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X-Ray Fluorescence as an analytical tool for studying the copper matrices in the collection of the Museum Plantin-Moretus

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Introduction

The collection of the Plantin-Moretus Museum consists of a large variety of historical typographical objects. Amongst them are sets of copper matrices, which are the 'dies' for casting lead printing letters ('type'). They are of foremost interest for the research of typography and have been studied thoroughly in the past decades, mainly through visual comparison and enduring research in the Plantin archives and books which were printed with type cast from the matrices. Until now, there has never been an attempt to apply analytical measuring techniques. The main reasons for this is the vast number of strikes and matrices (about 20000 pieces) and the fact that they may not leave the museum nor may they be damaged for destructive analysis. Also, the majority of matrices are made of copper and were never questioned towards possible material variations to differentiate them from each other. X-Ray Fluorescence (XRF) was investigated to provide analytical results on the copper compositions. Firstly, the basic idea for the use of XRF is explained and technically described. Secondly, historical and technical considerations on the use of copper for making matrices are addressed. Thirdly, the results are presented and discussed, including two case studies.

Motivation

Research on matrices can contribute to the knowledge of origin and history of typecasting and typefaces. It can help to define precise sources from which matrices for the types reached a printing office, in this case Plantin. Since the earliest records on hand printing, the use of steel punches to strike copper matrices is eminent according to the abundant presence of studies. Therefore it seems not very useful at first sight to try to analyse these pieces. However, copper was never completely free of trace metals, at least not until the 19th century when advanced chemical techniques strongly upgraded the copper purity. The reason for the trace metals lies in the way of unearthing the ore, the smelting processes and the refining techniques used at the time. Therefore it could be expected that each lot of 'pure' copper that was sold on the market had some trace metals incorporated. Since it is known that coppersmiths provided copper to the punch cutters / matrice maker in the form of rods which they had cut and hammered from large sheets or blocks[1], possible tendencies in the presence of trace metals could be found.

[1] (Fournier 1764)

The goal of this study, using XRF as an analytical technique, is to determine the alloy composition(s) used in each matrices set and to indicate possible alloy composition deviations of certain matrices within a set. Further, by performing extensive series of measurements on the matrices' collection, it could perhaps lead to the detection of possible tendencies in the copper composition used by a certain punch cutter to whom a matrices set is attributed. Similarly, possible tendencies of copper used in different locations and through the centuries could rise to the surface. A final result could be found with the identification of the punch cutter-matrice maker of non-attributed matrices sets, although this goal may be far beyond our reach. At least a complementary contribution to the extensive studies historians and typographers have executed over the last century is hopefully expected. As a start of this new trajectory, we concentrate in this paper on measuring with XRF as an analytical technique and the data retrieved from it.

Strikes and matrices

In hand press printing several subsequent stages and tools are needed to achieve a print on paper. Firstly, a design for a certain character was made considering style and dimensions (size, body or x-height of the letter). A complete alphabet of capital letters, lower case letters, numbers, accents, etc. is called a polis or fount. The main groups of styles (typefaces) are Blackletter, Roman and Italic. Secondly, from the design, each character had to be cut in steel to obtain a punch with the mirrored image. The steel punch was hardened and driven into a copper rod by striking it with a hammer. This imprint in the copper gives the 'strike', which produces a clear negative shape of the punch. However, because of the force that was used to drive the punch into the copper, it underwent deformations at the upper face and at the sides because of its plasticity. To make a strike suitable for further processing, it had to be justified. The justification is a process of filing down and smoothing all sides of the strike to fit in the casting mould, also called 'instrument'. When done, finally the matrice is ready to be placed into the moulding tool and metal can be cast into it, giving a reproduction of the steel punch model and is called 'type'. This casting process (typefounding) was done for each character as many times as the printer expected to need type for composing the pages of the book that had to be printed.

