ANDREAS KRONZ

# RHENISH GLASS – THE ROLE OF LATE ANTIQUE TO EARLY MEDIEVAL GLASS PRODUCTION IN THE RHINELAND

## A CHEMICAL INVESTIGATION

With the expansion of the Roman Empire into the region of today's Rhineland, a significant transition in socio-economic social structures took place, which also manifested itself in the changed use of everyday objects. Although the material glass was not unknown in the preceding cultures north of the Alps and was widely used within the Celtic culture in the form of beads and bracelets, hollow glass in the form of drinking vessels and bowls was reserved exclusively for a social elite. A significant upswing in the production of glass objects north of the Alps did not take place until the first centuries of the common era. The question of whether primary glass was produced at all from raw materials within Germania during the Roman and Early Medieval periods can now be largely answered in the negative. One of the most prominent examples of Roman glass production in the Rhineland, the glass workshops of Hambach, located between the modern cities of Cologne and Aachen, can now be shown to have been interpreted as secondary glass workshops[1]. According to the so-called regional model, »global players«, i.e. large (primary) glassworks on the Levantine coast and Egypt covered the predominant raw glass demand of the entire Roman Empire[2]. The trade goods were easily transportable raw glass ingots or chunks, as they can be found for example in numerous shipwrecks[3]. A well-developed infrastructure, both via river systems and an established road network, also supplied the Rhenish glassworks with sufficient raw glass in Roman times.
A very large number of chemical and isotopic analytical studies carried out in recent decades now give us a clear insight into the provenance of glass in antique and Early Medieval times. Although most of the studies represent localized evaluations of glass from specific sites, their totality now provides a clear overview of the production, origin, and distribution of the various ancient and Early Medieval glass types. The Rhineland was one of the centres of Roman glass manufacture and numerous authors refer to the typologically unique forms of the products[4]. They also like to refer to the regional continuity of the glass craft into the Middle Ages, a question that T. Rehren (2001) also took up, but could not sufficiently answer for the actual transition Late Antiquity-Early Middle Ages at that time. Since then, numerous other studies have been published, which contribute more and more to an overall picture of what is known about the origins and formulations of glass. This study presents new analyses of major and trace elements of glass from the well-documented and dated excavations from the urban area of Cologne and surrounding Rhineland in the context of published analyses of contemporaneous glass. In doing so, we can access a specially compiled database of more than 15000 analyses (as of spring 2022) of pre-antique up to modern glass.

### MAJOR GLASS TYPES AND PRODUCTION GROUPS – A BRIEF SURVEY

With the exception of pure lead glass, historical glass is based on the raw material quartz, which is obtained either as sand or quartz pebbles. Alkaline raw materials serve as fluxes. These are mineral soda, mostly

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trona ($Na_3H(CO_3)_2 \times 2H_2O$), or plant ashes. The latter are subdivided into Na-rich ashes, mainly from beach plants[5], or potassium-rich wood ashes. All glasses contain CaO, as »stabilizer« which is either derived from lime of various origins (recent clam shells or limestone), or is already present in the plant ash.

Thus, three main glass types can be defined[6]:

1. Soda lime glass, »SLG«.
2. Soda ash glass, »SAG«.
3. Wood ash glass, »WAG«, also often termed »Forest glass«.

Roman glass and also the glass of the following centuries was almost exclusively SLG in northern Central Europe. For the origin of the mineral soda raw material, the Upper Egyptian salt lakes (Wadi-El Natrun) have been identified[7]. However, some other evaporite deposits are certainly worth considering, even if they probably played a minor role in the first millennium CE[8].

In **figure 1**, the main glass types are shown in the $Na_2O$-CaO-MgO and $Na_2O$-$K_2O$-CaO ternary variation diagrams for the first millennium CE. The variation of these four elemental oxides provides an excellent discrimination of the three main glass types based on their different raw material components. The diagram is based on the database of the Archaeometry Workgroup, GZG[9], and includes here 9025 glass samples dated to the first millennium CE, from a total pool of 14426 type classified glass analyses derived from 214 publications and some unpublished data records.

Of crucial importance is the answer to the question of the provenance of the glass. Countless works deal with this topic. In the meantime, the SLG can be classified into numerous subtypes, but definitions and separations in the literature are not clear and often still confusing. In what follows, we will call these subtypes »production groups«, which includes both a technological distinction (»recipe«) and a chemical distinction of the substances used for production (»raw material signature«)[10]. These production groups allow us to differentiate the various supply regions of the major primary producers. Essentially, these differences are based on:

1. Different impurities in the raw material sand: these components, added as accessory minerals, particularly influence the main components $Al_2O_3$, $TiO_2$ and $Fe_2O_3$, as well as numerous trace elements[11].
2. The CaO raw material, which can be distinguished as recent shell lime or limestone added on the basis of Sr/CaO ratios or $^{86}Sr/^{87}Sr$ ratios[12].
3. A variation in the recipe, which can be expressed, for example, as a variation in the amount of trona and thus a different $Na_2O$/CaO ratio.
4. To produce desired properties in the glass, certain substances were intentionally added. These often significantly change the trace and sometimes also the major element composition: antimony (Sb), manganese (Mn) to decolourize the glass, Sb or tin (Sn) to opacify, other metals (Mn, Fe, Cu, Co, Pb, Sn, Sb) to stain the glass. It should be noted that with the intentional admixture of certain basic materials, which in the case of colouring elements, for example, can come from metal ores, other elements enriched in the basic material can also inevitably considerably change the glass chemistry in the trace element pattern.

In addition to the geographical origin of the various SLG subtypes, their chronological appearance is of great interest. This, of course, presupposes a correct dating of the glass finds. Conversely, as the dating of certain production groups becomes more and more precise, the dating of glass finds on the basis of their chemical composition or specific isotope ratios may become increasingly successful in the future. Possibly the first appearance of a new glass type can be regionally narrowed down to a few years or decades. The decline of a certain type, on the other hand, can certainly not be delimited too sharply. Here, an essential aspect of glass technology played a crucial role: the recycling of glass.

From an energetic point of view, glass recycling always makes sense and it can be assumed that it also took place in every secondary glass workshop. The only question remains: to what extent? Here everything would

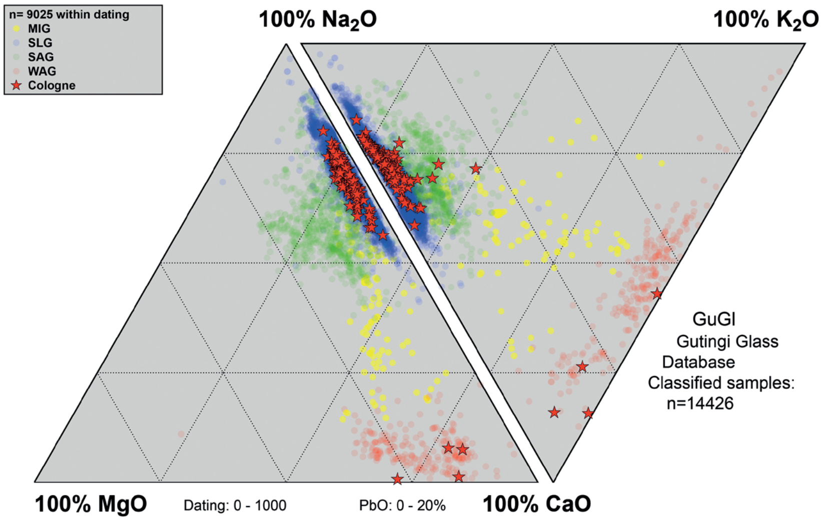


**Fig. 1** Double ternary diagram of the systems $Na_2O$-CaO-MgO and $Na_2O$-$K_2O$-CaO (mass-%). These ternaries are ideal to visualize the three major glass groups soda lime glass (SLG, blue colours), soda ash glass (SAG, green colours) and wood ash glass (WAG, red colours). – Glass from Cologne, first millenium CE (Roman to Early Medieval): red asterisks. – Glass from other sites within the »Harbor project«: green asterisks. – (Diagram A. Kronz, Göttingen).

be possible from »rarely« to the exclusive use of cullet. Numerous studies deal with this question and in the meantime some criteria have emerged to recognize glass recycling by its chemical composition [13]. On the one hand, these are trace elements that are intentionally used as colourants or decolourants (e. g. Co, Cu, Sn, Sb, Pb) and now occur in a certain glass below the effective concentration, but above the geogenic determined concentration of the raw materials. However, again, the limits of these concentrations are not clearly defined. SLG is relatively poor in some elements in its primary production. This offers the advantage that especially certain elements from the fly ash in the furnace, which originates from the wood ash component, can be absorbed into the glass and accumulate during (repeated) recycling: These are essentially K, Mg and P. Possible Na losses due to evaporation during repeated melting are also to be discussed as to whether they can serve as a recycling indicator in special cases. However, due to the different formulations (different mineral soda/trona content), this indicator can only be meaningfully estimated in precisely defined find contexts.

Already in the last centuries of Roman rule, a pronounced recycling of glass can be proven, which can certainly be attributed to the troubled times, especially in the peripheral areas of the empire and the temporary interruption of trade networks [14]. With the socio-cultural upheavals of the Migration Period, this effect can certainly be assumed to have intensified, and should be tested for Rhenish glass.

In the second half of the 8th century, another caesura with the scarcity of SLG led to the »invention« of wood ash glass (WAG). Its earliest evidence was found for the late 8th century in the Carolingian palatinate of Paderborn [15], and for contemporaneous glass in the French monastery of Baume-Les-Messiuers [16]. This transition from SLG to WAG led to the production of mixed glass (»MIG«) in some glass workshops [17]. However, the WAG will not be further investigated in this paper.

**Figure 2** presents an overview of the frequency of the main glass types in the first millennium of our era. Here, all recorded analyses of the data collection assigned to the major glass types are presented, regardless of their geographic distribution. Please note: For clarity of presentation, the frequencies of the main types are not shown relatively among themselves, but only within a respective group. A few outliers will not be discussed further here. E. g., the occurrence of early wood ash glass (WAG) < 700 CE is with some certainty due to erroneous dating.

For this presentation, we evaluated the absolute number of analyses available in the literature as well as our own unpublished analyses. There is, of course, the risk of a bias of the proportions, since especially more modern publications could overrepresent certain sites due to the newer applied analytical methods, which allow a larger sample throughput. Nevertheless, we do believe that this diagram reflects well the respective

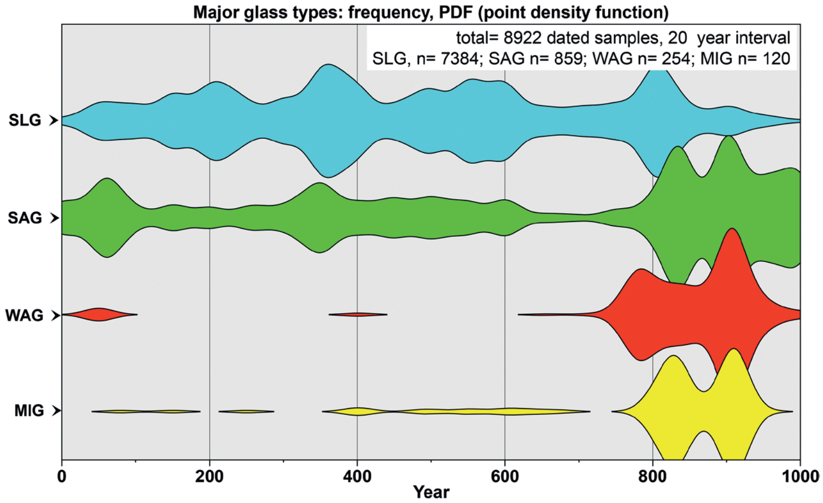


**Fig. 2** Frequency diagram of the three major glass types and a type defined as glass mixing from major types (»MIG«) in the first millenium CE. No geographical filter is applied. – (Diagram A. Kronz, Göttingen, Literature data, Göttingen Glass database, GuGl).

times of increased occurrence of certain glass types. It can be well seen that SLG dominates the first eight centuries of the common era. SAG, however, has never been completely displaced. It is, however, limited in its distribution to southeastern Europe. The strongest caesura occurred at the end of the 8$^{th}$ century with the emergence of WAG, an increased production of SAG and the already mentioned mixture of glasses of different main types, typical in this transitional period, the latter essentially between WAG and SLG. For the SLG, the periods of crisis with reduced glass production alternating with periods of larger glass production are clearly visible in the diagram: at the end of the 3$^{rd}$ century and during the Migration Period, there were downturns, while in Late Antiquity, for example, glass production increased significantly. Likewise, a renewed increase in SLG production can be seen in the Carolingian period, before this type of glass completely disappears in the 10$^{th}$ century.

