence is available for the transition period. The scarcity of glass artefacts has been interpreted as resulting from the production of chemically unstable glass low in lime rather than a complete hiatus in the glassmaking industry following the end of LBA [80]. Since natron does not naturally contain stabilising ions, they need to be introduced, either with the silica source (as impurities in the sand) or as a separate component (e.g., by adding crushed shells or limestone). Some of the earliest preserved examples of natron-fluxed glass artefacts, such as those from the early 10th century BCE burial of Nesikhons in Thebes (Egypt), various glass beads from 10th century BCE contexts at Pella (Jordan), as well as 9th–8th century BCE glass inlays from Nimrud (Iraq), contain low amounts of Mg, K, and Ca, which is taken as indicative of a pure sodium flux source—natron [74,81]. While natron was quickly adopted as a source of sodium flux, quartz pebbles continued to be used in accordance with the Late Bronze Age tradition documented at Qantir-Pi-Ramesse, Amarna, and Lisht (Egypt) [80,82]. This effectively eliminated Ca stabilisers from the glassmaking recipes, resulting in chemically unstable glass being produced during the 9th–7th centuries BCE, as evidenced by the aforementioned examples from the Eastern Mediterranean and the Near East, as well as from Europe, most notably Italy and France [35,37,38,74,81]. At one point during the 1st millennium BCE, easily available Ca-bearing sand replaced less available quartz pebbles, resulting in the optimisation of glassmaking recipes, but the exact timing is difficult to pin down without archaeological evidence of the primary production sites.

The transition from the Bronze Age plant ash-based glassmaking tradition to natron-fluxed glass is by no means clear-cut, as evidenced by the centuries-long parallel production and circulation of glass fluxed with natron and plant ashes throughout Europe during the early first millennium BCE, most notably in France and Italy [20,35,37,38,75,78,79,83]. In fact, studies of European glass beads and other artefacts in the last decade alone have shown that several different types of glass were in use throughout Europe during the 1st millennium BCE [33,37–39,84]. The subdivision of glass types is based primarily on their MgO and $K_2O$ contents, but other elements, most notably Al and Fe, can be used to define further sub-types. The following glass types have been identified in Iron Age (1st millennium BCE) European assemblages:

- Low-magnesium glass (LMG), which typically exhibits both MgO and $K_2O$ values below 1.5 wt. % [10,78]. Originally, this group was defined by samples of Roman glass, exhibiting 0.73–1.47 wt. % MgO and 0.22–0.63 wt. % $K_2O$ [73], but the upper MgO limit varies depending on the assemblage being studied, with values sometimes reaching 2 wt. % [8]. Here, we consider LMG to have both MgO and $K_2O$ contents of <1.5 wt. %. The fluxing agent used in the production of this type of glass is uniformly thought to be natron [20].
- High magnesium glass (HMG) was initially defined as Bronze Age glass with MgO values in the range of 2.9–4.6 wt. % and $K_2O$ in the range of 0.69–1.89 wt. % [73]. Since then, the range has been reported differently in the literature, broadly including values of MgO > 1.5 wt. % [10,28,78,85] and $K_2O$ values in the range of 1–4 wt. % [20,73,75,78,86]. Because of the broad range of reported values, some researchers prefer the term plant ash glass to HMG and require the MgO content to be higher than that of $K_2O$ [10,78]. This type is considered synonymous with Mediterranean Bronze Age glass production based on a pure silica source and halophytic

plant ashes, although $K_2O$ values as low as 0.5 wt. % have been observed for plant ash glass [32,37]. Typically, glass fluxed with plant ash will exhibit > 5 wt.% CaO, coupled with elevated Sr values [28].

- High magnesium–low potassium (HMLK) glass was first described in detail by Conte et al. [37] on a set of Early Iron Age glass artefacts from Italy. This was defined as an umbrella term covering samples which presented MgO values (ca. 1.5–5 wt. %, avg. 3.3 wt. %) higher than LMG and $K_2O$ values (<2 wt. %, avg. 0.8 wt. %) lower than HMG [37,38]. Although this group has only recently been formally defined, the first mentions of similar Iron Age glass with high MgO and low $K_2O$ content from the territory of Greece, Hungary, former Yugoslavia, and Czechoslovakia can be found in works by Henderson [87], Frána [88] and Braun [89]. There is no firm consensus on which type of flux was used in the production of HMLK glass, with both natron and plant ash being proposed based on the $Al_2O_3$, CaO, and MgO values; this is, in part, related to the glass colour [37]. CaO values in the range of 6.5–8 wt. % were interpreted as resulting from the use of plant ash as the fluxing agent, which also contributed MgO to the final glass composition. Lower CaO levels (<4 wt. %) were found to be consistent with the use of natron and impure sands. Natron-fluxed HMLK glass can further be divided into black-appearing/iron-rich HMLK and Al-Co blue HMLK glass, reflecting the specific raw materials used in the glassmaking recipes.

- Iron-rich HMLK glass contains elevated iron contents (c. 5–13 wt. % FeO), which are responsible for its dark colouration. Rather than being a separate ingredient, this iron is believed to represent the choice of glassmaking sand rich in impurities, or, rather, an intentional choice of an iron-rich sand which served as both the network former and the colouring agent [37,38]. In this case, high MgO and $Al_2O_3$ contents (1.5–5 wt.%) would also have resulted from the choice of impure sands.

- Al-Co blue HMLK glass exhibits high MgO (2.15–4.95 wt. %) and low $K_2O$ (<1 wt. %) [37]. Additionally, this glass type contains high amounts of $Al_2O_3$ (4.36–7.72 wt. %) and lower amounts of CaO (1.29–3.96 wt. %) than typical LMG [37]. The blue colour is derived from the use of cobaltiferous alums, which impart an increased Co content (ca. 250–2000 ppm) and the aforementioned high $Al_2O_3$ and MgO to the finished glass [37,79]. This type has been recognised as one of the earliest natron glasses in France [79] and Italy [37]. Coeval turquoise glass found in Italy with similar compositional characteristics (high MgO and $Al_2O_3$, low CaO, and noticeable Co contents) [83] could be considered as closely related to Al-Co blue glass, and was possibly produced by adapting the recipe to obtain a different hue.

- Low magnesium–medium potassium (LMMK) glass was defined by a set of Early Iron Age glass beads from Poland [33,39]. This type is characterised by MgO values compatible to those of LMG (<1.5 wt. %) and slightly elevated $K_2O$ values (1.4–2.2 wt. %). Further compositional characteristics noticed for LMMK glass include generally high $Al_2O_3$ (1.5–7.8 wt. %) and $Fe_2O_3$ (1.7–5.4 wt. %), as well as a notable presence of metallic inclusions related to the colouring technology [39]. Initially, plant ash was proposed as the fluxing agent used in the production of LMMK glass by Purowski et al. [39], but other authors argued for natron as the fluxing agent in LMMK glass from Poland and similar glass from Italy, with the heightened $K_2O$ content being ascribed to the use of impure sands [37].

- Low magnesium–high potassium (LMHK) glass is considered a relic of the European LBA glassmaking tradition, relying on the use of different plant ashes, contrary to the methods used in the Eastern Mediterranean. This glass is thought to have been fluxed with leached plant ash [20,75–77,87]. The type is characterised by low MgO (<1 wt. %) and high $K_2O$ (4–6 wt. %) content coupled with high $Na_2O$ content, leading to this type of glass also being called mixed alkali glass [10,87].

- "Early wood ash glass" presents a composition similar to that of medieval European early wood ash glass [55], with very high $K_2O$ (5.3–12.6 wt. %), high MgO (2–5.3 wt. %), and low $Na_2O$ (<1 wt. %) contents [38]. This type of glass has, so far,

only been identified in a single assemblage from Chotin (Slovakia), and is interpreted as evidence of small-scale local European glass production in the Iron Age [38].

The geographical and chronological distribution (Table 3) of these types will be explained in more detail in the following sections. However, regarding the links between the glass types according to their chemical compositions, it should be noted here that MgO and $K_2O$ content alone fail to provide unequivocal evidence for fluxing agents used during the Iron Age. This is because, in glasses made with quartz sand, the concentration of both of these oxides reflects the sand accessory minerals [12,37]. As a result, natron-fluxed glass made from impure sands rich in Mg- or K-bearing minerals (e.g., pyroxenes, feldspars) could exhibit MgO and/or $K_2O$ values evocative of plant ash glass. This results in a partial discrepancy between the concepts of glass types and glassmaking recipes because, while both are dependent on each other, the former focuses on the final composition and the latter considers the raw materials used.