Analytical technique

Fig. 1: XRF measuring stand at the museum

X-Ray Fluorescence (XRF) analysis is a fast and non-destructive physical method for measuring elements in materials. It's use increased strongly in the last years because of the improved detector sensitivity and software. Comparable analysis studies are found in measuring archaeological bronzes.[2] In this research, the measurements were executed at the museum since the unique matrices may not leave the building for insurance reasons. The setup in situ is shown in Fig. 1. Behind the laptop with the operational control window, there is the measuring stand with the instrument (yellow-black) below and the shielded cabinet on top, where the matrices are placed into for measuring.

[2] (Nicholas and Manti 2014)

The instrument used was an Olympus-InnovX Delta Professional equipped with a Rhodium X-Ray tube (4 Watt) and a Silicon Drift Detector (SDD). The energy range was 0-40 keV and the measurement time for each object was 20 s Live Time with a measurement spot of 5 mm. The instrument mode was set to ‘Metal alloy’, no filter. The used software for calculating the quantitative data from the spectrum was the Innov-X Delta Advanced PC Software. Measurements were taken on the cleanest side of each piece; for the strikes this was the upper face, for the matrices this was mostly a side or the backside.

The principle of operation is based on the fact that X-rays, emitted from the instrument, irradiate the specimen to cause its elements to emit characteristic x-ray line spectra. The detector determines the energies of the emission lines and their intensities. Elements in the specimen are identified by their spectral line energies or wavelengths (plotted on the X-axis) while the intensities are related to the concentrations of the elements (plotted on the Y-axis). This is shown in Fig. 2, where the left image of a spectrum with linear scales illustrates the abundant presence of about 99% of copper (Cu). When a logarithmic scale is applied to the Y-axis (right image), smaller peaks become more visible, such as those for lead (Pb), antimony (Sb) and silver (Ag). Other elements measured were zinc (Zn), tin (Sn), iron (Fe), nickel (Ni) and arsenic (As). Since only very small amounts of these elements are detected in the measured matrices’ alloys and the fact that in the region of 15-35 keV (kilo electron Volt) the signal to noise ratio is not optimal, leaves the quantification of specific elements more difficult. Empiric calibration on standardized alloys is used for the evaluation of the obtained quantitative results.

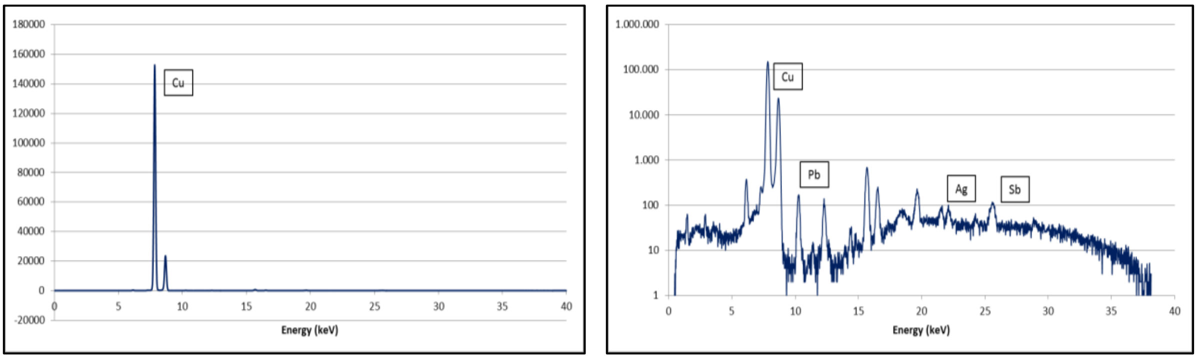

Fig. 2: XRF spectrum from a certain matrice; left with a linear Y-axis showing the abundant presence of Cu by the two distinctive peaks, while on the right with a logarithmic Y-axis, also the peaks for the metals with low concentrations become visible, i.e. Pb, Ag, Sb.

In measuring metals, attention must be given to the fact that XRF is a surface sensitive technique. Depending on the matrix of the metal, the depth of information is estimated to be maximal about 100 micrometres (µm)[3]. Surface dirt or corrosion are less dense since they are chemically bound with oxygen, carbon, sulphur, chlorides or other light elements which are not measured or quantified in the metal alloy measuring mode. Moreover, the measured matrices have no or at the most a thin oxide layer which should not affect the measurements.