## »ROMAN«, »HIMT«, »HLIMT«, »LEVANTINE«, »EGYPT«, »FOY-SÉRIES«: THE CONFUSING MULTIPLICITY OF DEFINITIONS OF SODA LIME GLASS

There are numerous suggestions in the literature for variation diagrams to distinguish the different production groups[18].

**Table 1** lists the most important production groups of the SLG. The analysis of the large amounts of data in our data collection certainly resulted in inconsistencies in the classification of the individual types. Therefore, some of the type classifications used in this work are not the definitions as used in the initial identification of a production group in the respective studies: This is not to detract from the value of these papers. Other suggestions for grouping are additionally given in **table 1**, but exact consistency of the various designations of a group is not always given. Also, certain groups have been grouped together, which will be justified in more detail below.

**Figure 3** is an attempt to classify the identified production types of soda lime glass in their temporal development during the first millennium CE, based on a large number of samples from our data collection.

In addition to the problems of actually assigning the glasses to specific production groups, it can be expected that for some of the samples there is also erroneous dating. Often, glass of different ages is mixed in the settlement context. However, the large number of samples evaluated somewhat compensates for

| Production group/ Subtype (this paper) | other definitions | approx. first oc-curence | approx. decline | Provenance | References |
|---|---|---|---|---|---|
| Roman-bg | | 3rd c. | 4th c. | Levant | Silvestri 2008; Brems/Degryse 2014; Schibille et al. 2012 |
| Roman-Sb | Foy groupe 4 | 2nd c. | end 3rd c. | Egypt | Silvestri/Molin/Salviulo 2008; Hoff-mann Barfod et al. 2020 |
| Roman-Mn | Foy groupe/sér 3.1 | | | Levant | Silvestri/Molin/Salviulo 2008; Foster/Jackson 2010 |
| Roman-SbMn | | end 2nd c. | end 4th c. | mixed | Silvestri/Molin/Salviulo 2008 |
| Levantine-1 | Foy groupe/sér 3.3 | late 7th c. | early 8th c. | Levant | Freestone/Gorin-Rosen/Hughes 2000; Phelps et al. 2016 |
| | Levantine *sensu lato* | 4th c. | 8th c. | Levant | Foster/Jackson 2009 |
| Levantine-2 | | | | Levant | Freestone et al. 2000; Phelps et al. 2016 |
| HIMT-1 | Foy groupe 1; HIMT *sensu stricto* (HIMTa, HIMTb) | 325 | 5th c. | Egypt | Mirti/Casoli/Appolonia 1993, group »E«; Freestone 1994; Freestone/Pon-ting/ Hughes 2002; Freestone/Wolf/Thirlwall 2005; Foster/Jackson 2009; Nenna 2014; Freestone et al. 2018 |
| HIMT-2 | Foy groupe/sér 3.2 | 300 | 5th c. | Egypt? | Foy et al. 2003; Foster/Jackson 2009; Gliozzo et al. 2019 |
| | Foy groupe/sér 2.1 | 1st h. 6th c. | 7th c. | Egypt? | |
| | Foy groupe/sér 2.2 | 2nd h. 7th c. | end 8th c. | Egypt? Recycled | |
| | HLIMT | | | | Ceglia et al. 2015 |
| | Saxon | 400 | 700 | | Freestone/Hughes/Stapleton 2008 |
| Egypt-1 | | 710 | 790 | Egypt | Phelps et al. 2016; Schibille et al. 2019 |
| Egypt-2 | | 720 | 970 | Egypt | Gratuze 1988; Phelps et al. 2016; Schibille et al. 2019 |

**Tab. 1** Production groups of Soda Lime Glass. – (Table A. Kronz, Göttingen).

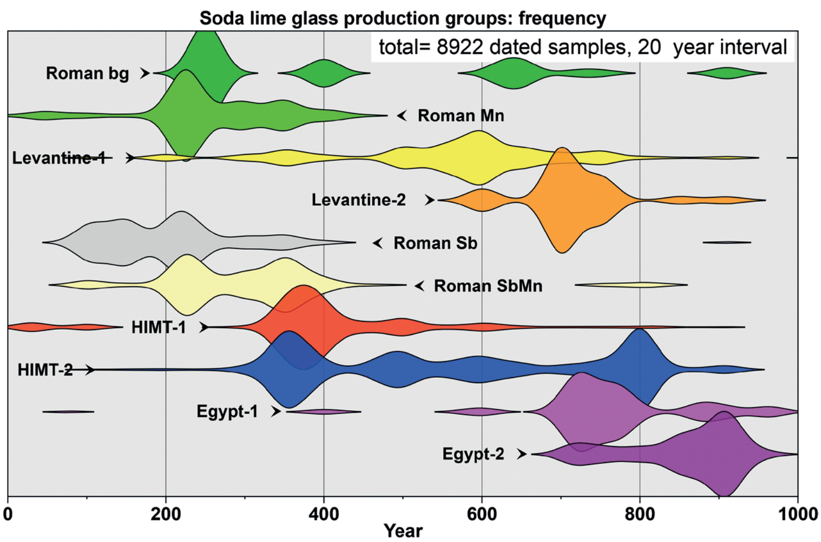


**Fig. 3** Classification attempt of the glass subtypes in a frequency diagram of production groups within the SLG major type. Regardless of remaining errors in dating and type assignment, clear temporal distribution trends are recognizable due to the large number of evaluated samples. – (Diagram A. Kronz, Göttingen, Literature data Göttingen Glass database GuGl).

this effect. As in figure 2, the different subtypes cannot be compared with each other because of their very different absolute numbers, but rather the relative proportions within a respective group are shown in their temporal variation. The temporal distribution is quite determinable for the SLG subtypes (production groups). Some observations, which are already sufficiently described in the literature, are briefly outlined again here:

Even in ancient times, glass could be decolourized very effectively. Antimony (Sb) and manganese (Mn) were used as the most important decolourants, with arsenic (As) being added only in modern times. An overview of the distribution of Sb-, Mn-, and SbMn-decoloured glasses regardless of production type is given by Elisabetta Gliozzo (2017).

1. »Roman« is used here to distinguish the early SLG glass types, which are seperated from the other SLGs by a relatively pure sand raw material and exclusively shell limestone as Ca raw material. Roman colourless glass begins with the Sb-rich type (Roman-Sb)[19]. It is apparently not replaced by the colourless glass that is decolourized with Mn (Roman-Mn), rather both variants exist contemporaneously from the beginning of the 3rd century (fig. 3). Consistently, they also seem to occur very early as mixtures (Roman-SbMn). This mixed glass is still common in the 4th century, while at least the pure Roman-Sb circulates only in residual quantities in this period.

   Roman-Sb is attributed to an Egyptian provenance, while Roman-Mn and Roman-bg are of Levantine origin (Hoffmann Barfod et al. 2020). In Late Antiquity, we then observe the marked increase in the mixing of both Roman types to Roman-SbMn a mixed type that can attest to the widespread recycling of old glass[20]. Figure 3 shows the occurrence of Roman-SbMn, however, in two temporal centers of distribution. The occurrence of early Roman-SbMn coincides with the occurrence of Roman-Mn. Thus, it cannot be excluded that either this mixed form was already produced in certain primary glassworks or that both raw glass types were fused together in secondary glassworks. A more detailed evaluation on the degree of recycling will answer this question, but is not the subject of this work.
2. »Levantine« refers to Late Antique, Early Medieval SLG: They have been defined at type localities such as Jalame, Apollonia, and Bet Eli'ezer for 6th-8th century glass[21]. Their demarcation from the »Roman« is rather fluid, but certainly there are differences that indicate changes in formulations, such as the much lower $Na_2O$ contents of the Levantine type. In its pure form, Levantine glass contains no intentionally added manganese or antimony, and the natural Mn content is less than 300 µg/g. In the literature, however, we repeatedly find manganese-containing glass referred to as »Levatine«; in most cases, this is probably glass that has already been recycled[22]. Also, in some studies certain older ancient glasses (before 400) are classified with the group designation »Levantine«, because they show characteristics that correspond to the Levantine *sensu stricto* of the 6th/7th century[23]. The basic chemical similarities of both Levantine and Roman-Mn glass types in terms of non-intentionally added elements point to the same or similar sand sources and a common origin from the coastal region of the Levant. The classification »Roman-Mn« or »Levantine« is therefore probably more a question of the recipe and less one of the sand raw material source.
3. Two groups of very late SLG are identified as Egypt-1 and Egypt-2. Based on their chemical signature, these are quite easily distinguishable from other production types. Due to their original definition on the basis of stamped glass weights, it has been possible to date the occurrence of both groups very precisely. The Egypt types can be subdivided into further subtypes whose temporal distribution is well recorded[24].
4. The designation HIMT (high iron, manganese, titanium) *sensu stricto* is used to define SLG types that are well distinguished from the Roman and Levantine groups by significantly increased contents of the specified elements[25]. In addition to the designations HIMT-1 and -2, »weakHIMT«, »strongHIMT«, and

HIMT-a, -b, and HLIMT[26] are also found in the literature for further subdivision[27]. HIMT- 1 as well as the earliest HIMT-2 (also: Foy série 3.2) appear apparently very spontaneously on the markets, HIMT-2 even slightly earlier. In Hambach, both types are used contemporarily. The types Roman-Mn and -SbMn compete at the same time (**fig. 3**).

While HIMT-1 does not have a too long persistence and is of minor importance already in the 5th century, glasses with a chemical signature of HIMT-2 continue to persist, even after the disappearance of the actual Roman types, to compete with Levantine glasses. HIMT-1, or HIMT *sensu stricto*, are thereby chemically quite clearly distinguishable from the other production types. This group was already conspicuous in earlier work[28] and can today be assigned with some certainty to an Egyptian provenance, since also certain trace element patterns agree with the Egypt types. The HIMT-2 glass type is more difficult: its complexity and differentiation from other production types will be discussed in detail below. This designation is delineated for glass that is similar in chemical signature to HIMT-1 but does not contain quite as high concentrations in the designating elements (Fe, Mn, Ti), hence referred to by Rosenow and Rehren (2014) as weakHIMT or otherwise as HIMT-2[29].

If we try to distinguish the HIMT-2 group from the Roman, Levantine and HIMT-1 type glasses on the basis of the chemical composition, we come across a large group that is difficult to define, characterized by a greater dispersion of the data, as well as a large temporal depth starting from the beginning of the 4th century and persisting until the 9th century. This observation is not new and has prompted various authors to undertake further subdivisions within this group, among others. D. Foy et al. (2003) distinguish their »groupe/série-3.2« for Late Antique, and the »groupe/série-2.1/2.2« for Early Medieval glass, the youngest series with higher proportions of recycled glass. A. Ceglia et al. (2015) defined a chemically well delimitable HLIMT group as a separate type. I. Freestone et al. (2008) introduced the term »Saxon« for Anglo-Saxon glasses in the 5th-7th century period (**tab. 1**), but these also show considerable chemical scatter in the variation diagrams.