**Table 3.** Summary of general trends for the glass types of the 1st millennium BCE found in Europe, as discussed in the text.

| Glass type | Chemical Composition (wt. %) | | | | | Additional Significant Compositional Characteristics | Flux | Date | Distribution (Europe) | References |
|---|---|---|---|---|---|---|---|---|---|---|
| | Na$_2$O | MgO | K$_2$O | CaO | Al$_2$O$_3$ | | | | | |
| LMG | c. 10–20 | <1.5 | <1.5 | c. 4–10 | usually < 4 | | natron | 8th c. BCE onwards | across Europe | [7,10,27,31,33,37,39–42,56–59,66,78,79,84,89–102] |
| LMG (low Ca) | c. 10–20 | <1.5 | <1.5 | <4 | c. 0.4–4.3 | | natron | 9th–7th c. BCE, rare post 6th c. BCE | Italy, Poland, Slovenia, Spain | [31,33,37–39,41,83] |
| HMG | c. 10–20 | >1.5, >K$_2$O | >1.5, <MgO | >5 | usually < 2, up to 10 | | plant ash | LBA-7th c. BCE, sporadic finds post-6th c. BCE | Pre-7th c. BCE. Italy, Poland, Slovenia; post-6th c. BCE: Georgia, Bulgaria, Poland | [31,33,37,39,58,91] |
| LMHK | 4–8 | <1 | 4–6 | <4 | 1–10 | | plant ash | 9th–6th c. BCE | Italy, Slovenia | [31,83] |
| LMMK | c. 13–19 | <1.5 | 1.4–2.2 | <3 | 1.5–7.8 | increased FeO content (c. 2–5 wt. %) | plant ash/natron | | Poland | [39,103] |
| HMLK ("black") | c. 10–20 | c. 1.5–5 | <2 | c. 1.2–6 | 1–4.8 | high FeO content (c. 5–13 wt. %) | natron | 9th–6th c. BCE | Italy, Hungary, former Yugoslavia & Czechoslovakia | [37,38,86,87] |
| HMLK (colourless and blue) | c. 16–19 | c. 1.5–5 | <2 | 6.5–8.5 | <1 | | plant-ash | 9th–6th c. BCE | Italy, Hungary, former Yugoslavia & Czechoslovakia | [37,86,87] |
| Al-Co blue (HMLK) | c. 15–22 | c. 2–5 | <1 | <4 | c. 4–8 | noticeable Co content | natron | 9th–6th c. BCE | Italy, France | [37,79,83] |
| "Early wood-ash" | <1 | 2–5.3 | 5.3–12.6 | c. 5.5–10 | 1.3–3.7 | High FeO content (c. 13–16 wt. %) | wood ash | 7th–5th c. BCE | Slovakia | [38] |

Nevertheless, by the 6th century BCE, the Eastern Mediterranean, with primary production centres somewhere in Egypt and the Levant, was considered the dominant producer of Iron Age glass [20,90], owing this apparent monopoly to readily available sources of natron flux and sand as well as centralised government systems. Evidence of ancient primary glassmaking sites is extremely rare [82,104] and no sites dating to the Iron Age are known, making archaeometric analyses essential in understanding the provenance of Iron Age glass. To this end, trace element (e.g., Zr, Sr, Mn, Ti) concentrations are most often used to discern between Egyptian and Levantine sands as sources of silica for glasses made in the Eastern Mediterranean, and by extension, in primary glass production centres [7,20,37,38,40–43,59,83,91,92].

## 4. Iron Age European Glass Compositions as Indicators of Changing Trends in the Economic Structures of the 1st Millennium BCE

### 4.1. The Plurality of Recipes (ca. 9th–7th c. BCE)

As stated previously, the initial period of Early Iron Age is characterised by parallel production of natron-fluxed glass (LMG) from the Eastern Mediterranean, as well as several unique glassmaking types (Figure 1). Chemical analyses of glass from the 9th–7th centuries BCE were extensively conducted on material from France [35,78,79] and Italy [37,38,83,86], and, to a lesser degree, Poland [33,39], Slovenia [31], and Slovakia [38]. Glass types found during this period can be characterised as LMG, LMHK, HMG, HMLK, and Al-Co blue glass. A common characteristic of glass dating from this period is that there is no clear link between the glass type as defined by the alkali content and the fluxing agent used. Many glass samples from this period, regardless of the glass type, contain lower levels of lime

relative to other alkali and earth alkali oxides, making them appear as an intermediary group between natron soda-lime glass and plant ash soda-lime glass (Figure 1a).
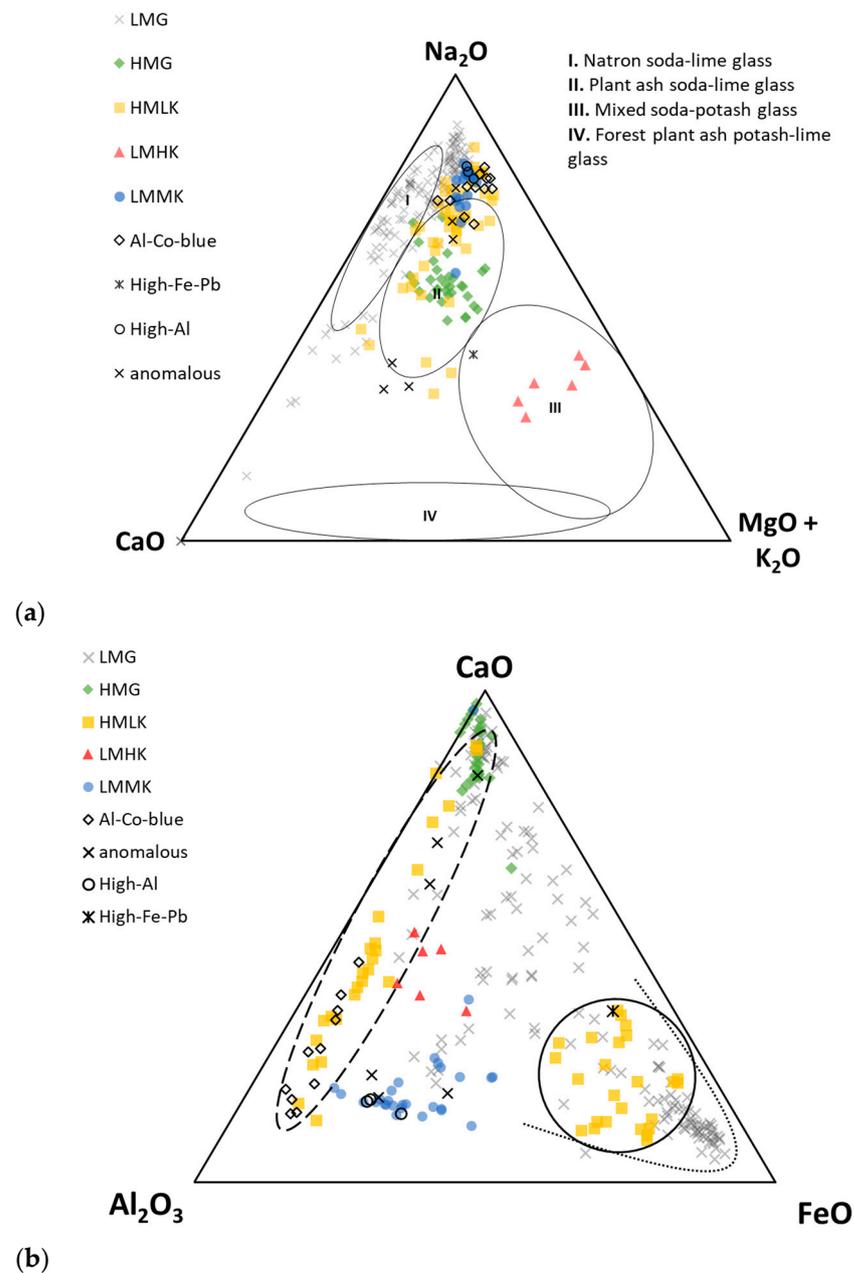


(**a**)



(**b**)

**Figure 1.** (**a**) CaO-Na$_2$O-MgO+K$_2$O ternary diagram of 9th–7th century BCE glass from Europe, showing the relationship between Ca stabilisers and fluxing agents. Leaching of alkali accounts for high CaO outliers. Data were obtained from various publications [31,33,37–39,83,86]. Glass family ranges were obtained from [10]. Glass types were assigned according to MgO and K$_2$O content. Al-Co blue, High-Fe-Pb, and High-Al glass types from [37] and anomalous glass type from [86], were left as in original publications. (**b**) Al$_2$O$_3$-CaO-FeO ternary diagram of 9th–7th century BCE glass from Europe. Dashed ellipse delineates the range of alum-coloured blue and turquoise glass belonging to the LMK, HMLK, and Al-Co blue compositional groups; circle delineates the range of "black" HMLK glass defined by [38]; dotted line encompasses Fe-rich dark/black-coloured glass, including LMG. Data were obtained from various publications [31,33,37–39,83,86].