The quantification of the elements has been verified with measurements on IMMACO standardized copper alloys. These alloys were designed to act as references for historical copper alloys.[4] The composition of the five IMMACO copper alloys is listed in Table 1. Since the measurements are on the limits of detection by the XRF instrument, the numeric results given in weight percent (wt%) for the elements, some slight deviations may be present. It also has to be remembered that XRF is a semi-

[3] (Nicholas and Manti 2014), p.2
[4] (Beldjoudi 2001)

quantitative technique where the values in wt% are derived through software calculations from the spectrum. The quantified XRF-results are shown in Fig. 3, showing the most prominent metals that are present in the matrices. The limit of detection of trace metals in the copper lies around 0.1 wt%. Smaller concentrations may not be picked up by the instrument nor are they taken into account in the results. For the purpose of the study at hand, we can conclude that the technique permits sufficient accurate data to detect tendencies, groups and deviations in alloy compositions.

Table 1: Composition of the five respective IMMACO standardized copper alloys, elements expressed as weight percent (wt%)..

IMMACO N°	Cu	Pb	Sb	Zn	Sn	Fe	Ni	Mn	As	S
1.7.52	76.50	9.00	0.50	6.00	7.00	0.20	0.10	0.20	0.20	0.30
2.7.54	93.40	0.20	0.50	0.00	0.20	0.20	0.00	0.20	5.00	0.30
3.2.24	90.20	0.20	0.70	0.10	7.00	0.30	0.50	0.30	0.20	0.50
4.1.41	78.80	10.00	0.30	0.10	10.00	0.10	0.30	0.10	0.30	0.00
5.9.2	81.10	0.40	0.00	15.00	2.00	0.50	0.20	0.40	0.10	0.30

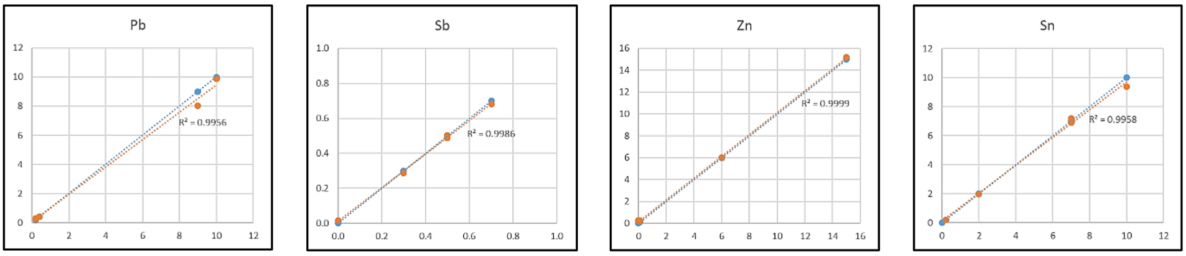

Fig. 3: Deviations of the measured results plotted against the values of the IMMACO reference alloys, for the elements lead (Pb), antimony (Sb), zinc (Zn) and tin (Sn). All results are expressed in weight percent on the X- and Y-axis.

Historical-technical aspects of the use of copper for matrices

Copper as a metal has been in use for about 10.000 years and found its widest practical use in the Bronze Ages.[5] In the Middle-ages copper was used as a metal for cooking gear or as a base metal for plating with silver or gold. In most cases however it was used as a alloying metal for bronzes, brasses, gold and silver alloys. The use of copper for matrices in the context of hand printing is a logic choice amongst the metals that were available at the time. It has a high melting point (1083°C), is fairly soft (harder than lead, tin, gold or silver but softer than iron), has very good heat conductivity properties, is in ambient atmospheric conditions corrosion resistant and is relatively cheap.

Processing of copper in the 15-16th century is described by Biringuccio.[6] Chapter 3 in Book 1 concerns the localisation and mining of copper ore, chapter 4 is about refining operations. Chapter 8 of Book 3 goes into the parting of copper and lead from the 'matte', which is left from the assaying of silver. He mentions that the process leads to '*very pure and beautiful copper, which is called* rosette copper *and which comes from Germany in rough cakes*.'