In a review article, I. Freestone and co-authors (2018) reject the designations weak HIMT, or HIMT-2, and speak only of »HIMT« (*sensu stricto*) for glasses classified in this paper as HIMT-1, but with a further subdivision (HIMTa, b) that can be distinguished on the basis of different correlation trends of Ti and Fe. And indeed, some trace/major element ratios (e. g., Th/Zr vs. La/Ti) show that the Egypt groups and HIMT (*sensu stricto*, referred to in this article as HIMT-1) form a definable group relative to the Roman-Levantine and just such groups that we group together here as HIMT-2 (including Foy-3.2, -2.1, -2.2, HLIMT)[30]. However, if we try to classify the SLG, which can neither be assigned to the Roman or Levantine types (due to higher Fe, Mn, Ti contents), nor classified as HIMT-1, as the types defined by Foy et al. (Foy série 3.2, or group 2) or HLIMT, substantial difficulties of an unambiguous assignment arise for many literature data and the glasses we studied. We therefore group all glasses of a particular signature (shown in **fig. 5** and **6**) in the mutual Ti-Fe-Al ratios here as »HIMT-2«, even though we are aware that some subgroups could be more clearly assigned to Foy séries 3.2, 2.1/2.2, or HLIMT. What our HIMT-2 group has in common is that the glasses probably obtained their sand raw material from a largely uniform deposit.

I. Freestones[31] arguments for a deliberate addition of (a yet unknown!) Fe-Mn-ore to the HIMT (*sensu stricto*) glass as a »branding« and quality proof, appear comprehensible and conclusive. Consequently, however, it cannot be ruled out that glass made from the same raw materials (the same sand source and the same trona) may exist without this particular additive. At least Foy's série 3.2 appears to be such a glass type, because of its contemporary and often local occurence (as Hambach shows!). Therefore, the designation »HIMT-2« still seems appropriate to us to express the same basic glass type of probably Egyptian (?) provenance.

## METHODOLOGY

From the glass samples chips of 1-2 mm in diameter were cut off and mounted into epoxy 1-inch discs. After grinding a final surface polish up to 1 µm finish was established. The surface was coated by a 15-20 nm carbon layer to enable electrical conductivity. 20 major and minor elements were analysed by electron microprobe (EPMA) at the Geowissenschaftliches Zentrum, Göttingen University. The JEOL JXA 8900 instrument is equipped with 5 wavelength dispersive spectrometers for quantitative measurements. Synthetic and natural reference material are used for primary calibration (Appendix, tab. A1). The correction of the raw data is done by the »phi-rho-z algorithm (z)« of the CITZAF program according to Armstrong (1995). The CORNING glasses COR-A, -B, C, -D were analysed as secondary references[32]. Deviations for COR-A and COR-B and COR-D which represent the typical element ranges of SLG, SAG and WAG respectively are found in general to be less than 2 % relative for element oxides abundant in concentrations > 1 wt. % and 1 to 10 % for concentration ranges of 0.1 to 1 %.
Table A1 (Appendix) gives an overview of the analytical conditions:
Five individual measurement points are averaged. The standard deviations in comparison to the predicted errors by counting statistics enable an assumption about the homogeneity of each sample.
The electron beam was defocussed to an effective diameter of 22 µm to avoid the well known fact of Na-loss in Natron glass due to electron bombardment.
Major and trace elements were measured at the Geowissenschaftliches Zentrum, Göttingen University using a ThermoFisher Scientific Element2 single-collector, high-resolution magnetic-sector inductively-coupled plasma mass spectrometer, combined with a Resonetics Resolution M50 Laser ablation system (Coherent COMPex 193 nm ArF eximer laser). The laser operates at a pulse rate of 10 Hz and a focused laser energy of about 3 J $cm^{-2}$. Focusing was adjusted at 100 µm diameter spots and a sampling depth of approx. 100 µm was aquired. The NBS610 Standard glass (NIST) containing nominal 450 ppm of almost all elements except the major elements was used for primary calibration. In total, we obtained a methodologically compatible data set of 68 elements.

## SAMPLES AND SAMPLING

The study focuses on the extensive glass finds from the Harbour area in Cologne, especially from the excavations on Heumarkt and Kurt Hackenberg Platz[33].
An important aspect was also the analysis of other Early Medieval glass finds from trade centers that may have been supplied with Rhenish glass. These include the North Sea islands but also finds along the trade routes within the Frankish Empire.
A total of 276 glass samples were characterized with 287 analyses. Of these, 147 analyses were of material from the immediate urban area of Cologne and 140 analyses were of comparative samples from potential trade locations. In order to obtain a more comprehensive picture of the development of the glass typology, previously described glass finds from the Rhineland were also re-analysed due to the changes in analytical methods in the meantime. Even though the quality of the older analyses is generally good, in some examples systematic deviations in the data occurred, which even affected the interpretation of the glass provenance[34]. Furthermore, trace element data were often not available from the older analyses, but these are important for understanding the glass interpretation.
Repeat analyses were performed on:

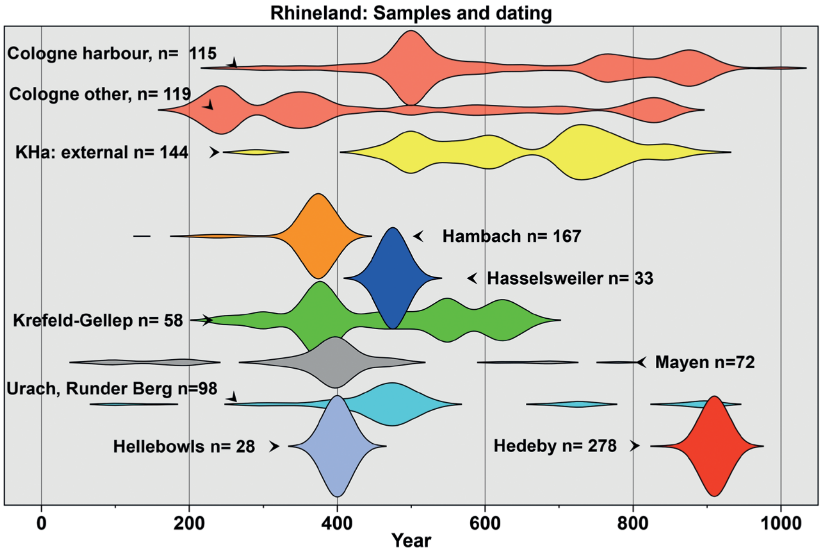


**Fig. 4** Temporary distribution of SLG discussed in this paper: Cologne Harbour excavations (Heumarkt and Kurt-Hackenberg-Platz), other Cologne sites (e.g. Tel-Aviv-Straße), external sites investigated within the »Harbour project«, Hambach (new analyses within this project; data of Rehren/Brüggler 2020), Hasselsweiler (new analyses; Päffgen/Wedepohl 2004), Krefeld-Gellep (new analyses; Wedepohl/Pirling/Hartmann 1997), Mayen (unpublished data RGZM Mainz; Grünewald/Hartmann 2014), Urach, Runder Berg (Czygan 1987; Koch 1987), Hellebowls (Rehren/Brüggler 2015), Hedeby / Haithabu (Dekówna 1990; Kronz et al. 2016). – (Diagram A. Kronz, Göttingen).

1. 89 samples (92 new analyses) from Hambach (74 analyses) and comparative Roman glasses (18 analyses, mainly barrel jugs from potentially Hambach production of different sites)[35].
2. 17 new analyses on 16 glasses of Merovingian glass production at Hasselsweiler near Jülich[36].
3. 24 new analyses of glass from the Romano-Frankish cemetery of Krefeld-Gellep[37].

Recent systematic studies carried out on Late Antique/Early Medieval glass in the Rhineland, for example the »Hellebowls«[38] and Mayen[39], are also included for evaluation. Other earlier analyses with possible relation to the Rhineland are available from the »Runder Berg« near Urach[40]. **Figure 4** lists the comparative samples of Rhenish provenance discussed here in their temporal frequency distribution of finds. The ages are based on archaeological dating. They are presented as point density functions, with values randomized over the dating period, assuming symmetrical distributions.

## RESULTS

### Glass from Hambach, Hasselsweiler, Mayen, and Krefeld-Gellep – new analyses, new assessment

**Figure 5** and **6** are used to distinguish the different subtypes of the SLG. We have selected three different binary diagrams whose axes are each represented by an element ratio[41]. There are numerous other options to make differentiations also on the basis of specific trace elements, which would go beyond the scope of this paper[42]. The distribution fields (coloured areas) have been determined from the variation of the respective production types, since the diagrams are no longer clear when all individual analyses are displayed. These fields are a momentary snapshot, which will most certainly change again with the extension of the database and possibly new definitions. Also, different production groups overlap in certain diagrams, so that a single diagram does not allow a clear assignment of a glass to a specific production type.
The very extensive glassworks of Hambach prove that the Rhineland certainly belonged to an important center of secondary glass processing in the Late Antique northwestern Roman Empire[43]. The discoveries and excavations of these extensive furnace sites were made during archaeological explorations in preparation for the Hambach open-pit lignite mine. It is quite likely that there were several other large Roman

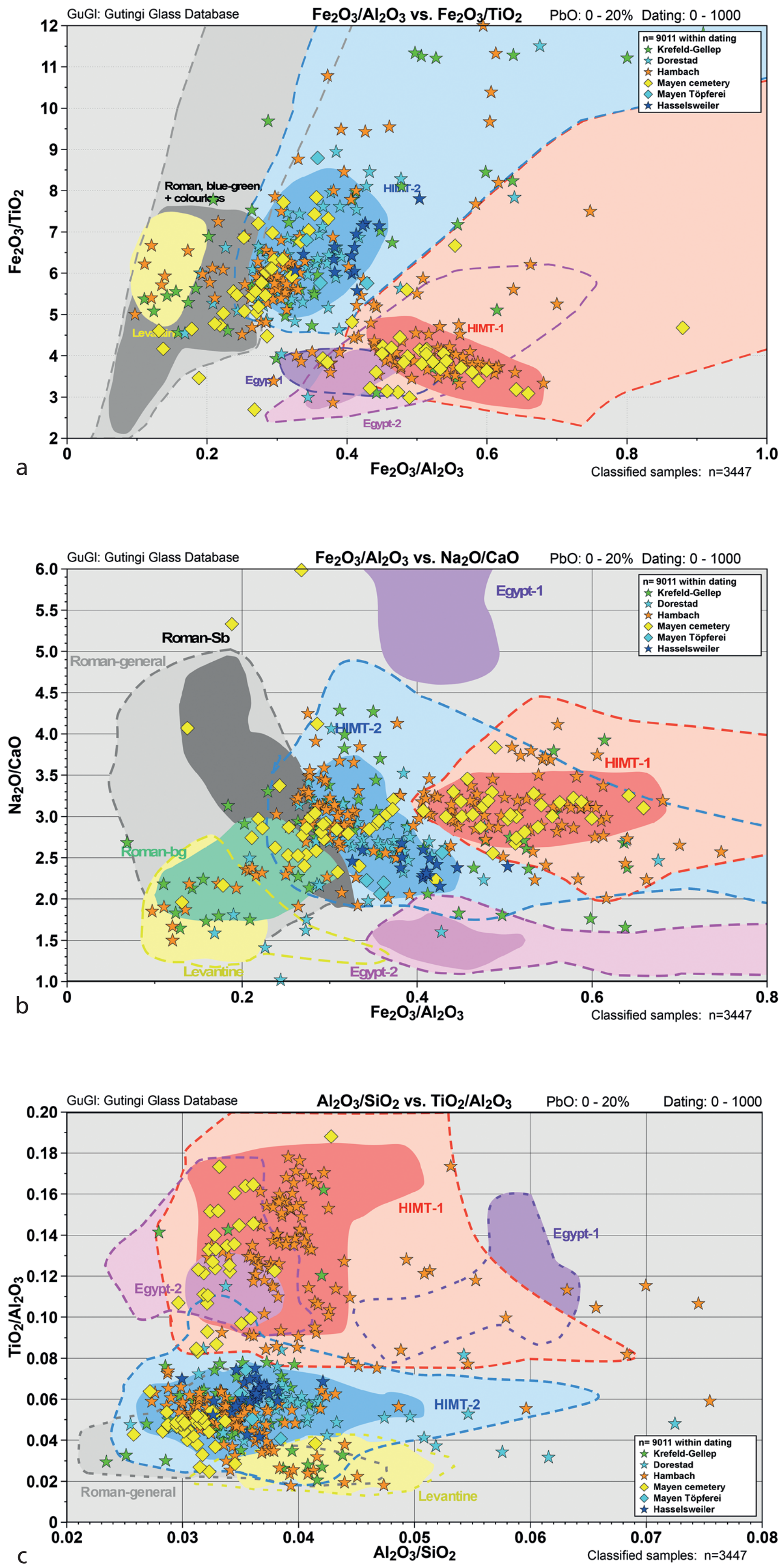


**Fig. 5** Binary element ratio diagrams useful for the separation of SLG productiongroups: **a** $Fe_2O_3/Al_2O_3$ vs. $Fe_2O_3/TiO_2$. – **b** $Fe_2O_3/Al_2O_3$ vs. $Na_2O_3/CaO$. – **c** $Al_2O_3/SiO_2$ vs. $TiO_2/Al_2O_3$.
The coloured fields are estimated distributions of the respective SLG production groups, based on the GuGI database on its current status (3447 classified samples, SLG subgroups). Glass from Hambach, Krefeld-Gellep, Mayen. – (Diagram proposal after Schibille et al. 2017, diagrams A Kronz, Göttingen)