HMG remained in use in Europe during the first few centuries of the first millennium BCE, as confirmed by finds from Italy [37] and Poland [33,39]. These glasses are of uniform

base composition, with, on average, 16.8 wt. % $Na_2O$, 4.3 wt. % $MgO$, 2.1 wt. % $K_2O$, and 7.3 wt.% $CaO$. As seen in Figure 1a, the $CaO$-$Na_2O$-$MgO$+$K_2O$ ratios indicated that they were fluxed with plant ashes. HMG samples from Slovenia, dated to the time of the LBA/IA transition [31], might also be considered here. Due to its generally rare occurrence, these high-magnesium glasses could be considered a relic of Bronze Age glassmaking traditions, although it is unclear whether HMG found in Europe was actively being produced during the 9th–7th centuries BCE or whether these examples represent older objects that remained in circulation. The available Sr (avg. 360 ppm) and Zr (avg. 13 ppm) data for HMG from Poland and Italy [33,37,39], coupled with low Nd (up to 5 ppm) for Italian HMG [37], would suggest they were made from a pure quartz source and fluxed with plant ashes, most likely in Mesopotamia.

LMG has been detected in European contexts since the 8th century BCE [78,79]. Only a few samples are known from ninth century BCE contexts in Slovenia [31] and the second half of the eighth century BCE in Poland [33,39], while the majority of examples come from Italy [37,38,83,86] and France and Switzerland [79]. A considerable number of 9th–7th century BCE LMGs do not fit neatly into the natron soda-lime group (Figure 1a). In fact, early LMG tends to cluster with other coeval glass types, such as HMLK, Al-Co blue, LMMK, and high-Al glass (Figure 1a). The most defining characteristic for many of the early LMGs is a comparatively low $CaO$ contribution: avg. 2.6 wt. % for black LMG from Italy [37,38] and 2.5 and 4.4 wt. % for blue LMG from Poland [39] and Italy [86], respectively. When all colours of LMG are considered together for LMGs from Italy [37,38,83], Poland [33,39], and Slovenia [31], $CaO$ averages 3.5 wt. %, while $Na_2O$ values fall in the 9–21 wt. % range. The overall low $CaO$ content of 9th–7th century BCE glass in Europe is in agreement with the standing opinion about lime not initially being recognised as an effective stabiliser for natron glass [37,80], resulting in Ca-depleted and unstable glass being produced in the initial stage of the Iron Age. There is also a notable relationship between the colour and composition of early LMG, in particular black glass (Figure 1b). Italian black LMG is characterised by high $FeO$ (avg. 8.9 wt. %) and low $MgO$ (avg. 0.8 wt. %), but lower $Al_2O_3$ content (avg. 1.3 wt. %) than that of black HMLK glass [37,38]. Their closest parallels are black glasses from France and Switzerland with similarly high $FeO$ (avg. ca. 15 wt. %) and $Al_2O_3$ and $CaO$ contents lower than those of Roman black glass [79]. A compositional match for black LMG glasses outside of Europe can be found in Early Iron Age (11th–10th century BCE) black beads from Pella (Jordan), which, on average, contain 17 wt. % $Na_2O$, 1.5 wt. % $MgO$, 1.6 wt. % $Al_2O_3$, 1.3 wt. % $K_2O$, and 2.3 wt. % $CaO$ [74].

Along with LMG, a similar glassmaking recipe employing natron and different sands is evidenced by contemporary glass finds from Italy [37,38,83,86]. A peculiar characteristic of these samples are $MgO$ values (1.5–4 wt. %) higher than those expected for natron-fluxed glass (up to 1.5 wt. %) coupled with $K_2O$ values (<2 wt.%) lower than those of glasses fluxed with plant ash (2–4 wt.%), representing a sub-group of natron-fluxed glass possibly fused with impure sands rich in pyroxenes or amphiboles [37,38]. These HMLK glasses show colour-specific compositions, and have accordingly been presented as Al-Co blue glass and iron-rich black glass [37,38,79] (Figure 1b). In the case of Al-Co blue glass, low $K_2O$, $CaO$, and trace amounts of P are used as an argument against plant ash flux, while the high $MgO$, $Al_2O_3$ and Co contents, which also show a positive corelation, are explained by the use of Egyptian alums as colouring agents [37,74,79]. These characteristics were also observed in two LBA/IA blue glass beads (corresponding to HMLK criteria presented here) from Slovenia (samples Novo Mesto 6 and 7) [31]. Aside from $K_2O$, $MgO$, $Al_2O_3$, and Co values compatible with those reported for Sarno and Capua [37], the two samples from Novo Mesto also had low Rb contents (BDL[1] to 8 ppm for Novo Mesto samples, avg. 3.8 ppm for Italian Al-Co blue glass) [31], which, when lower than the Sr content, can be used to rule out the use of precipitated plant ash as a fluxing agent [20]. Thus, the two Slovenian samples might be considered some of the oldest examples of natron glass in Europe, depending on the accuracy of their dating into LBA. One turquoise and two blue (FiBo4, FiBo16) glass

samples from Bologna (Italy) [83] also fell outside of the typical flux-stabiliser range for plant ash soda-lime glass, showing more similarities with natron-fluxed Ca-depleted glass such as early LMG, Al-Co blue glass, and black HMLK glass. Further links with Al-Co blue glass can be found in the high $Al_2O_3$ content (avg. 4.8 wt. % for blue and turquoise beads), which is positively correlated with MgO, and Co concentrations in the range of 200–2000 ppm [83]. Based on the presented characteristics, Arletti et al. [83] determined these samples to be early natron-fluxed glass coloured with cobaltiferous alums but produced using Mesopotamian sands, as indicated by their low Zr content (avg. 31 ppm). Egyptian cobaltiferous alums from the Western Oases were used as glass colourants until the 13th century BCE, and were still employed in the production of blue pigments as late as the 11th century BCE in Egypt [36]. With the end of the New Kingdom (late 11th century BCE), cobalt pigments, as well as glassmaking evidence, disappeared from Egypt until the 7th century BCE, when a different cobalt source began to be used [36]. The archaeological lacunae are filled with 9th–8th century BCE Co-coloured glass found outside of Egypt, namely in Nimrud (Iraq) [74], France [79], Italy [37,83], and Slovenia [31], evidencing that Egyptian alums continued to be used in glassmaking—but most likely not for glass made in Egypt—after the shift to natron flux occurred. On the other hand, black and darkly coloured glass corresponding to HMLK criteria from France [79] and Italy [37,38] might have been produced in Egypt. Italian black HMLK glasses typically contain increased amounts of FeO (avg. 8.9 wt. %), MgO (avg. 2.5 wt. %), and $Al_2O_3$ (2.3 wt. %) content than those of black LMG, which was interpreted as resulting from the use of sands containing more Fe-Mg minerals such as amphiboles and pyroxenes [37,38]. Lower Sr and heightened Zr and Hf content compared to that of natron glass produced in Mesopotamia is taken as proof of the Egyptian origin of black HMLK glass [37].

Another glass type, LMMK, was described by Purowski et al. [39] on a set of (mostly) blue Hallstatt C (8th–7th centuries BCE) glass beads from Poland. Although the beads have an overall similar composition to early LMG (on average 0.9 wt. % MgO and 2.0 wt. % CaO), their slightly increased $K_2O$ content (avg. 1.8 wt. % compared to avg. 0.7 wt. % for the previously discussed LMG glass), high $Al_2O_3$ (avg. 5.7 wt. %) accompanied by elevated FeO (avg. 3.4 wt. %), and the presence of unfused raw materials in the glass structure led the authors to initially argue for a local origin of LMMK glass. A subsequently published analysis of an LMMK glass-bow fibula (Glasbügelfibel) from Groszewice complicates this interpretation [103]. Glass-bow fibulae are mostly known from 8th–7th century BCE contexts in North Italy and nearby regions [17], meaning that the Groszewice example is an import testifying to contacts between the Apennine peninsula and continental Europe. More importantly, as the manufacturing centre for glass-bow fibulae is believed to have been located in Northern Italy, it would imply that LMMK glass was produced either in Italy or somewhere else in Europe, whereupon it was transported to both Poland and Italy. Although some analyses have been conducted on glass-bow fibulae, not many have been published [31,103]. The seven Early Iron Age (five broadly dated to EIA, two dated to the first half of the 6th century BCE) glass-bow fibulae from Slovenia fit neatly into the LMG glass type (avg. 0.8 wt. % MgO, 0.8 wt. % $Al_2O_3$, 0.4 wt. % $K_2O$, 7.8 wt. % CaO, 0.9 wt.% FeO) [31]. Even higher MgO and $Al_2O_3$ content (avg. 1.9 wt. % and 9.6 wt. %, respectively), and $K_2O$ (2.2 wt. %), CaO (avg. 2.5 wt. %), and FeO (3.9 wt. %) content compatible with the LMMK glass from Poland was recorded in three 8th century BCE samples from Italy [37]. Interestingly, both these glasses, referred to as high Al glass by Conte et al. [37] and LMMK by Purowski et al. [39], contain elevated concentrations of most trace elements, including B (avg. 4300 ppm for glass from Italy and 3500 ppm for glass from Poland). Aside from the aforementioned high Al glass from Italy, we are aware of only one other publication on coeval Iron Age glass of similar composition. Three black and yellow glass beads from 8th–7th century BCE Sardis (Turkey) show a similar average composition (avg. 0.9 wt. % MgO, 7.6 wt. % $Al_2O_3$, 2.5 wt. % $K_2O$, 2.5 wt. % CaO, 3.0 wt.% FeO, 5000 ppm B) [105] to LMMK glass and high Al glass [37,39]. Regarding the sand source for these types of glass, Purowski et al. [39] argue for the European origin of the LMMK