[5] (Scott, Copper and Bronze in Art 2002), p.4-5
[6] (Biringuccio, Smith and Gnudi 1540; 1990)

In the second half of the 17th century Moxon states in his work 'Mechanick exercises' that '*The Steel Punches being thus finish'd, as afore was shewed, they are to be sunk or struck into pieces of Copper, about an Inch and an half long, and one quarter of an Inch deep; but the thickness not assignable, because of the different thicknesses in Letters…*'.[7] In the chapter 'Of Sinking the punches into the Matrices', he stresses on the fact that the copper pieces must be large enough to enable the strike of long or wide formed letters. The coppersmith had to make rods of different sizes as the lettercutter would have instructed him by handing over wooden patterns that indicate the sizes he needed to produce successfully strikes. He further instructs the coppersmith to choose the softest copper he can get, that the steel punches may find no hazard of breaking upon striking. Moxon continues: '*The rose copper is commonly accounted the softest: But yet I have many times Sunk Punches indifferently into every sort of Copper. Nay, even cast Copper, which is generally accounted the Hardest: Because Copper, as well (as some other Mettals) Hardens with Melting.*' He also explains why the copper rods must be 'deep' enough: '*That the more substance of Copper may lie under the Face of the punch: For if the Rod have not a convenient depth, the Face of the punch in Sinking, does the sooner ingage with the Hardness of the Face of the Stake it is Sunk upon: And having with a few Blows of the Hammer, soon hardned copper just under the Face of the Punch, as well the hardness of the small (thus hardned) Body of Copper just under the Face of the Punch, as the Hardness of the Face of the Stake contribute a complicated assistance to the breaking or battering the Face of the punch. But if the Rod be deep, the Substance of Copper between the Face of the punch and the Stake is less hardned, and consequently the punch will Sink the easier, and deeper with less Violence.*' It is correct to observe that copper hardens quite readily under cold deformation such as striking with a punch. Locally, indeed this hardness increases from 45 to 90 Rockwell when reduced in thickness with 50%.[8] Moxon continues saying '*But sometimes it has happ'ned that for the Sinking one Matrice or two, I have been loath to trouble my self to go to the Copper-Smiths, to get one Forg'd: and therefore I have made shift with such Copper as I have had by me. But when it has not been so deep as I could have wisht it, I have just entred the Punch into the Matrice upon the Stake, and to Sink it deep enough, I have laid it upon a good thick piece of Lead, which by reason of its softness has not hardned the Copper just under the Face of the Punch; but suffered the Punch to do its Office with good Success.*' Here, he points out the possibility to use lead as an alternative base which allows the copper to deform further even when the local hardness became too high. It results in a deformation of the bottom face of the copper rod but since they still have to be justified for use later, this poses no problem to produce a successful strike. It is noticeable that all of the described techniques are very recognisable in the 16th century matrices from the Plantin collection. The copper sizes, the finishes, the justification etc. seems not to have developed in the century that had passed.

Fournier describes at the end of the 18th century again fairly the same workflow: '*Ce sont de petits morceaux de cuivre rouge, de quinze à dix-huit lignes de long pour l'ordinaire, sur trois lignes environ d'épaisseur, mais dont la largeur est relative à celle des lettres, des ornemens, &c. que l'on veut frapper.*'[9] Saying: Matrices are small pieces of red copper, fifteen to eighteen '*lignes*' long for the normal upon three lines thick, but the width is relative to the kind of letters or ornaments which are to be struck. About the kind of copper he mentions that rods are cut from large plaquettes of red copper, called '*monnoie de Suède*' ('Swedish money'). These plates had a weight of six to eight 'livres'

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[7] (Moxon 1683)
[8] (Scott 2011)
[9] (Fournier 1764)

and carried at each of the four corners the arms of Sweden as a trade mark and guarantee for its monetary value. After being cut into long rods, they were forged manually to an even thickness and to the desired dimensions. After cutting the rods into smaller pieces to serve for the strikes, they are heated in a fire or a kiln to a red colour where after they are thrown in water to cool rapidly. This makes the microstructure more uniform and the copper overall softer. Fournier advises this process only for the larger letters to strike them more easily. For the smaller punches, the slightly harder 'as cast' copper is sufficient or even preferred to retain sufficient strength and hardness after striking. To finish the upper face that will retain the strike, it is filed flat with a coarse and a fine toothed file while adding a little oil. The surface is then polished using a burnishing rod, that is a long, round and highly polished steel rod. It is to be noticed that also this way of finishing was already used on the 16th century strikes in the Plantin-Moretus Museum (MPM) collection.