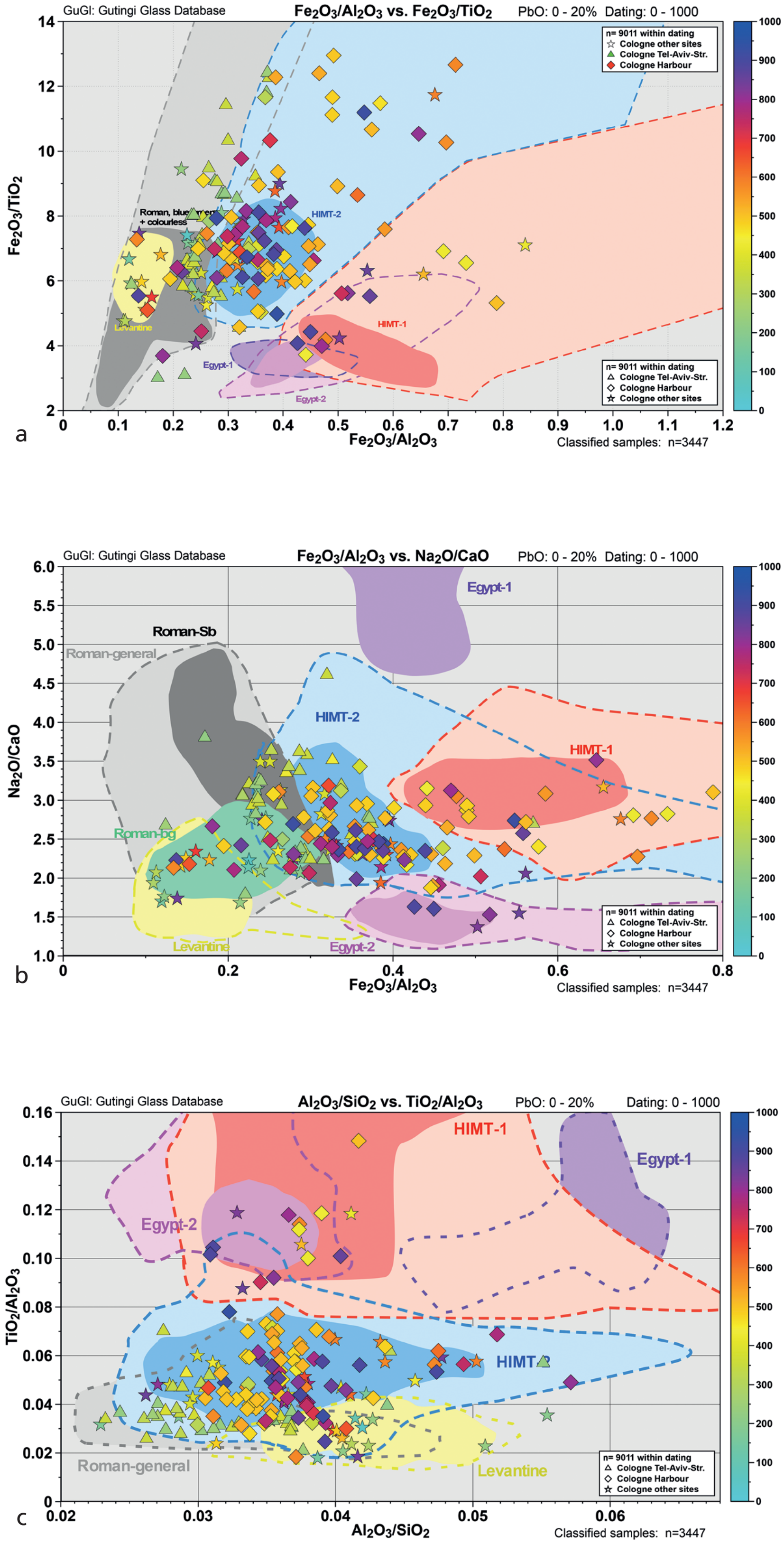


**Fig. 6** Binary element ratio diagrams useful for the separation of SLG productiongroups **a**, **b**, **c** as in fig. 5. Cologne samples. – (Diagrams A. Kronz, Göttingen).

production centers regionally where raw glass was processed into finished objects[44]. A renewed evaluation of the Hambach glass is of importance in so far as, firstly trace element data are now available, and secondly the transition from Roman antiquity to the Early Middle Ages in the Rhineland can be recorded with regard to the glass craft. The main focus is on the question of changes in the glass recipes and thus in the composition of the glass, which firstly can give conclusions about the raw glass sources, but secondly should also help to answer the question about the recycling of the glass in particular. At the same time as our analyses, a new analysis of the Hambach glass was carried out by Rehren and Brüggler (2020).

While Karl Hans Wedepohl, based on the minor element signature, considered the Hambach glasses to be an independent Roman primary glass production[45], recent studies clearly show that it is not a regionally typical chemical glass signature, but rather the typical Late Antique glass types HIMT-1 and HIMT-2 (Foy série 3.2), which were traded throughout the Roman Empire[46]. According to Wedepohl, the arguments in favor of a primary glass production at Hambach were, first, the installed large tank furnaces and, second, a divergent minor element signature that followed the local sand deposits in the northern Eifel region, especially with regard to iron contents. However, due to the typical $^{87/86}Sr$ isotopic signature, which follows the recent seawater Sr isotopic signature and the high Sr/CaO ratios, the use of imported shells for this glass production must be postulated[47]. Meanwhile, we know that Roman tank furnaces were obviously not restricted to the production of primary glass from raw materials. The iron contents of the original analyses from Hambach were systematically determined too high and now by no means result in an independent group that can be distinguished from the other glass subtype[48]. Rather, the now newly determined iron contents clearly fit into the variation ranges of the Late Antique HIMT-1 and HIMT-2 glass compositions. The HIMT-2 group of Hambach glass is chemically still close to the Roman types. The agreement with the série 3.2 group defined by Foy et al. (2003) is not particularly satisfactory, contrary to what is presented in Rehren and Brüggler (2020). For Hambach, we do not see a continuous transition between HIMT-1 and HIMT-2 glass. Rather, the foci of chemical variation of the two groups are well separated, and mixing of these production types apparently did not occur at the site (**fig. 5a-c**).

Only a short distance away from the Hambach glass workshops, the glass finds from Hasselsweiler near Jülich prove the existence of a glass workshop from the 2nd half of the 5th century[49]. The find material is well dated and covers a not too large temporal spread of only about 50 years. The glass finds are significant because they are located near Cologne and Hambach and chronologically follow the Hambach glassworks. This glass, however, dating to less than a century later, is easily distinguishable from the Hambach HIMT-2 group and shows a clear change in glass chemistry. The Hasselsweiler glass corresponds quite well to the Foy série 2.1. They also coincide with the group of glasses from Cyprus defined as HLIMT by Ceglia et al. (2015). Obviously, a change in the source of the raw glasses occurred in the 5th century. The compositions scatter only slightly, which is evidence for a temporary activity of the workshop and the supply of the raw glass from probably only one source (**fig. 5a-c**).

For glass research in the Rhineland, the finds from the Krefeld-Gellep cemetery are of considerable importance: In contrast to other burial sites, the occupation of the cemetery around Fort Gelduba did not stop with the end of the Roman period, but continued well into Frankish times. K. H. Wedepohl recognized this importance and has analysed Roman and Frankish glass[50]. Fundamental changes in glass compositions were recognized in the transition from Roman to Frankish glass, but the analysis at that time of only 27 samples is quite modest compared to the large number of glasses found. With our new analyses of the same sample assemblage, trace element data are now available. The compositions of the Gellep glasses vary widely, which on the one hand is due to the great temporal depth of this glass assemblage, but on the other hand certainly also to the find itself. Unlike in a workshop, here glasses were collected as grave burial objects from certainly very different origins. Only one vessel can be assigned to the early Roman-Sb type, five others are either Roman-Mn or Roman-bg, only two HIMT-1 vessels can be identified. The large remaining group is HIMT-2, with the older, Late Antique group (corresponding to Hambach HIMT-2) and the younger group (corresponding to Foy série 2.1/2.2) being about equally represented. The analysis of a claw beaker (»Rüsselbecher«) can be regarded as a remarkable finding: Except for some colouring trace elements (Cu + Pb) and production-related slight differences in Na2O content, the fragment of the claw beaker from Gellep (Gel-27) is chemically identical within the analytical error limits to a claw beaker fragment from Norddorf on the island of Amrum (KHa 137). Thus, they not only originate from the same workshop, but must have been made at the same time in the first half of the 6th century from the same raw glass batch[51].

Late Roman glass from Mayen/Eifel roughly coincides with the period of operation of the Hambach Ironworks. Glass analyses were carried out by S. Hartmann and S. Greiff at the Römisch-Germanisches Zentralmuseum in Mainz (RGZM) on samples from a Late Roman cemetery[52]. Four other samples of younger glass (5th, 7th, 8th/9th century) from a settlement context were analysed by us[53]. The majority of the Roman glass from the cemetery of Mayen shows a remarkable agreement with the two Hambach groups HIMT-1 and earlier HIMT-2. The deviation still to be noticed in the diagrams of Grünewald and Hartmann (2014, fig. 6.5) is, as already mentioned, to be attributed to the too high determined iron contents of the earlier analyses in Gaitzsch et al. (2000). In addition, a very clear separation of both Hambach glass groups can be observed. It shows that both glass sources were not mixed. However, the separation between the classical »Roman« types and HIMT-2 remains blurred. Here we see, especially for Mayen, a continuous development from the classical colourless Roman types towards the higher titanium and iron contents, the Late Antique glasses called »série 3.2« by Foy et al. Also, this early HIMT-2 group is intermediate between HIMT-1 and the Roman types in many variation diagrams.

## Glass from Cologne

Cologne glass workshops can be traced back to the 1st century AD[54].