glass, while Conte et al. [37] refrain from pinpointing the region, but confirm both types were probably made with a highly impure sand source. For the glass from Sardis, Van Ham-Meert et al. [105] successfully demonstrated the similarity of REE patterns to those of local sand and Byzantine glass from Sardis, demonstrating that at least experimental glass production occurred in ancient Lydia [105]. The location(s) of the primary production sites producing the glass found in Italy and Poland remain unknown [105 but the similarity of their composition to the glass from Sardis could mean they were produced within the same region, or at least outside of the established centres in Egypt and the Levant.

As of yet, the relationship between these early types of glass is poorly understood. The overall similarity of many LMG, HMLK, LMMK, Al-Co blue, and high-Al glasses found in the 9th–7th centuries BCE regarding the modifier/stabiliser ratio might suggest a common underlying glassmaking recipe employing sand, as opposed to quartz pebbles, as a network former. The variable amounts of sand-related oxides, such as $MgO$, $Al_2O_3$, and $FeO$, could be seen as experimentation with sand sources. Whether and where natron, plant ash, or a combination of both were being employed as fluxes has been debated [37–39]. For black HMLK and Al-Co blue (and turquoise) HMLK glass, researchers agree on natron as the fluxing agent [37,74,79,83], but point out their different origins in Egypt and Mesopotamia, respectively. Both plant ash [39] and natron [37] have been hypothesised as fluxing agents for LMMK glass from Poland. Based on the overall similarity of LMMK glass to natron-fluxed HMLK and Ca-depleted LMG (Figure 1a), we consider it more likely that LMMK glass was produced with natron, and the increased $K_2O$ content is likely derived from the feldspar impurities in the glassmaking sand.

In summary, it could be said that the initial period of the Iron Age in Europe saw a relatively abrupt shift to natron-fluxed glass. Since previous glassmaking traditions did not rely on the addition of a Ca stabiliser, the need for its addition to glass fluxed with natron was not immediately recognised, resulting in an overall low CaO content of early natron glass, which has also been noted for 10th–8th century glass from the Nesikhons burial in Thebes (Egypt) [81], various glass beads from Pella (Jordan) [74], and 9th–8th century BCE glass inlays from Nimrud (Iraq) [74]. Trace element concentrations point to a colour-based organisation of production, with black Fe-rich glass probably produced with iron-rich Egyptian sands, and Al-Co blue glass most likely produced in Mesopotamia, but using Egyptian cobaltiferous alums [37,38,83]. The origin of other types of glass appearing in Europe during this period is harder to discern. Nonetheless, it seems that different regions employed "specialised" recipes for the production of natron glass. A similar model was established for Bronze Age glass production in Egypt [82], where glass workshops were producing some glass colours and importing others. A comparable system transposed on a larger scale could have persisted throughout the beginning of Early Iron Age. Historically, this is the period of Phoenician expansion to the West, and it may well be that the newly explored maritime networks not only brought Egyptian and Levantine/Mesopotamian glass to Europe, but that the rising demand for glass forced the exploration and, ultimately, the optimisation of natron-fluxed glass.

### 4.2. The Turning Point in Natron Glass Production (ca. 6th Century BCE)

Pinpointing the end of "specialised" glassmaking recipes in the Early Iron Age is nearly impossible due to the overlap of context dating. Nevertheless, as can be seen in Figure 2, the low-Ca glasses all but disappeared during the 6th century BCE, and the glassmaking recipes became more uniform.
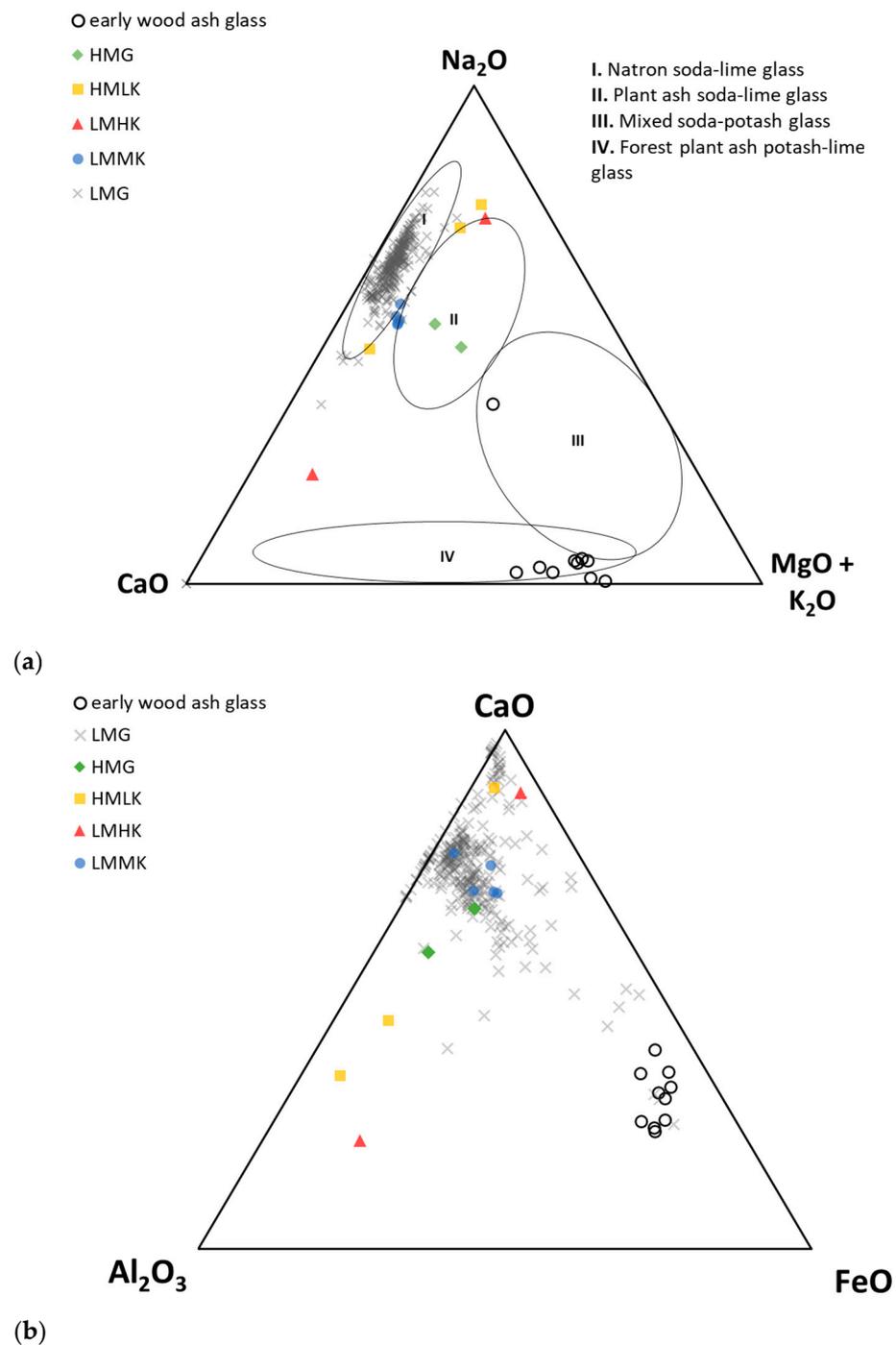
**Figure 2.** (**a**) CaO-Na₂O-MgO+K₂O ternary diagram of glass from Europe roughly dated to the 6th century BCE, showing the relationship between Ca stabilisers and fluxing agents. Leaching of alkali accounts for high CaO outliers. Data were taken from various publications [31,33,37–40,42,84,86,90,92,93]. Glass family ranges were obtained from [10]. Glass types were formally assigned according to MgO and K₂O content. (**b**) Al₂O₃-CaO-FeO ternary diagram of glass from Europe roughly dated to the 6th century BCE. Data were taken from various publications [31,33,37–40,42,84,86,90,92,93].