Metallurgical considerations

From metallurgic point of view, the copper used in the 16 to 19th centuries was not completely free of other metals, coming from the copper ores, from the smelting and refining techniques or from the casting, hammering or finishing techniques applied. In the pre-industrial times copper was drawn from the ore by roasting and refined with a combination of heat and oxidation. This left often small amounts of lead and antimony behind. Since no analytical techniques were at hand, the quality of the 'pure' copper was tested by striking a cast test piece with a hammer while it was still red hot. If it stayed solid, is was considered pure and suitable for further processing. When the test piece broke under the hammer, it had to be refined still further. This gives way for certain variations in composition although the copper was usable for different operations such as making matrices, but also for making sheets for copper engravings or oil paintings and for alloying with precious metals.[10]

Another aspect to take in account is the possible segregation of certain elements at the surface of a metal ingot cast. This is widely known as a surface enrichment, where in most cases the metal with the highest melting point solidifies first (in this case copper, 1084°C), leaving the presence and the detection of certain constituting elements more difficult with a surface measuring technique. In the case of the matrices, the copper content of 99 wt% or more makes that the antimony stays dissolved in the copper grains. As can be seen in the phase diagram, antimony (melting point 817°C) forms an α-phase (Fig. 4, phase 'Cu') with the copper. The highest dissolution is present at temperatures above 500°C, diminishing with cooling down towards room temperature. With the very low levels (less than 1 wt%) that are to be expected in 'pure' copper, we can assume that all the antimony is kept in solution. Lead however does not dissolve at all into copper (Fig. 5) but stays as globules at the grain boundaries. Moreover, lead has the lowest melting point (327°C) and therefore solidifies last on cooling down from the casting. Another aspect that can change the lead distribution in the copper are annealing and forging processes that could have been applied to the copper rods. If this was the case for some of the copper rods (e.g. strikes and matrices), the initial lead content may have been re-distributed throughout the matrix.

[10] (Scott, Copper and Bronze in Art 2002)

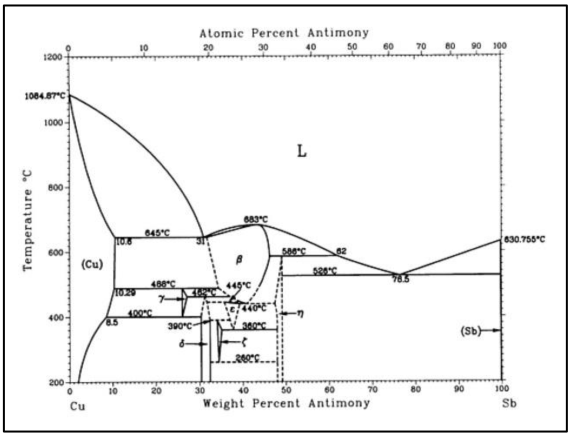


Fig. 4: Phase diagram of Cu-Sb where the dissolution of Sb in Cu is shown as the broad α-phase at higher temperatures, narrowing down towards room temperature. (ASM Vol. 3 Alloy Phase Diagrams, 1992)