Roman glass from Cologne, the Colonia Claudia Ara Agrippinensium (CCAA), could be analysed from the finds of the excavation Tel-Aviv-Straße, with 44 samples. 43 samples are commodity glass, while one sample turned out to be waste of metallurgical processing. The findings date to the period from the 3rd to the beginning of the 5th century[55]. The most outstanding find is a shard of a conchylia cup (»Konchylienbecher«, a variety of a fish beaker)[56]. The finds, among others, also glasspipe cuttings, raw glass and crucible fillings indicate a small-scale glass processing workshop (*officina*), which probably produced high-quality hollow glassware. The result of the analyses is remarkable: As expected, all Roman types (Roman-Sb, Roman-Mn and Roman-SbMn, with a tendency towards HIMT-2) are found. However, not a single HIMT-1 glass can be detected, although the specimens date until the beginning of the 5th century. Apparently, the yellow-green coloured HIMT-1 glass was not desired for the production spectrum of glassware of this small specialized workshop, although it was supplied in masses at the same time (at least for the Late Antique phase) in Hambach, hardly 30 km away. If one follows the arguments of Freestone et al. (2018), the HIMT-1 glass was intentionally coloured to denote its easier smeltability on the merchandise product compared to the Roman types. HIMT-1 is, relative to the classically colourless Roman type, easier to melt and has a lower viscosity at a given temperature than the glasses of Levantine provenance. Thus, it is plausible that the HIMT types were well suited for simple mass-produced goods, such as bottles. The Tel-Aviv-Straße workshop in Cologne may have produced primarily sophisticated, high-end glassware for the luxury market: only the colourless Roman-type glasses could be considered for this purpose. With some certainty, this raw glass also had a higher price. Both groups could apparently still be easily obtained in the Rhineland in the late 4th century. Foster and

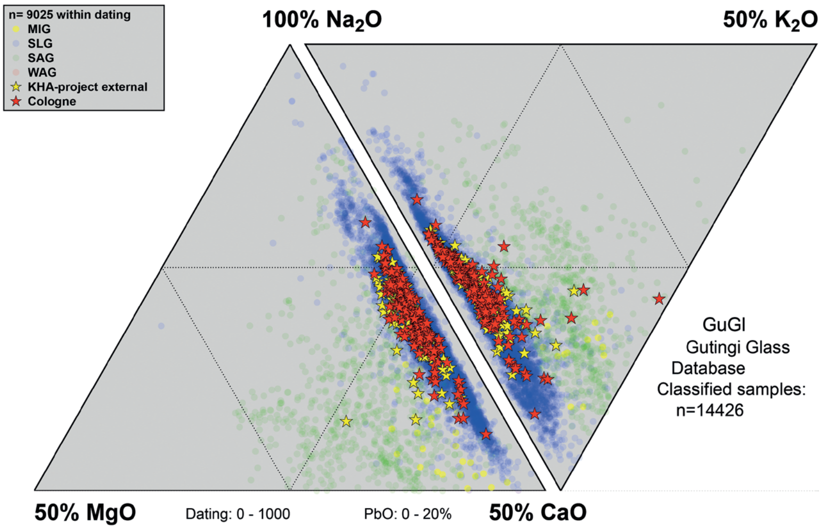


**Fig. 7** Double ternary diagram of the systems $Na_2O$-CaO-MgO and $Na_2O$-$K_2O$-CaO (mass-%), 50-100 % of the $Na_2O$-corner. Glass from Cologne (red asterisks) and other investigated sites within the »Harbour project«. – (Diagram A. Kronz, Göttingen)

Jackson (2010) also already noted that Levantine glass was used for higher quality glassware in Britain and was also less recycled. In the element ratio variation diagrams shown, the transitions of the glass analysed from the Tel-Aviv-Straße workshop to the HIMT-2 type are not quite clearly separated (**fig. 6**). However, the generally lower absolute contents of the heavy mineral-derived elements Fe and Ti make it possible to distinguish them from the Hambach HIMT-2 group.

## Cologne Harbour

Antique and Early Medieval glass from Cologne Harbour (excavations Heumarkt and Kurt-Hackenberg-Platz)[57] is despite a few exceptions restricted to the SLG type (**fig. 1**). Only a few late dated Carolingian window panes are exceptionally WAG. They will be not discussed further here.

In **figure 7** glass samples from the Cologne and the comparative localities of the Harbour project are shown (restricted to the 50-100%-$Na_2O$-component corner of the ternary systems described in **fig. 1**). The potassium enriched »outliers« (ternary system $Na_2O$-$K_2O$-CaO) are glass from thin layers of crucible fillings, enriched in Al and K, low in MgO, hence contaminated glass and thus not interpreted as soda-plant-ash glass (SAG). To our knowledge, SAG cannot be confirmed in the Rhineland so far.

In the following, we try to trace the transition from Roman glass technology to the Early Middle Ages on the basis of the Rhenish glass workshops. As already evidenced by the burials of the numerous glass vessels in the Krefeld-Gellep cemetery, the Rhineland seems to be characterized by a continuity of glass use beyond the end of the Roman Empire. With the evidence of glass processing in the frankish settlement area at the harbour of Cologne, this also applies to the manufacture of glass objects. This raises the question of whether, in the times of enormous socio-cultural upheavals during the Migration Period, the supply of raw glass from the Levantine and Egyptian primary glassworks completely ceased and possibly only the abundant Roman glass was recycled? With the extensively investigated glass workshops in Hambach and the Tel-Aviv-Straße in Cologne, we now have a very good overview of the types of glass processed in the Rhineland in Late Roman antiquity. It should be noted that this production spectrum also largely corresponds to the types of glass in circulation in the region (in the chemical sense), as shown, for example, by the findings from Mayen and numerous other investigations of Roman utility glass[58].

If we now look at the chemical composition of the Frankish glass from the Cologne Harbour (**fig. 6**) under this aspect, the spectrum of glass types changes:

1. The original pure Sb- or Mn-decoloured Roman glass types can no longer be traced. The recycled mixed Roman-SbMn glass is only sporadically encountered.
2. HIMT-1 is almost completely extinct: A few unspecific finds, which can be identified as HIMT-1, could well be relicts of older material in the respective post-Roman dated strata, if they are not clearly to be addressed as Frankish objects. For a few finds from HIMT-1, which can clearly be attributed to Frankish glass processing, the reuse of Late Roman cullet must be postulated.
   The generally observed rapid decline of the HIMT-1 type (**fig. 4**) in the beginning of the 5$^{th}$ century can thus also be traced in the finds from the Cologne Harbour.
3. We assign the majority of all SLG of the Cologne port to the type HIMT-2.
   There is more variation in their compositions than we observe for other SLG production types. Good correspondences exist for age-matched samples to the Hasselsweiler glasses, while the Hambach HIMT-2 chemical signature is less represented. Roman inherited glass seems to play hardly any role in the stock of 5$^{th}$/6$^{th}$ century Cologne Harbour glass, although they could certainly have been recycled in masses from old glass. This raises the question whether the broad spectrum of HIMT-2 types found in the Cologne harbour might represent a mixture of various other glass types (Roman + HIMT-1) and thus always recycled glass. This question will be addressed in the following. A very clear chemical demarcation of the Roman types to HIMT-2 is not possible for the Cologne glasses, rather the transition is a continuous one. In contrast, HIMT-1 is clearly defined. Within the Cologne HIMT-2 type, chemical variants can be traced which, with some caution, correspond to the subdivisions between the Foy groups »série 3.2« and »série 2.1/2.2«. However, a complete congruence is not given. Ultimately, it remains to be examined to what extent analytical systematic deviations also complicate a group assignment here. In particular in the ratio/ratio diagrams of **figure 5** and **6** the analytical errors are quite two to three times the symbol sizes used in the diagrams.
4. The Levantine glass type, which was strongly represented in Europe around the 6$^{th}$/7$^{th}$ century, is found only sporadically in the Cologne Harbour glass. However, this period is also not particularly well represented in the find material of the Cologne harbour (**fig. 3; 4**). Nevertheless, Levantine Glass seems to have reached the Cologne market only in smaller quantities, while it is found in larger proportions in the other trading locations studied.
5. Egypt types are nearly absent. Only three glass analyses of two Reticella decorated hollow jars and one funnel beaker of the 9$^{th}$ century can be assigned to the Egypt-2 type. This is out of proportion to the number of specimens dated to the 8$^{th}$/9$^{th}$ century. Consequently, the Cologne area seems to have been hardly supplied with glass from the Egypt groups. Since no processing remains of Egypt-type glass were found, it can be concluded that this glass was not processed in Cologne and that the finds are imports of finished goods.

## DISCUSSION: RHENISH GLASS

In the Rhineland, the general development of glass types in Roman times hardly differs from the other regions of the northwestern Roman Empire, as shown, for example, by the detailed studies of British sites[59]. In a very comprehensive study J. Komp has analysed Roman window glass from the Rhine area by means of major and trace element analysis[60]. Of 287 samples analysed from the dating period from the 1$^{st}$ to the 4$^{th}$ century, 271 are from SLG, of which 219 samples are window glass. The analysis of window glass offers the advantage that it is often produced in larger batches, is less susceptible to production-related contamination, and was hardly intentionally coloured, at least in Roman times. Of disadvantage is the very vague dating, which, since window glass is quite nonspecific, can only be given with quite wide variation by the author. Komp's Roman window glass of the Rhineland and surroundings represents the Roman types and illustrates again the great chemical uniformity of Roman glass. By means of hierarchical cluster analysis, however, she distinguished 10 composition-specific groups. The window glass can be assigned predominantly to the Roman and early Levantine types and only in smaller quantities to HIMT-1 or HIMT-2. Glasses with manganese contents in the geogenic background concentration range are rarely found in Roman window glass of the Rhineland, while a large number do not contain intentionally added antimony. The composition of Roman window glass in the type spectrum is thus quite different from that of the circulating glass vessels.

In the Roman workshops of Cologne, the typical Roman-type glasses decolourized with antimony, manganese or both elements are found. In contrast, at Hambach, apart from only a few examples of early Levantine glass, almost exclusively the two HIMT types can be found. The Hambach glassworks are typical for the mass production of glassware, such as bottles and simple vessels, hence the use of larger furnaces, while the small workshop (*officina*) of the Cologne Tel-Aviv-Straße produced sophisticated and expensive glassware for the luxury market.

With the beginning of the 2$^{nd}$ quarter of the 5$^{th}$ century, the HIMT-1 glass seems to disappear almost completely from the market in Cologne. Only isolated pieces, including raw glass, mostly dated to the 5$^{th}$ century, appear in the finds of the harbour. It is possible that this is old glass that was processed in the workshops. The dominant glass in Cologne from the Frankish period onwards is HIMT-2 in variable chemical composition. Levantine-type glass plays only a minor role in the Rhineland in Late Antiquity, and its Early Medieval counterpart is hardly present in the glass of the Cologne Harbour, with a few exceptions.

### Is all of the HIMT-2 glass from the Cologne Harbour recycled and a result of glass blending?

There is now no doubt that glass has been recycled everywhere and at all times, at least in secondary workshops, because reuse saves energy and costs and is technically simple to carry out[61]. Glass recycling can obscure its original origin if glass from different primary sources has been mixed and remelted. Recycling appears to be easily identified by unusually high levels of those trace elements that are intentionally added to the mixture to achieve desired effects such as colouration or opacification. If the concentrations are above the assumed content of the original raw materials (the »geogenic background value«), e.g., a few 10 µg/g for Co, Cu, Sn, Sb, Pb, but too low to achieve the desired effect, recycling seems to be quite obvious. However, it is not easy to set general thresholds, and of course the amount of recycled glass can vary in a wide range[62]. Moreover, in coloured glass, where the concentration of colouring elements is high anyway, it is impossible or at least difficult to judge whether a particular glass contains recycled fractions. However, the specific combination of »useful« and »unnecessary« elements may indicate recycling. Also, a mass balance of all elements can prove or disprove the use of various recycled components to produce a particular glass. In **figure 8a**, we reviewed the percentages of glass that exhibited recycling indicators compared to glass that appeared to have no recycling or, at best, low recycling percentages in a timeline for the entire database, where trace values were available. We then reviewed each production group separately. Any glass containing higher levels of a particular element believed to have been intentionally added to produce a coloring or opacifying effect is not included in this chart. Only the Roman types -Mn, -Sb, -SbMn, HIMT-1, HIMT-2, and Levantine are shown. A »recycled« glass is defined if any of the elements Co, Cu, Sn, Sb, Pb are between the