The few exceptions that exhibit MgO and K₂O values similar to those of previously described glass types are some 6th–5th century BCE samples from Stična (Slovenia) which show MgO and K₂O values compatible with the LMMK group (Stična 42, 44, 45, 51,51) [31], a mixed alkali (LMHK) sample from Slovenia (Stična 41) [31], and two coeval HMG (Wicina 68d) and HMLK (Mechlin 41) samples from Poland [39]. Slightly younger examples, dating

from the 6th–4th centuries BCE, are even scarcer: a HMG sample (Bg) from Bulgaria [84]; two samples from Italy (IG 11b, IG 44b) [93] corresponding to LMHK and HMLK glass, respectively; and another HMLK sample (Stična 29) from Slovenia [31]. Of the mentioned samples, only the HMG glass plots falls into the plant ash soda-lime silica range (Figure 2a). Two other examples—a bead from Poland (Mechlin 41, broadly dated to Ha D/ca. 650–475 BCE) [39] and blue glass of a spindle whorl from Bologna (Italy) (broadly dated to the mid-6th–early 4th century BCE)—are reminiscent of the early natron glass (CaO < 4 wt. %). The rest of the non-LMG samples do not group on the stabiliser–flux graph as would be expected of the aforementioned types (in between natron and plant ash soda-lime glass), and it is likely that they reflect the effects of glass alteration rather than unrecognised glassmaking recipes.

Given the dubious nature of other non-LMG instances past the 6th century BCE, the two low-Ca HMLK beads from Italy (IG44b) [93] and Poland (Mechlin 41) [39] could potentially be explained as heirlooms that remained in circulation for some time after their production ceased. Heirloom artefacts, such as bronze spearheads and antennae daggers, can be found in the northwest Iberian Peninsula from as late as the 1st century BCE, despite having dropped out of use from the rest of Europe at least five centuries before [106]. Glass beads are also noted as potential heirlooms encoding family or territorial power during the Early Iron Age, but since typochronological dating for beads is imprecise, they are seldom recognised as such [106–108]. The blue glass (IG11b) of a blue and white spindle whorl from Bologna is much more puzzling, since the white glass of the same artefact has a typical natron glass composition [93].

The so-far-unique set of wood-ash "black" glass from Chotin (Slovakia) [38], dated between the late 7th and the early 5th century BCE, should also be mentioned here. Chotin glass samples were interpreted as evidence of a European glassmaking tradition operating independently of Mediterranean glassmaking centres, employing impure, iron-rich sands and, most likely, wood ash as the fluxing agent. Without analogous examples, it is impossible to fully understand the spatiotemporal extent of this group, but it is interesting to note that the one known instance overlaps with the period of a mass shift to uniform natron glass production. The Chotin glass was found to have low $SiO_2$ (56–61 wt. %) and $Na_2O$ (0.1–1 wt. %) but high MgO (2–3.5 wt. % and $P_2O_5$ (1.7–4.6 wt. %), indicative of values determined for wood-ash glass [38,55]. Other chemical features of Chotin glass, including high alumina (1.3–3.7 wt. %) and trace element (Zr, Hf, Th, Y) content and a positive Eu anomaly, were interpreted as choices of impure quartz sand enriched in feldspars [38]. Much like the HMLK black glass from Italy, dated somewhat earlier, the choice of iron-rich sand is related to the dark colour of the beads (Figure 2b). Since this glass relies on a completely different type of raw material than other Iron Age glass, glass from Chotin represents the only indisputable argument for the existence of European glassmaking during the Early Iron Age so far.

Despite the dominance of natron-glass after the 6th century BCE, plant-ash glass remained in circulation in some regions. Examples from the 5th century BCE necropolis Pichvnari (Georgia) [91] and 6th–4th century BCE necropolis of Dren-Delyan (Bulgaria) [84] could indicate that the Black Sea and neighbouring regions maintained some form of contact with Asian glassmakers, as well as actively traded with Mediterranean workshops. For the 6th–4th century BCE necropolis of Dren-Delyan, Tzankova and Mihaylov [84] proposed as many as four different glass batches differing in the type of flux and the amount of feldspar impurities in the sand. Their most intriguing conclusion concerns the origin of a plant-ash sample, B, the high $Al_2O_3$ (5.2 wt. %) and $K_2O$ (4.4 wt. %) content of which best matches the compositional characteristics of Central Asian plant-ash glass [84,109]. Similarly, Shortland and Schroeder [91] linked the extremely high $Al_2O_3$ (ca. 10 wt. %) content of plant ash beads from Pichvnari to coeval glass produced in the Indian subcontinent. If organised trade with Asian artisans existed, it is not well-reflected in glass assemblages. A more probable scenario as to how these beads came to be so far from their usual distribution would include "down-the-line" exchanges probably occurring during mass movements

such as the expansion of Achaemenid Empire or mediated by nomadic tribes of the Pontic steppe [84,91]. Most recently, high-Al plant ash glass (3.2–3.7 wt. % MgO, 2.8–5.4 wt. % $Al_2O_3$, 2.7–2.9 wt. % $K_2O$), interpreted as evidence of (indirect) contact with the Black Sea littoral during the 5th century BCE, has been reported from Modlnica (Poland) [33], showing that these types of contact could have reached rather far into Europe.

Natron-fluxed glass (LMG) became the dominant type of glass found in Europe after the 6th century BCE, and it is universally believed to have been produced in the Eastern Mediterranean and imported into Europe. The optimisation of the flux/stabiliser ratio can also be followed through the diversification of forms, namely, the re-emergence of core-formed vessels in the Mediterranean, and Celtic glassworking somewhat later. Beads continued to be in use in the second half of the first millennium BCE, but discerning between different manufacturing regions is not straightforward because these beads show greater variability in their $Al_2O_3$ and $SiO_2$ content (Figure 3), implying more sand sources or primary production sites involved in their production; at the same time, they are more difficult to accurately date.
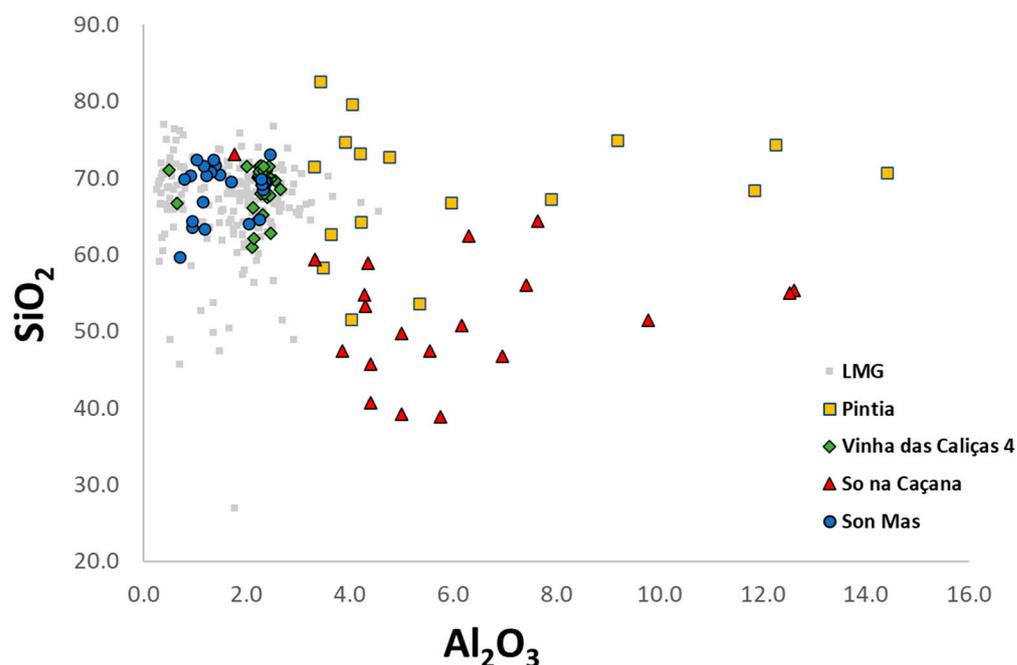


**Figure 3.** $SiO_2$-$Al_2O_3$ biplot for beads, pendants, and other small LMG objects dated to the 6th–1st century BCE from Spain [41,94,95] and Portugal [40], compared to a larger dataset for LMG small finds from Europe. Data for LMG were obtained from various publications [31,33,37,39–42,57,84,90–97].