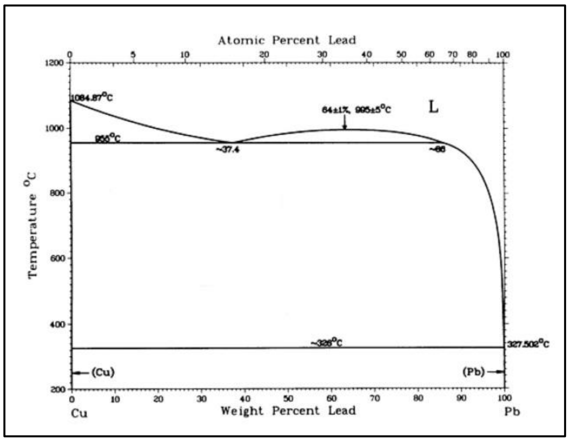


Fig. 5: Phase diagram of Cu-Pb showing no dissolution of Pb in Cu, since no α–phase is present at any temperature. As a result, lead is present as distinctive globules throughout the copper matrix. (ASM Vol. 3 Alloy Phase Diagrams, 1992)

The above mentioned possible segregation is to be taken in account since the measurements are performed at the surface of strikes (Fig. 6, Fig. 7) or on justified matrices (Fig. 8), where an important part from all sides has been removed mechanically. For the non-justified strikes, the measurements are taken at the upper surface with the punch mark, which was previously prepared to a smooth surface (Fig. 6) while the sides and bottom remained with fire scale from the casting and/or hammering, or with saw marks (Fig. 7). However, more detailed information on the lead concentration and distribution throughout each strike or matrice can only be obtained by destructive sampling and consequent analysing techniques such as metallography or scanning electron microscopy (SEM-EDX). This is however not applicable for the studied valuable historic objects.

Fig. 6: The striking face, previously prepared by probably sawing and/or planing, filing, sanding and polishing (most probably burnishing)

Fig. 7: The side and back of a strike, showing resp. oxide scale from casting or hammering and saw marks from parting larger sheets into rods

Fig. 8: Corrosion has occurred on some of the matrices set MA 71. This presence of corrosion could not be attributed yet.

Selection of matrices

For the purpose of evaluating the XRF technique, a choice amongst matrices sets in the collection of the MPM was made. Five sets of strikes, attributed to Robert Granjon (°1513-†1590) were selected. These are the strikes MA 120, 134, 156, 184 and 191, for 'Jolie Cursive'. Besides the facts that they have a very similar visual appearance and they were most probably never used to cast type in them, we know from the Plantin archives that the strikes were purchased as a lot of 11 identical sets from Granjon in 1574 together with the punches (ST 30). The six other sets are no longer known to exist.

Fig. 9: The five selected strikes sets from the Plantin collection. MA 156 lacks the capitals.

Robert Granjon was born in Paris about 1513 as the son of the bookseller Jean Granjon. He was a bookseller and a punch cutter, travelling frequently to Lyons and after having married and lived there, leaving in the mid-1560s for Antwerp, to work for Plantin and Silvius. With van den Keere, mainly for type and in some cases punches and matrices, Granjon was Plantin's main purveyor of punches and matrices from the end of the 1560's up to the 1580's. In the early 1570s, Granjon stayed in Frankfurt, Paris and Lyons, which he left in 1577 heading for Rome and to die in 1590. Granjon was a talented man, had innovative ideas and was very productive. He has developed and cut nearly ninety typefaces: 30 Italics, 7 civilités, 9 Greeks, 20 Romans, two or three Hebrews, a dozen exotics, half a dozen music typefaces, and an unknown number of initials, arabesque ornaments and fleurons. His average production comes close to two typefaces per year. Besides selling books and cutting punches, it is in this context important to note his trade of selling matrices to the whole of Europe. Printing types from his matrices were available in France, Italy, Spain, the Netherlands, and the German-speaking and Scandinavian countries until the end of the eighteenth century.[11] The MPM preserves 25 sets of punches or matrices attributable to Granjon.