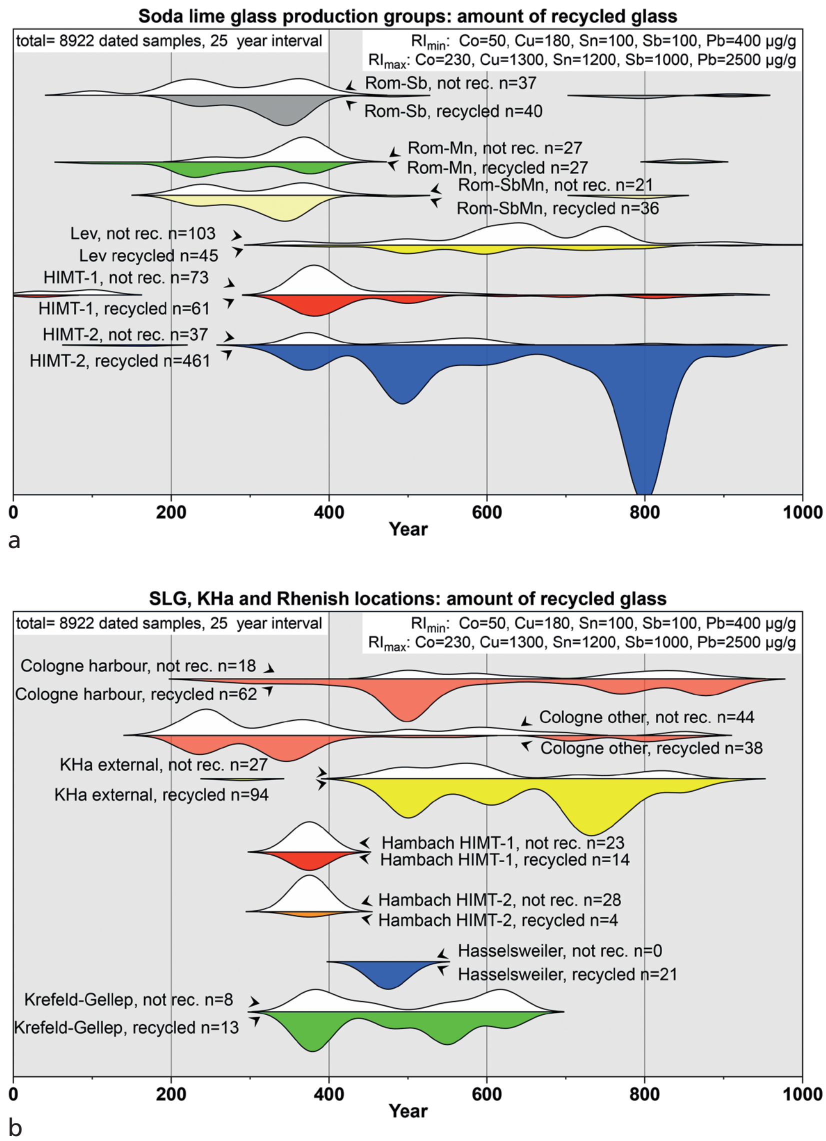


**Fig. 8** **a** Ratio of recycled to non-recycled glass for different SLG types. Point density functions, symmetrical distribution assumed, GuGI database of 8922 classified and dated glass samples, Roman-Sb, -Mn, -SbMn, Levantine and HIMT types only. – **b** Ratio of recycled to non-recycled glass from different Rhenish sites, regardless of SLG production type. – (Diagrams A. Kronz, Göttingen).

respective »recycling index« $RI_{min}$ and $RI_{max}$ values, which here range from 50 to 400 µg/g for the minimum values and 230 to 2500 µg/g for the maximum values (lower coloured portion of each distribution pattern in **fig. 8a**). When all values are below $RI_{min}$, the glass is classified as »not recycled« (upper white areas in the graph). No filters were applied in the graph for object types (e.g., window glass, hollow glass), geographic origin or for archaeological sites (such as primary workshops, settlements, cemeteries, secondary workshops). What we can learn from **figure 8a** is that almost all SLG production groups have significant amounts of recycled glass. However, the amount of recycled glass is different for each group and each cumulation over time. Of course, we recognize that the distribution patterns represent only a snapshot. First, accurate trace element analyses compared to the total amount of glass in our database are rare, so the results are likely biased by the few projects where trace elements were analysed. Second, the picture could change if the grouping of SLG production types is defined differently in the future. As mentioned earlier, we have grouped a large set of glass data as »HIMT-2« in this work, although we note compositional changes within this group over time. Roman Sb glass begins in the 1st and 2nd centuries with a purely non-recycled group. The proportion of recycled glass increases within this group until its decline at the end of the 4th century. Three maxima of occurrence can be observed.

Roman Mn behaves somewhat strangely: of the two maxima in the 3rd and 4th centuries, the first has an unexpectedly high proportion of recycled glass. We cannot explain this behavior, but the number of samples belonging to the Roman-Mn group is very small here anyway. It seems that fresh primary glass entered the markets in the 4th century. Roman-SbMn is a mixed glass anyway. In the diagrams for Roman-Sb and Roman-SbMn, we have of course not included Sb and Pb as recycling indicators. Therefore, only the Co, Cu and Sn concentrations were used here as an indication of recycling to check whether pure raw glass or partially coloured cullet was mixed. When primary glasses of Roman-Sb and Roman-Mn were mixed, they appear here as »not recycled«. However, we observe an increase in recycled content with time.

The glass identified here as Levantine is consistent over a long period of time (though we would define the 4th century group as »Levantine«). Its appearance on the European markets is always characterized by a high proportion of fresh raw glass, especially from the 2nd half of the 6th century to the end of the 7th century.

It has already been mentioned that HIMT-1 seems to have existed for a relatively short time. **Figure 3** supports this assumption, but a large »tail« of the distribution pattern is evident by the end of the 9th century. HIMT-1 has a high proportion of glass from the beginning, which is referred to here as »recycled«. It could be that the uniform thresholds used for each glass type are different for HIMT-1. The ratio of »recycled« to »non-recycled« is very sensitive to Pb concentration for HIMT-1. It is likely that even fresh HIMT-1 contains somewhat higher Pb concentrations, although we set the $Pb_{min}$ threshold to a relatively high value (400 µg/g). If a Mn-Fe ore (and other elements such as Ba, Sr) was intentionally added for staining purposes, as noted by Freestone et al. (2018), Pb could also be somewhat higher concentrated.

In almost all diagrams in which element concentrations or element fractions are plotted against each other, the glass group HIMT-2 is sandwiched between the other »primary« end members HIMT-1 and the Roman or Levantine types. Therefore, it would be reasonable to assume that HIMT-2 is not an independent primary production group at all, but always a mixture of, for example, Roman or Levantine types and HIMT-1. Jackson and Foster (2014) also discuss whether HIMT-2 could be a mixture of HIMT-1 and the Roman colourless glasses. This is the case, for example, in the diagrams with different ratios of elemental oxides $Na_2O$, $Al_2O_3$, $SiO_2$, $Fe_2O_3$, $TiO_2$ (**fig. 5; 6**).

The distribution pattern for all glasses classified as HIMT-2 in the database shows three or even four maxima (**fig. 8a**). Among all SLG production groups, HIMT-2 has the highest proportions of recycled glass. Nevertheless, it does not consist entirely of remelted glass cullet. If we assume that no fresh material was added in the primary workshops, we observe an influx of fresh primary glass in the 4th and late 6th centuries, but also intensive recycling in the 4th century and around 500.

HIMT-2 glass in Carolingian times consists almost entirely of recycled material. It appears that almost all Carolingian HIMT-2 is a mixture of possibly inherited glass fragments from various groups.

If we analyse the degree of recycling for the Rhenish glass, we find a high proportion of recycled glass for the Cologne Harbour (**fig. 8b**). Nevertheless, we also recognize proportions of fresh glass for the 5th and 6th centuries. The proportion of non-recycled glass in the 8th-9th centuries is also unusual here, but the small number of samples is hardly significant. In the other Cologne sites (here with emphasis on the glass from the Tel-Aviv-Straße workshop) the fraction of fresh glass is significantly higher.

For the SLG external sites analysed in the project, the fraction of glass without recycling markers is only about 22 %, which proves that the peripheral sites, such as at the eastern North Sea and southern Baltic Sea coasts, e. g. Haithabu were mainly supplied with cullet that was mixed and further processed.

The HIMT-2 from Hambach is obviously made from original raw glass, while HIMT-1 has higher RI, but the latter is not necessarily certain for this glass type, as discussed above. The glass produced at Hasselsweiler (2nd half of the 5th century) seems to have been produced exclusively in secondary use of recycled glass, while the glass from Gellep shows a balanced ratio of recycled to fresh glass (**fig. 8b**).

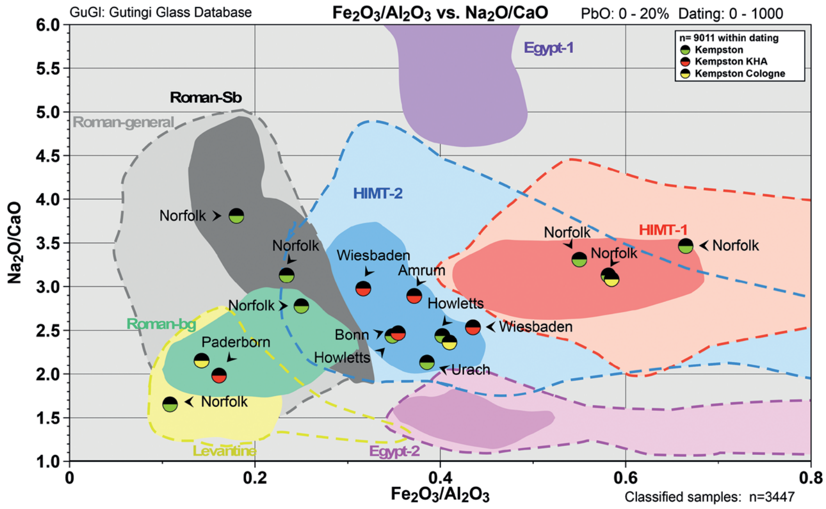


**Fig. 9** Chemical composition of conical beakers, type Kempston. – (Diagram A. Kronz, Göttingen).

The Hambach glass in particular makes it clear that HIMT-2 cannot generally be a mixture of other types of glass. If the HIMT-2 glass from Hambach should represent a mixture (conceivably a mixture of the original Roman-Mn and Roman-SbMn types and the abundantly detected HIMT-1 glass, continuous mixed series would be expected here. But this is clearly not the case: Especially the HIMT-2 glass is very clearly separated from the HIMT-1 in the case of Hambach and only isolated samples are not clear in their assignment (**fig. 5**). Obviously, there are clear and different sources of origin for these raw glasses.

## OBJECT STUDIES: KEMPSTON BEAKER AND CLAW BEAKER

From the variety of vessels, which have a specific and well assignable design, two Early Medieval »leading forms« are considered here in more detail. These are firstly the so-called Kempston beaker (Early Merovingian) and secondly the claw beaker (Early and Mid Merovingian). These are relatively well datable vessel types, whose typology and making style may allow the conclusion that they could have come from singular workshops. Similar questions have been formulated and investigated by Rehren and Brüggler (2015) for Late Antique »Helle« type glass vessels. Kempston beakers are pointed conical beakers with loop decoration. Their distribution is concentrated in southern England and the Rhineland[63].
Chemical analyses of Kempston-type beakers are sparse in the literature[64]. While Evison (2008) considers a production of the Kempston beakers due to their accumulation in England, she also recognizes a possible production in different workshops, also on the continent, with regard to the distribution of finds. The rather large chemical variability of the beakers seems to confirm this finding (**fig. 9**). However, Kempston beakers are chemically quite variable even within a single site (Cologne, Paderborn, Spong Hill) so that they could be considered as imports rather than belonging to one workshop. While the $TiO_2/Fe_2O_3$ ratios of all Kempston beakers are still relatively uniform, other element ratios vary in such a way that the Kempston beakers are distributed among the glass types Levantine, HIMT-1 and HIMT-2. Here the Spong Hill (North Elmham, Norfolk) beakers stand somewhat outside the typical variation fields, though this may also indicate an analytical problem with this older paper[65]. But also a Cologne beaker, two from Paderborn and the one Dortmund specimen[66] deviate from the central group of HIMT-2 glasses, although they were analysed with uniform methods. As far as trace element data are available, the recycling indicators for the Kempston beakers from both the Cologne and the external sites are quite high. This makes it difficult to prove whether the Kempston

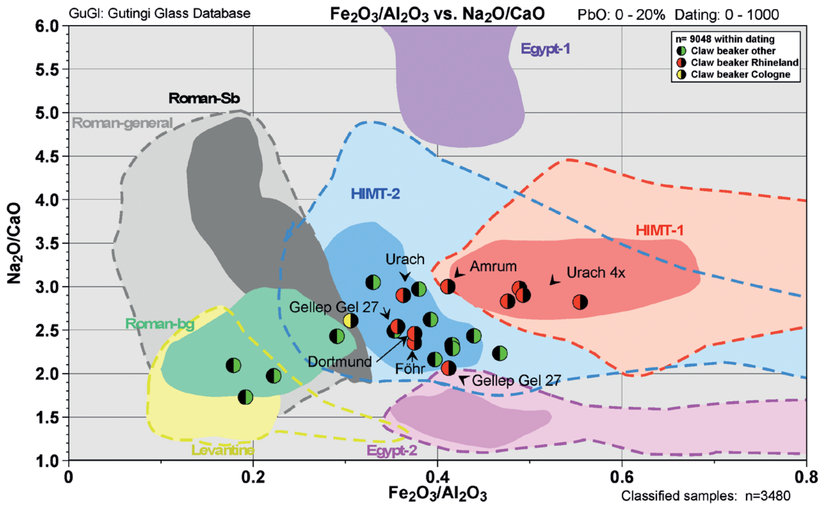


**Fig. 10** Chemical composition of claw beakers. – (Diagram A. Kronz, Göttingen).

beakers were made in a singular workshop. T. Rehren and M. Brüggler (2015) recognized the same problem for »Helle« type vessels: they found that all Roman-type and about half of the glasses classified as HIMT-1 contained high proportions of recycled glass. Some Kempston-type beakers are concentrated in the HIMT-2 field within a narrower range that may suggest at least a single workshop grouping. These are two Cologne specimens, the Wiesbaden beakers, Amrum, Bonn, Howletts and with the restriction of missing $TiO_2$ values also to some extent the specimens from the Runder Berg near Urach[67]. Very probably we record with this concentration a Rhenish workshop, possibly even in Cologne.