It is definitely interesting to note that only the natron-fluxed glass from the western Mediterranean exhibits unusually high alumina contents [94,95]. It still remains to be seen if this could signify the existence of a Western glass supply chain, because data for Iron Age vitreous materials from the Iberian Peninsula and the islands are overall limited [40,41,94–112]. Beads with a high $Al_2O_3$ content (3.42–14.41 wt. %), compared to the "typical Hellenistic" composition of other beads from the Iberian Peninsula found in Portugal [40] and on Menorca [41], come from Pintia (Spain) [95] and So na Caçana (Spain) [94]. Links with Asian high-Al plant ash glasses can easily be ruled out since a completely different flux was used to make those beads [109]. In the case of So na Caçana, this composition might reflect an older glassmaking tradition, akin to the high-Al glass found in France and Italy before the 6th century BCE, although the associated material from the stratigraphic unit in which the beads were found is dated somewhat later, between the 4th and 2nd centuries BCE [94]. In this case, the collection of beads from So na Caçana could be an heirloom deposited in a chronologically later context. Similar examples of

centuries-old house hoards, and particularly glass and other jewellery items, are abundant in the northwest Iberian Peninsula [106]. The SEM-EDS results for the 4th–1st c. BCE Pintia beads also show higher $Al_2O_3$ values, seemingly providing an argument in favour of a high-Al glass being in circulation in the western Mediterranean in the second half of the first millennium BCE. Other published Iron Age glass samples from the western Mediterranean dated to the sixth century BCE [40] and the second half of the first millennium BCE [41] fall within the expected $Al_2O_3$ range (ca. 1–2.5 wt. %) for LMG. It is unclear whether the heightened $Al_2O_3$ of the glass from So na Caçana stems from a systematic error of the handheld XRF (HH-XRF) quantification method used in analysing the beads [94]. In their experimental comparison of the performance of HH-XRF instruments, Brand and Brand [113] demonstrated that HH-XRF instruments have poor accuracy when in-built quantification procedures are used, and the difficulties in using HH-XRF to quantify light elements are well known [114,115]. However, aluminium and other light elements are usually underrepresented by XRF, implying that the $Al_2O_3$ content in LMG from So na Caçana [94] is overall higher than that of typical LMG from the Eastern Mediterranean. Furthermore, the roughly coeval samples from Pintia, when analysed by SEM-EDS, also exhibited heightened $Al_2O_3$ values [95]. These high-alumina LMG examples from Spain could reflect the use of feldspar-rich sands, and, by extension, another glassmaking centre supplying glass to the western Mediterranean in the second half of the first century BCE. The origin of these beads could potentially be found somewhere along the coast of North Africa, since similarly high $Al_2O_3$ values have been reported for 8th–4th c. BCE beads from Carthage [116], but more data are needed to confirm this hypothesis. Alternatively, the heightened Al content of both the samples from Pintia and So na Caçana could have resulted from the combined effects of surface leaching and the presence of deposition sediments on the surfaces of the beads, since information on the sample preparation was not provided for either of the sample sets [94,95]. Trace element data are only available for glass from Son Mas and Vinha das Caliças 4, leaving the nature and origin of high-Al glass from the western Mediterranean unaddressed.

A more recent sample set for European contexts included glass beads and other small glass finds from Georgia [91], Poland [33,39,103], Slovenia [31], Italy [57,90,93], Spain [41,94,95], Portugal [40], Bulgaria [84,96], and Hungary [97]. Overlaps in artefact dating from different sites, and broad or uncertain dating for artefacts found within a single site, make it extremely challenging to observe changing trends for the production of glass used in beadmaking in the second half of the first millennium BCE. Low- and high-Al beads often feature on the same sites and display no apparent typological association, which could imply that the bead market was not as specialised as was the case with the core-formed vessels discussed in the following section. Regarding the possible primary production sites, the Zr and Sr values for the beads and small glass artefacts displayed the same characteristic split into low Zr/high Sr Levantine and high Zr/low Sr Egyptian glass (Figure 4).

The fact that glass beads, pendants, and other small artefacts showed greater compositional variability than coeval core-formed vessels might be explained by their artefact properties. Beads are easily transportable, which certainly explains how they can end up far from the place of their origin, as is the case of the possibly Asian beads in the Pontic region. Furthermore, some bead forms, especially monochrome beads, would have required significantly less time and skill to manufacture than other glass artefacts, and could have been made in workshops other than those catering to the demand for core-formed vessels. If high-Al glass from Spain is any indication, bead workshops could have been organised in smaller regional markets, relying on natron (possibly imported) and local sands. Alternatively, the greater variability of glass beads compared to vessels could also mean that less care was put into treating the sand before fusing, or that lower-quality chunks of raw glass were used. Since well-fused glass seldom contains remains of sand minerals, sand processing methods are virtually impossible to prove from glass composition alone. Nonetheless, it is highly improbable that glassmaking sands were used "as

found" in nature, and cisterns discovered on Rhodes were thought to have been used as levitation tanks for local sand [20] prior to being disputed by isotopic evidence [43].
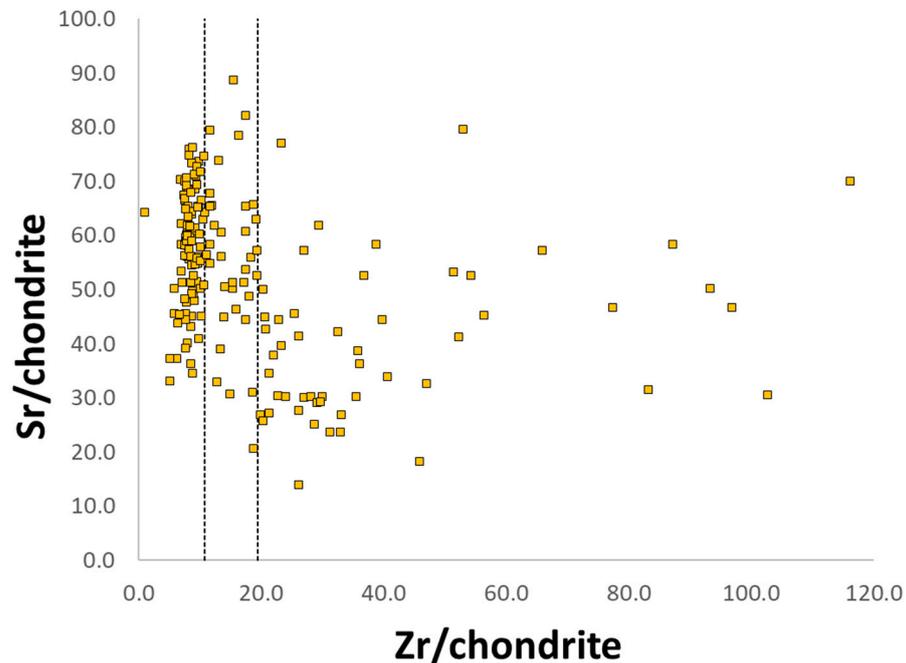


**Figure 4.** Chondrite-normalised biplot of Sr and Zr 6th–1st century BCE natron-glass beads and other small finds from Europe. Dotted lines represent cut-off Zr values for Levantine (**left**) and Egyptian (**right**) sands, proposed by Van Strydonck et al. [41]. Data were obtained from various publications [31,33,39–42,57,84,91,92,94,96].

*4.3. Hellenistic Core-Formed Vessels (6th–1st Century BCE)*

In the Mediterranean, Hellenistic core-formed vessels feature prominently alongside glass beads [20]. These vessels are considered their own topic of study, both from an archaeological [18–20,117] and archaeometric perspective [20,42,43,56,66,90–93]. Core-formed vessels appeared in Mediterranean Europe during the sixth century BCE, about two centuries after they reappeared in Mesopotamia following the BA/IA glassmaking hiatus [19,42]. A great number of core-formed vessels was concentrated in the Eastern Mediterranean, most notably in the Aegean region, where they were manufactured to imitate Greek ceramic forms. Typologically and chronologically, three distinct industries/groups of core-formed vessels are known. The oldest is dubbed Mediterranean Group I, and lasted from the second half of the sixth century BCE to the first half of the fourth century BCE (roughly corresponding to the late archaic and the classical period in ancient Greece). Archaeological evidence places the production of these vessels on the island of Rhodes (Greece), where they featured prominently [117]. It has been proposed that Rhodian workshops were established by Mesopotamian artisans in the 7th century BCE [18]. The Mediterranean Group II is younger in date (second half of the fourth century BCE to the late third century BCE), and introduced new colour combinations as well as a new form of vessels—hydria. South Italy and the Middle East have been proposed as production centres for this group [18]. The youngest group of core-formed vessels is named Mediterranean Group III (mid-2nd to 1st century BCE). This group includes modified forms of alabastra and amphoriskoi. Based on stylistic evidence and find distribution, production centres for vessels of this group are thought to have been located somewhere on Cyprus and along the Levantine coast [18,19]. With the strengthening of Roman control over the Eastern Mediterranean, core-formed vessels were overtaken by Roman glass production [20,118]. While the need for a Ca-bearing stabiliser in natron-based recipes was uniformly recognised by the end of the 7th century BCE, multiple primary production sites using slightly different glass are thought to have

operated into the late 5th century BCE. The difference between these glasses can be noted in the amount of sand impurities reflected in $Al_2O_3$, $TiO_2$, FeO, and other oxides [56,57]. At least three different sand sources were proposed for Mediterranean group II vessels from Satricum (Italy) based on $SiO/Al_2O_3$ content [57], and the groupings hold true for core-formed vessels from Spina (Italy) [66] and Rhodes (Greece) [98]. Variable alumina content was also noted among core-formed vessels from other sites in Italy [56,92] and Greece [42]. In some cases, $SiO_2$ values of core-formed vessels can be as low as 45 wt. %, accompanied by extremely high PbO content in the range of 30 wt. % [56,92,93], which is linked to the yellow colour of the glass, although other opacifier-based colours such as white, and turquoise have a similar effect on glass silica content (Figure 5a). Contrary to the long-standing opinion for primary production sites in Rhodes [18,117], recent Nd and Sr isotope studies of potential glassmaking sands from the Aegean and Rhodes ruled out both regions as primary glassmaking sites for Mediterranean group I vessels [43]. Similar conclusions were also reached by comparing trace element patterns and isotopic data of core-formed vessels with Roman "Levantine" glass [42,56,91]. When a large dataset was considered, Mediterranean core-formed vessels showed a link between their chrono-typology and possible sand provenance (Figure 5b): the earliest core-formed vessels belonging to the Mediterranean Group I had high Sr and low Zr content [42,56,91,92], typical of glass made with Levantine or Mesopotamian sand [7,29,40–42,58–62]. Mediterranean II vessels were split into low- and high- Zr clusters, suggesting that Egypt started supplying raw glass to Hellenistic workshops during the 4th century BCE alongside Levant. Samples belonging to Mediterranean Group III had both the lowest Sr and the highest Zr values [56].

The picture that emerged in the light of the last two decades of archaeometric research on Hellenistic core-formed vessels was similar to the two-step model of Roman glass production [20,118]. Raw glass was made in a limited region in the Eastern Mediterranean [42,56,91] and distributed to specialised workshops, such as those hypothesised for Rhodes. Additional colouring could have occurred in workshops, resulting in glass of highly variable $SiO_2$ content with respect to colouring and opacifying agents represented by extremely high Pb and Ca contents [42,56]. Initially, the glass used in Hellenistic workshops probably came from the Levantine coast. Available trace element data for Group I vessels suggest shell-rich, very pure silica sand [42,56,57,91,92]. Comparisons of Cr, Ti, and Sr content with Sr-Nd isotope ratios have shown that Group I glass is remarkably similar to both Roman Levantine glass and the Belus River sand [42,56,91]. For the subsequent two groups (II and III), other, likely Egyptian, primary workshops joined the new glass trade model. Group II glasses were split into high Sr–low Zr Levantine glass and a low Sr–high Zr group, which could have been of Egyptian origin, based on analogous behaviour for Bronze Age glass from Egypt [29]. The presence of Egyptian glass since the 4th century BCE could be a result of the rising demand for raw glass, not only in Hellenistic workshops, but also in central Europe. As long-distance contacts became more intense, the exchange of knowledge led to elaboration of glass artisans' skills. Consequently, the value of glass artefacts might have shifted more to the possibility of creating innovative forms and colour combinations rather than the rarity of glass as a material. As such, primary production sites would have been forced to forgo making finished products in favour of increasing the production of raw glass. Compositional data for group III vessels are very limited [56], making it appear as if there was a complete shift from Levantine to Egyptian glass in the late 1st millennium BCE. In reality, the fact that Levantine glassmakers continued supplying raw glass to Hellenistic glassworkers, as well as continental Europe, is evidenced by glass of Levantine signature from Celtic oppida and Mediterranean shipwrecks [7,27].
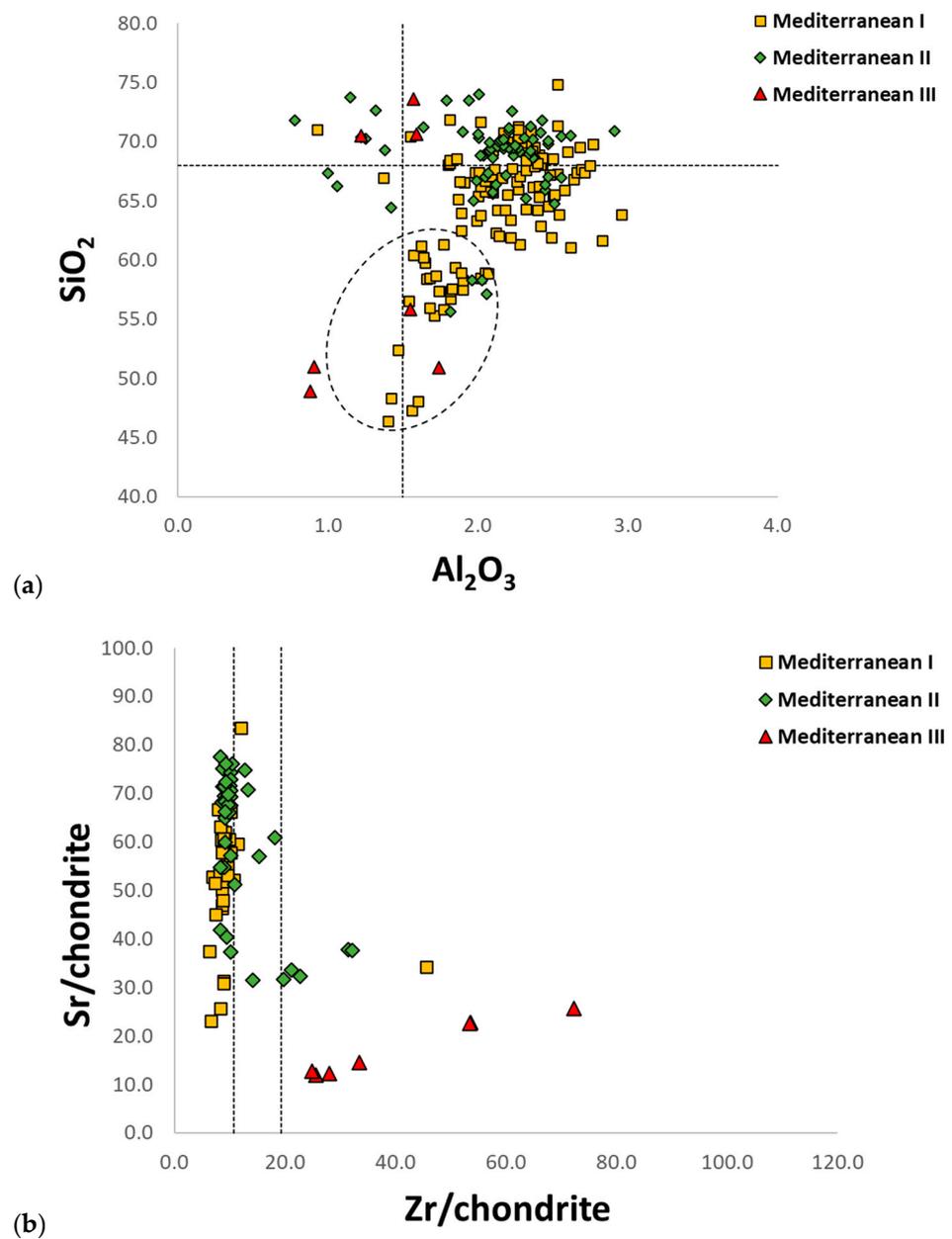
**Figure 5.** (**a**) SiO$_2$-Al$_2$O$_3$ biplot of Hellenistic core-formed vessels from various sites [42,56,57,66,90–93]. Dashed ellipse indicates opacifier-rich, low-silica glass. Dashed lines represent splitting of Mediterranean II group noticed on Satricum samples [57]. (**b**) Chondrite-normalised biplot of Sr and Zr for Hellenistic core-formed vessels of various groups found in Europe [42,56,57,91,92], illustrating the Sr-Zr split between glass of Levantine origin and high-Zr glass made with Egyptian sands. Dotted lines represent cut-off Zr values for Levantine (left) and Egyptian (right) sands proposed by Van Strydonck et al. [41].