## Claw beaker

Claw beakers represent a particular elaborate Early Medieval glass form that was common in Central Europe and England[68]. Predominantly they are dated from the mid 5$^{th}$ to the 7$^{th}$ century with a focus around 500-530[69]. Fragments of claw beakers dated around 750-800 (Borg and from Witsum on the island of Föhr [KHa 165]) are either inherited old glass (Witsum) or the correct determination of the fragments is doubtful. With our investigations a total of 24 analyses are now available, which include besides one Cologne specimen also fragments from Krefeld-Gellep, the Runder Berg near Urach, the North Sea islands, Borg (Lofoten)[70] and England[71] (**fig 10**). We also encounter the problem in the comparative analyses from the literature data that important data are missing (no $TiO_2$ analyses for the Urach beakers) or, in the case of the SEM-EDX analyses, are subject to larger analytical uncertainty. Overall, however, the claw beakers appear much more uniform in chemical composition than is observed for other vessel forms, so one might well assume a singular manufacturing site. However, the longer period of their persistence, variations in shapes, different quality and variety of colourations rather indicate a popular model produced in several workshops. Chemically, a core group again corresponds to the HIMT-2 type. Higher proportions of recycled glass must also be assumed for some claw beakers, as far as can be judged for intentionally coloured glass. I. Freestone notes for the »Saxon« type vessels the higher $K_2O$ and MgO contents[71], which he attributes to an increasing intentionally added wood ash content. This is not yet the case for the English claw beakers (»Period-1«), which is indicative for the processing of raw glass.
As already mentioned, the double analysis of the claw beaker from Krefeld-Gellep (Gel-27) is chemically almost identical to the Amrum specimen (KHa 137). There is also a close chemical match to a specimen from

Castle Eden, which confirms V. Evison's observation. She has already noted the specimen as being similar to the Gellep beaker. The Urach claw beakers show a higher Fe-Al ratio. Likewise, an olive-green vessel from Taplow cannot be assigned to the core group due to lower $TiO_2$ and MgO contents and still has a Roman or Levantine signature. V. Evison, in her comprehensive study on the claw beakers, refers to the accumulation of this type of vessel in the county of Kent and concludes that it was produced in this region and exported to the continent[73]. Whether the clustering of the claw beakers in southeast England actually suggests origin from there cannot be judged here. From the comparative interpretation of the chemical analyses it can be concluded that the majority of the claw beakers were made from a raw glass of a uniform source, which as HIMT-2 corresponded to the typical glass of the time.

## CONCLUSIONS

The investigations of the glass from the Cologne Harbour have shown that in the Rhineland the manufacture of soda lime glass from Roman times exhibited a remarkable continuity up to the Carolingian period. While in the peripheral regions of the empire other types of glass, here especially the WAG, were already introduced from the end of the 8$^{th}$ century, in the Cologne area SLG could be obtained without any problems. From this a shortage situation can be derived for the peripheral workshops, which forced the glass craftsmen in the monasteries of Fulda or Lorsch and the Palatinates of Paderborn, Werla to first obtain a well workable glass predominantly by mixing WAG and SLG. The shortage finally led to the exclusive production of WAG. The Rhineland still had access to imported raw glass of the SLG type in Late Roman and Early Medieval times. However, there is no question that this glass always contained a proportion of recycled waste glass. The proportions of recycled waste glass nevertheless fluctuated over the centuries, and especially in the final phase of SLG, waste glass, very likely even recovered from Roman ruins, seems to have played a dominant role at the end of the 8$^{th}$ and the 9$^{th}$ century.
Some of the post-Roman production types appear only very subordinately in the glass of the Cologne Harbour. These include the Early Medieval Levatine types and the Egypt types. Egypt-2, which as a separate production group of Egyptian provenance supplied parts of Europe from the 9$^{th}$ century onwards, can be recorded in large proportions at Haithabu, for example (Kronz et al. 2016), but is not represented in the Cologne excavations, with a few rare exceptions.
For the Cologne Harbour, HIMT-2 in its specific composition is the dominant glass type. It is certainly not an exclusive mixed glass of Roman, Levantine, or HIMT-1 types. Mixing tendencies of compositions between potential end-members are lacking, and several other chemical features confirm that HIMT-2 with its time-varying variants is rather a distinct raw glass group. This thus proves a still existing raw glass supply from the eastern Mediterranean area in the Early Middle Ages. Its distribution to peripheral smaller processing centers in the North- and Baltic Sea area can be traced with our work, even if a direct proof of provenance succeeds only in rare matches.

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## Notes

1) Nenna et al. 2014. – Rehren/Brüggler 2020. – Freestone/Ponting/Hughes 2002. – »Primary workshops« is understood here as the production of glass from raw materials, »secondary workshops« process raw glass and/or recycled waste glass into glass objects. If appropriate, »tertiary workshops« can also be defined as workshops that exclusively process cullet and do not have any special requirements in terms of technology and equipment. These are, for example, small-scale artisanal kilns for bead production.

2) For an overview see Freestone 2021.

3) Examples in Foy/Nenna 2003. – Foy et al. 2003. – Extensive analyses of such a cargo, which however did not consist of raw glass but cullet, were carried out by Silvestri/Molin/Salviulo 2008 on the glass cargo of the wreck of the Iulia Felix.

4) See discussions and distribution of Romano-Medieval glass workshops, e.g. Päffgen/Wedepohl 2004; Grünewald/Hartmann 2014.

5) Wedepohl 2003. – Henderson 2016, and references therein.

6) Wedepohl 2003. – Wedepohl/Simon/Kronz 2011.

7) Henderson 1985; 2016, and references therein. – Wedepohl 2003. – Shortland et al. 2006.

8) Schibille et al. 2011.

9) The database is termed »GuGi« (Gutingi Glass).

10) Freestone/Gorin-Rosen/Hughes 2000. – Freestone/Wolf/Thirlwall 2005. – Foy et al. 2003 and extensive discussions therein; cf. Rehren/Freestone 2014.

11) Wedepohl/Simon/Kronz 2011. – Brems/Degryse 2014.

12) Wedepohl/Baumann 2000. – Freestone et al. 2003.

13) Cf. Silvestri/Molin/Salviulo 2008, recycling index.

14) Freestone et al. 2015. – Freestone 2021.

15) Wedepohl/Winkelmann/Hartmann 1997.

16) Van Wersch et al. 2016.

17) See e.g. Fulda monastery: Kind/Wedepohl/Kronz 2003. – Lorsch monastery: Sanke/Wedepohl/Kronz 2002; 2003. – North-East France: Pactat et al. 2017. – Haithabu: Kronz et al. 2016

18) Some of them are used in this work, partly modified. Significant fundamental work on these subtypes has been done by: Freestone/Gorin-Rosen/Hughes 2000. – Freestone/Wolf/Thirlwall 2005. – Freestone/Hughes/Stapleton 2008. – Freestone et al. 2018. – Foy et al. 2003. – Foster/Jackson 2009; 2010. – Ceglia et al. 2015. – Schibille/Sterrett-Krause/Freestone 2017. – Schibille et al. 2019.

19) See e.g. Foster/Jackson 2010. – Gliozzo 2017.

20) About the mixing of Roman-type glass: Freestone et al. 2015.

21) Freestone/Gorin-Rosen/Hughes 2000. – Freestone et al. 2003.

22) Jackson/Foster 2014. – Phelps et al. 2016.

23) See e.g. Foster/Jackson 2009.

24) Gratuze 1988. – Gratuze/Barrandon 1990. – Summarizing: Schibille et al. 2019.

25) Freestone 1994.

26) Ceglia et al. 2015.

27) Summarized in Freestone et al. 2018. – Comparative definitions in: Nenna et al. 2014.

28) Group »E« = HIMT-1, Mirti/Casoli/Appolonia 1993.

29) Among others Foster/Jackson 2009; Gliozzo et al. 2019.

30) Freestone et al. 2018, fig. 8.5.

31) Freestone et al. 2018.

32) Brill 1999.

33) Cf. contributions of Dodt, Messal/Kronz, Majchczack/Kronz und Segschneider, this volume.

34) For Hambach see Rehren/Brüggler 2020.

35) First analyses by Wedepohl in: Gaitzsch et al. 2000.

36) Analyses in: Päffgen/Wedepohl 2006.

37) Analyses in: Wedepohl/Pirling/Hartmann 1997.

38) Rehren/Brüggler 2015.

39) Grünewald/Hartmann 2014.

40) Koch 1987. – Analyses by Czygan 1987.

41) Figs. 5a, 6a based on Ceglia et al. 2015; figs. 5c, 6c based on Schibille/Sterrett-Krause/Freestone 2017.

42) See e.g. Gliozzo et al. 2019.

43) Gaitzsch et al. 2000. – Brüggler 2009.

44) Comparatively: Grünewald/Hartmann 2014.

45) Wedepohl in: Gaitzsch et al. 2000.

46) Rehren/Brüggler 2020 and analysis data in this study.

47) Wedepohl/Baumann 2000.

48) Methodology in Gaitzsch et al. 2000: The reason for these generally too high iron contents can no longer be determined exactly. However, they are very probably due to the contamination of the larger quantities of sample powders required for the bulk chemical analysis (X-ray fluorescence analysis), since no firm mathematical relationship between the iron contents of the earlier analyses and the data of Rehren/Brüggler 2020 (s. note 46) or this study could be established.

49) Päffgen/Wedepohl 2004.

50) Wedepohl/Pirling/Hartmann 1997.

51) Cf. Segschneider, this volume. Notes on the term »batch« cf. Freestone/Price/Cartwright 2009.

52) Hartmann/Grünewald 2010. – Grünewald/Hartmann 2014. We would like to take this opportunity to thank S. Hartmann, M. Grünewald, Römisch-Germanisches Zentralmuseum Mainz, and S. Greiff, now University of Tübingen, for kindly providing the data.

53) See Dodt, this volume. – Dodt et al. 2018b.

54) For an overview see Grünewald/Hartmann 2014.

55) Cf. Schäfer, this volume.

56) Cf. Schäfer, this volume. The archaeological results will be reported in a doctoral thesis by E. M. Hetzel.

57) Dodt, this volume.

58) Gaitzsch et al. 2000 with Roman comparative samples: Barrel jugs, squared bottles.

59) Foster / Jackson 2009; 2010. – Jackson / Foster 2014.

60) Komp 2006; 2007.

61) For the Rhineland: Grünewald / Hartmann 2015.

62) First definition of a recycling index (RI) in: Silvestri / Molin / Salviulo 2008.

63) Evison 1972. – Dodt, this volume.

64) Sanderson / Hunter 1982. – Hunter / Sanderson 1982. – Freestone / Hughes / Stapleton 2008.

65) Sanderson / Hunter 1982. – Hunter / Sanderson 1982. – Analysis methods: Neutron Activation Analysis (NAA) and Energy Dispersive X-Ray Fluorescence (EDXRF).

66) Dortmund: unpublished data. – Kronz, forthcoming project.

67) Czygan in Koch 1987: Unfortunately, $TiO_2$ concentrations were not analysed in this very significant group of Early Medieval glasses, so that the Urach samples cannot be represented in some variation diagrams.

68) See distribution map in Evison 2008.

69) Evison 2008.

70) Henderson / Holand 1992.

71) Freestone / Hughes / Stapleton 2008.

72) Freestone / Hughes / Stapleton 2008.

73) Evison 1982.

74) EPMA: Electron Probe Microanalysis; LaICPMS: Laser ablation Inductive Coupled Plasma Mass Spectrometry.

75) Publications within the framework of the »Cologne Harbour project«: Dodt 2019. – Dodt / Kronz 2021. – Dodt / Kronz / Simon 2018; 2019; 2021. – Dodt et al. 2018.

76) P & H developments, 24 Shackleton Close, Whitby, YO21 1NR, North Yorkshire, UK.

77) Jarosewich / Nelen / Norberg 1980ab.

78) There exist different batches for the olivine San Carlos sample. See discussion in Fournelle 2011. Here we use a »commercial« single crystal, which is not the original USNM, NMNH111312-44 sample (Jarosewich / Nelen / Norberg 1980ab). But, our SC-olivine was extensively analysed and found to be homogeneous. We crosschecked the material with original NMNH111312-44 to obtain reliable values.

79) Sanidine Eifel is a Lab-internal RM. The gem quality single crystal was extensively checked by different methods and tested for homogeneity. The values give a perfect mineral formula stoichiometry.

80) Donovan et al. 2002; 2003. – Jarosewich / Boatner 1991.

81) GZG: Lab internal RM, tested for stoichiometry and impurities.

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## *Zusammenfassung*

Die Analyse (EPMA, LaICPMS) zahlreicher archäologisch gut datierter Gläser aus römischem und frühmittelalterlichem Kontext aus Köln und weiteren Handelsorten erweitert unsere Erkenntnisse zur Erzeugung von Gebrauchsglas im Rheinland. Die fränkischen Werkstätten im Bereich des Hafens von Köln führen die Glasverarbeitung aus römischer Zeit fort. Mit der erneuten Analyse der Gläser des Gräberfeldes Krefeld-Gellep (römisch-fränkisch), der Glaswerkstatt Hasselsweiler (fränkisch) dem Glas der Hambacher Hütten (römisch) liegen nun verbesserte umfangreiche Datensätze auch zu Spurenelementen vor. Die aus inzwischen zahlreichen Arbeiten bekannten chemischen Variationen innerhalb der einzelnen Produktionsgruppen des Soda-Kalk-Glases (SLG) können für die untersuchten archäologischen Glasbefunde weitgehend bestätigt werden. Die Produktionsgruppen werden in ihrem zeitlichen Kontext sowohl allgemein für eine große Anzahl in einer Datenbank vorhandener Analysen (n = 15270, Stand Juni 2022), als auch für die Gläser im Rheinland, besonders Köln, vorgestellt. Dabei werden auch Kriterien genutzt, um den Anteil rezyklierten Glases in den jeweiligen Zeitabschnitten zu ermitteln. Römisches Gebrauchsglas wurde in Hambach als Massenware überwiegend aus den SLG-Typen HIMT-1 und HIMT-2 (Foy série 3.2) gefertigt, während Antimon- und Mangan-entfärbtes Glas (Typ »Roman«) in Kölner Werkstätten zur Erzeugung hochwertigerer Glaswaren diente. In Hambach ist der Anteil von rezykliertem Altglas recht gering, sodass davon ausgegangen werden kann, dass überwiegend Rohglas als Ausgangsmaterial verwendet wurde. Im 5./6. Jahrhundert ist der Anteil von Altglas in den Werkstätten des Kölner Hafens sehr hoch, Hasselsweiler verarbeitete wohl ausschließlich Altglas. Für das im Kölner Hafen verarbeitete Glas gibt es Anteile ohne Rezyklierungs-Anzeiger, sodass ein Warenfluss von frischem Rohglas aus der Mittelmeerregion auch für die nachrömische Zeit zu fordern ist. Dieser Warenstrom bestand für das Rheinland überwiegend aus einer Produktionsgruppe, die wir als HIMT-2 zusammenfassen. Die Glastypen HIMT-1 und die reinen Roman-(Sb-Mn) Typen verschwinden in den ersten nachrömischen Jahrhunderten relativ rasch, sind aber noch in wenigen Fällen zu beobachten. Das Levantine-1 hat nur eine untergeordnete Bedeutung für den Kölner Hafen und ist, wie HIMT-1, in nachrömischer Zeit fast ausschließlich als Glasrezyklierung bzw. als ererbtes römisches Glas zu interpretieren. Glas der Egypt-Typen tritt nur in wenigen Ausnahmefällen auf, sodass davon ausgegangen werden kann, dass dieser Produktionstyp in den Kölner Werkstätten selbst nicht verarbeitet wurde. Bis in das 9. Jahrhundert dominiert das SLG die Glaswerkstätten im Rheinland, während in Gegenden vor allem rechts des Rheines bereits erstes Holzascheglas hergestellt und verarbeitet wird. Allgemein steigt der Anteil von genutztem Altglas in karolingischer Zeit stark an. Dabei tauchen auch im Rheinland im 8./9. Jahrhundert Gläser mit Roman- oder HIMT-typischer Zusammensetzung wieder auf. Das deutet auf die beginnende Verknappung des Soda-Kalk-Glases hin, da man größere Mengen Altglas aus den antiken Ruinen bezog.

## *Summary*

The analysis (EPMA, LaICPMS[74]) of numerous archaeologically well-dated glasses from Roman and Early Medieval contexts from Cologne and various trading sites extends our knowledge of the manufacture of commodity glass in the Rhineland[75]. Frankish workshops in the area of the harbour of Cologne continued the glass processing from Roman times. With the renewed analysis of the glass from the Krefeld-Gellep burial site (Roman-Frankish), the Hasselsweiler glass workshop (Frankish), the glass from the Hambach glass workshops (Roman), improved extensive data sets are now also available on trace elements. The chemical variations within the individual production groups of soda lime

glass (SLG), which are known from numerous studies in the meantime, can be confirmed to a large extent for the examined archaeological glass records. The production groups are presented in their temporal context both in general for a large number of analyses available in a database (n = 15 270, status June 2022), as well as for the specimens of glass in the Rhineland, especially Cologne. Criteria are also used to determine the proportion of recycled glass in each time period. Roman utility glass was manufactured in Hambach as mass-produced glass mainly from SLG types HIMT-1 and HIMT-2 (Foy série 3.2), while antimony- and manganese-decoloured glass (»Roman« type) was used in Cologne workshops to produce higher-quality glassware. In Hambach, the proportion of recycled waste glass is quite low so that it can be assumed that mainly raw glass was processed. In the 5th/6th century the proportion of recycled glass in the workshops of the Cologne Harbour is very high, Hasselsweiler probably processed only waste glass. For the glass manufactured in the Cologne Harbour, there are proportions without recycling indicators, so that a flow of fresh raw glass from the Mediterranean region can also be postulated for the post-Roman period. This flow of goods (glass supply) consisted for the Rhineland predominantly of a production group, which we summarize as HIMT-2. The glass type HIMT-1 and the pure Roman (Sb-Mn) types disappear relatively quickly in the first post-Roman centuries, but are still observed in a few cases. Levantine-1 has only a minor importance for the Cologne port and, like HIMT-1, is to be interpreted in post-Roman times almost exclusively as glass recycling or as inherited Roman glass. Egypt-type glass occurs only in a few exceptional cases, so that it can be assumed that this production type was not processed in the Cologne workshops themselves. Until the 9th century, SLG dominates the glass workshops in the Rhineland, while in regions mainly to the right of the Rhine, the first wood ash glass is already produced and processed. In general, the proportion of recycled glass increases strongly in Carolingian times. Glasses with Roman- or HIMT-typical composition also reappear in the Rhineland in the 8th/9th century. This indicates the incipient scarcity of soda lime glass, as larger quantities of older glass were probably recovered from the ancient ruins.

## APPENDIX A1

Accelerating voltage: 15 kV
Beam current: 15 nA
Beam diameter: 22 µm

| Element/ Oxide | X-ray line | Analyser crystal | Primary RM | RM provider | Counting Time Peak [s] | Total Counting Time Back-ground |
|---|---|---|---|---|---|---|
| $SiO_2$ | K$\alpha$ | TAP | Wollastonite | Wo: Willsboro, New York, USA, P&H [76] | 15 | 10 |
| $TiO_2$ | K$\alpha$ | PET | $TiO_2$ | synthetic, Earth Jewelry Co. Japan, P&H | 30 | 30 |
| $Al_2O_3$ | K$\alpha$ | TAP | Anorthite | NMNH 137041, Great Sitkin Island, Alaska, USA [77] | 15 | 10 |
| $Fe_2O_3$ | K$\alpha$ | LIFH | Hematite | Rio Marina, Elba, GZG | 15 | 10 |
| MnO | K$\alpha$ | LIFH | Rhodonite | North Mine, Broken Hill, NSW, Australia, P&H | 30 | 30 |
| MgO | K$\alpha$ | TAP | Olivine | San Carlos, Arizona, USA 3) | 15 | 10 |
| CaO | K$\alpha$ | PET | Wollastonite | Wo: Willsboro, New York, USA, P&H [78] | 15 | 10 |
| $Na_2O$ | K$\alpha$ | TAP | Albite | Amelia Co., Virginia, USA, P&H | 15 | 10 |
| $K_2O$ | K$\alpha$ | PET | Sanidine | Volkesfeld, Eifel, Germany, GZG [79] | 15 | 10 |
| $P_2O_5$ | K$\alpha$ | PET | $ScPO_4$ | NMNH 16849 [80] | 30 | 30 |
| $SO_3$ | K$\alpha$ | PET | Baryt | GZG, Hunsrück, Germany [81] | 30 | 30 |
| Cl | K$\alpha$ | PET | NaCl | synthetic, Merck | 30 | 30 |
| SrO | K$\alpha$ | PET | $SrTiO_3$ | synthetic, Crystec, Berlin | 30 | 30 |
| BaO | L$\alpha$ | LIFH | Celsiane | Micro-Analysis Consultants Ltd. | 30 | 30 |
| CoO | K$\alpha$ | LIFH | NaCl | synthetic, Merck | 60 | 30 |
| CuO | K$\alpha$ | LIFH | $Cu_2O$ | GZG, J. Zang, Idar-Oberstein, Germany [81] | 30 | 30 |
| PbO | M$\alpha$ | PET | Cerussite | GZG, Tsumeb [81] | 30 | 30 |
| $SnO_2$ | L$\alpha$ | PET | Cassiterite | Hemerdon open cast, nr. Plympton, Devon, Engl., P&H | 30 | 30 |
| $As_2O_5$ | L$\alpha$ | TAP | AsGa | synthetic, Alpha Products, P&H | 30 | 30 |
| $Sb_2O_5$ | L$\alpha$ | PET | Sb | Koch Chemicals Ltd., Hertford, England, P&H Developments | 30 | 30 |

**Tab. A1** EPMA instrument and analytical protocol. – (Table A. Kronz, Göttingen).

## APPENDICES A2-A4

https://doi.org/10.11588/data/QRQSSZ

**Tab. A2** Analysed samples.

**Tab. A3** Major element composition, EPMA, oxides given in mass-%.

**Tab. A4** Trace element composition, elements given in µg/g.