### 4.4. Celtic Glass and the Mediterranean Glass Monopoly (3rd–1st century BCE)

While the second half of the first millennium BCE in the Mediterranean was marked by core-formed vessels, the glass assemblage of continental Europe is best represented by Celtic glass. These artefacts constitute the most characteristic elements of the Celtic material repertoire and are found, sometimes in enormous numbers, throughout France in the west to the Dniester river in the east [99,100]. Bracelets are the most common form, but ring-beads and tiny rings (Fingerringe) were also produced by Celtic glassworkers. Much like Hellenistic vessels in the Mediterranean, typological studies of Celtic glass have a long tradition in Europe, and as a result, Celtic bracelets serve as good chronological

markers [22–25,99]. The bracelets appear in virtually all larger settlements dated to the middle and late La Tène periods (250–1 BCE), but are rarely encountered in burials in the eastern extent of La Tène culture, which favoured cremation over inhumation. It is widely recognised that glassworking occurred in Celtic settlements, since Celtic glass forms do not occur outside of the area of Celtic expansion. The lack of glassmaking structures, such as kilns, and associated tools or production waste point to the fact that raw glass was imported from the Mediterranean and shaped into products that were adapted to express a common identity/aesthetic among La Tène populations. Glass used in the production of Celtic artefacts has uniformly been found to contain high $Na_2O$ and low $K_2O$ and $MgO$ content, consistent with natron-fluxed glass [7,27,99–102]. Other Mediterranean goods, such as wine, as attested by imports of amphorae, were also part of Celtic–Mediterranean trade [100]. The best evidence for Celtic import of Mediterranean glass are raw glass fragments from oppida which are compositionally identical to raw glass retrieved from coeval Mediterranean shipwrecks Sanguinaires A, near Corsica, and Lequin 2, along the Provence coast [7,27,59]. The proposed model [100] for distribution of glass in the Celtic world includes primary production in the Levant and Egypt as well as maritime transport of raw glass chunks to southern Gaul, where Greek emporia such as Massalia (Marseilles) were prospering since the 5th century BCE and no doubt played a role in initiating the glass trade further north. From there, raw glass was distributed among Celtic settlements, where it was shaped into finished products. Proposed routes likely followed major rivers to facilitate transportation. Like for Hellenistic core-formed vessels, Sr/Zr ratios show a division of Celtic glass into Egyptian and Levantine groups [7,25,27,58,59,99,100,102]. High-Zr Egyptian glass was found to be more common among earlier Celtic glass dated to the Lt C1 phase [59,99]. The transition to high-Sr Levantine glass occurred during the late Lt C2 phase, around the early 2nd century BCE, and is typical of the late La Tène period (early 2nd–late 1st century BCE) [59,99,102]. This chrono-compositional picture is directly opposite from that noticed for Hellenistic core-formed vessels, where Egyptian compositions feature in chronologically younger Groups II and III [56], which raises the question of what, if further data on Mediterranean Group III vessels do not dispute it, caused this reversal to occur. It could very well be that the more homogenous composition of Levantine sands made the process of fusing and colouring more predictable, which in turn allowed for faster production that overtook Egyptian glassmaking. In her study of Celtic glass from France, Rolland [7] demonstrated that the Egyptian glass can be split into two groups, characterised by low (0.3–1.3 wt. %) and high (3–6 wt. %) $Al_2O_3$ values, while Levantine glass is more homogenous and typically contains $Al_2O_3$ values in the range of 1.6–3.15 wt. %, most closely resembling Foy's Syro-Palestinian group 3 [60]. Intermediate compositions between Egyptian and Levantine glass are taken to indicate glass mixing, but here, the case is probably that raw glass of different origins was worked on together as a batch, rather than intentional recycling on a scale such as that encountered in the Roman empire. Increasing the reliance on homogenous sand sources could be seen as an attempt to standardise glass production in order to lower the time and costs associated with adjusting the recipes to the quality of various regional sands. Another piece of evidence for the standardisation of glassmaking recipes with the aim to optimise production is the systematic increase in Mn content of post-3rd century BCE Celtic glass of various colours [7,99]. Since Mn serves as a decolourant, the proposed hypothesis is that its addition to base glass would have allowed for better control of the subsequent colouring [7]. By the late 1st century BCE, the Levant had all but reached the industrial scale of glass production and seized the monopoly over the Mediterranean and European market. Indeed, with the Roman Empire seizing control over the territory in the 1st century CE and the invention of the glassblowing technique, the production of natron glass reached its culmination point, becoming an easily available commodity.

## 5. Open Issues about Iron Age Glass in Europe

Several factors hinder a better understanding of the evolution of the production and distribution of Iron Age glass found in Europe. Practical issues include the way in which compositional data are reported, the visibility and accessibility of publications, and the level of agreement on how findings are presented. The main theoretical issues concern the chronological discrepancy between various life-stages of glass and the way we conceive of the geographies of the past.

Not all Iron Age periods and regions of Europe are equally represented in scientific publications. While some older data covering the period of the early Iron Age exist for central and southern Europe [88,89], in the last two decades, the focus has mostly been on the Mediterranean part of Europe. In terms of the late Iron Age in continental Europe, most research on glass is conducted in the scope of Celtic studies [27,58,59,100,101]. Glass and faience from the western Mediterranean have only recently started to be investigated. Modern-day Portugal is represented by a single 6th century BCE site [40,110–112]. The Spanish territory is somewhat better represented [41,94,95,119–121], but the reliability and usefulness of some older data has been called into question [41,121]. Moreover, to our knowledge, southeast Europe, namely the Balkans, remains unexplored territory, barring some published data for Slovenia [31] and Bulgaria [84,96] and the yet-unpublished data of IA glass from the territories of Slovenia, Croatia, and Bosnia and Herzegovina [122]. As far as we are aware, the only other recent publication on glass compositions from Slovenia provides only qualitative assessments of 9th–2nd century BCE glass compositions from Novo Mesto, attributing the raw materials to the Near East, but failing to provide an in-depth discussion [123]. Older data exist for Hungary and the territory of former Yugoslavia [89], but cannot fully be used for raw material provenance because of the high uncertainty regarding trace elements. Closely tied to the development of research methods is the scope of the reported data. Trace element analyses, while recognised as the gold standard in glass provenance studies, are not always available. When they are conducted, reported, and even analysed, elements differ between publications, meaning that different datasets might not be comparable for certain elements. Some examples of omitted elements include Hf [57,59,91], Eu [33,37,57], and Nb [57,83,91], but discrepancies can also be found among other elements. Moreover, the results of analyses for standard reference materials, which serve as quality control for the results of the archaeological glass analysis, are sometimes omitted from publications. This problem is especially pertinent when the chosen techniques are known to have significant variations in accuracy and precision, as is often the case with handheld/portable XRF [114,115]. A related issue is the fact that there is no single way of interpreting glass chemical composition. To determine the nature of the fluxing agent alone, $MgO/K_2O$ biplots, ternary diagrams [10], and fractions of $Na_2O$ and $K_2O$ in the total sum of earth and earth alkaline oxides [31] may be used, but as was demonstrated in the previous section, not all of these accurately reflect the nature of the fluxing agent used due to the combined effects of overlap in elemental contributions of raw materials, glass leaching and contamination, and the analytical capabilities of the selected technique. Understanding the composition of glassmaking sands is even more problematic, as iron, aluminium, and lead contents may be obscured by the use of other colourant-related components [12,37,38,56,79].

From a theoretical perspective, the main issue concerning archaeometric studies of ancient glass is that of chronology. This issue manifests on two levels: that of chronology intrinsic to the artefact itself and that of an external chronology, or the object's historical context. Conceptually, the two chronologies reflect the differences between the concepts of chaine operatoire and the life-cycle of an object, and ancient glass studies tend to place more emphasis on the latter, but in a manner that does not always fully incorporate the social complexity encoded in other forms of material culture. This by no means implies that researchers specialising in glass analyses are not equipped to address these issues, but rather that the formats adopted for scientific publications do not leave enough room for in-depth discussions. As a result, the chronologies which many glass studies seek to

elucidate risk boiling down to quantifying, rather than explaining, the timing of changes in glass production. In his work on glass beads from the "Loughey" burial (Northern Ireland), Henderson [124] concluded that glass chemical composition cannot be considered a reliable dating tool. More than three decades have passed since then, and the corpus of chemical data for Iron Age glass (and archaeological glass in general) has significantly increased. Furthermore, trace element and isotopic data have become more available, even if not uniformly reported. As a result, changes in glass composition are, once again, seriously considered as a chronological marker, and multiple subdivisions for the Iron Age are emerging [31,37,38,56]. Given what has been presented in this review, we would argue that glass composition is a chronological proxy worth exploring, but one that must be assessed on large datasets and considered within broader chronological and geographical ranges. Since glassmaking requires a high degree of organisation, from raw material procurement to kiln maintenance, it was no doubt sensitive to external socio-political factors such as wars, trade embargos, and market demands. Another issue to consider is the fact that glass production and glass circulation are two chronologically separate systems, and in most cases, archaeological evidence belongs to the latter. We have already mentioned the case of glass heirlooms in the northwest Iberian Peninsula [106] to illustrate this issue. Rather than discouraging researchers from the use of glass composition as a dating tool, similar examples could be used to further explore the questions of collective memory and inheritance [106,107].

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## Notes

[1] below detection limits.

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