About the strikes in sets MA 120, 134, 156, 184 and 191 (further denoted as 120 etc.) the Inventory of Parker-Melis mentions: "Caps, lc, lig, acc, nos, punct, 111 strikes. MA 156 consists of lc, acc, nos, punct, 77 strikes.[12] Furthermore it is read in the archives that Plantin paid Granjon in October 1574 for 11 strikes with the punches [Ar. 98, pp. 257, 265, 277]. One of these he exchanged with van den Keere for his Jolie Romaine in May of 1575 [42 p 7v°; 153 pp 160, 165]. On van den Keere's death in 1580 he had a strike of '*Jolye Cursyve van granjon*' [Inv vdK 1580] which Plantin bought back from his widow since it does not occur in De Vechter's post-1581 Inventory. None of the others appear in the 1581 Inventory and so were probably at Frankfurt. In 1588 there are at Frankfurt '*7. Sept frappes de corsive*

[11] (Vervliet 2010)
[12] (Parker and Melis, Inventory of the Plantin-Moretus Museum Punches and Matrices 1960)

nonpareille de Grandion contenant chacune frape 111 matrices' [Inv Frank 1588]. In 1589-90 there were 6 at Frankfurt, 1 at Antwerp and 4 at Leyden, all of 111 strikes, in sum the original 11 sets [Invs Ant 1589, Frank 1590, Leid 1590]. The [1590] Frankfort and 1590 Frankfort-Leyden Inventories show that five of the Frankfort six went to Moretus, the other one to Raphelengius, making five at Leyden and six at Antwerp; these six were still there c. 1612. The 1652 Inventory seems to list only one strike, but the 18th century inventories list ten. The surviving sets of strikes, MA 120 etc., all have cursive capitals which appear to be original with the sets. At any rate they were cut before March 1579 when van den Keere justified matrices for them for MA 71.[13]

The van den Keere Inventory of 1580 (Ar.153, p.295-303), List C (Ar.153, p.299-297)[14] mentions under '*Dit syn rau afslaghen'* (These are unjustified strikes) MA 120 etc. *'Jolye Cursyue'*. A note is also added: '*Noch so esser seer veel copere dwelck al bereyt is an afslaen van alle soorten'* (And also there is a lot of copper which has been already prepared to make strikes of all sorts). When Plantin and De Vechter (foreman of van den Keere's foundry) acquire all the material from van den Keere's workshop, we can assume that for strikes they would produce their selves or as an order with a subcontractor, these copper rods could have been used until the stock was exhausted.

MA 71 is a set which has been justified, while the other sets (above) are strikes which were ordered by Plantin for resale to other printers. All above matrices were struck with the punches ST 30 from Granjon's hand (except for the numbers). The lower case would have been cut in 1573 or before, whilst the capitals date probably from 1573 or 1574 and were used by Plantin from 1575 on. It is thought that the first set was only equipped with roman capitals, making this set a composition of different series of matrices. Between August 26th and November 29th 1573 Plantin paid 71 s 5d for '*Les matrices de granjon cursive nonpareille'* and 4s 8d '*A s granjon pr reste des matrices susdites'* [116 p 285]. Between October 14th and 17th van den Keere charged Plantin '*Pour la preparation du moulle de la plus petite nonpareille / et celuy de ma nomp flam pour faire les 2 epreuves de la petite Cursive de Granjon'* [153 p 150]. On September 24th 1574 he sent '*la première partie de la Cursive Jolie en 2 mandes'* [153 p 153, 42 p 1]. On November 25th he sent '*La fonte des pet. Cap. De la Coronelle sur la Jolye / qui est l'accomplissement de la fonte entiere de la Cursive Jolye'* [153 p 155, 42 p 2]. The Bible small capitals seem to have been found more suitable later. On March 10th 1579 he sent a font of '*Capitales couchées de la Jolye Cursive'* and charged for justifying the 23 matrices [153 p 197, 42 p 18]. The 1581 Inventory lists a Jolie 'C*ursive de Robert Granion / justifiée and the 1590 Leiden '164 Jolie Italique justée*'. The c 1612 Octavo Inventory lists '*164 Jolie Cursive'* and the 1652 '*140 Jolie Cursive'*. Possibly in the latter the matrices for the Coronelle small caps have been removed, perhaps to MA 76 (N° 82).[15]

[13] (Parker, Melis and Vervliet, Early Inventories of Punches, Matrices and Moulds 1960)
[14] (Parker, Melis and Vervliet, Early Inventories of Punches, Matrices and Moulds 1960)
[15] (Parker and Melis, Inventory of the Plantin-Moretus Museum Punches and Matrices 1960)

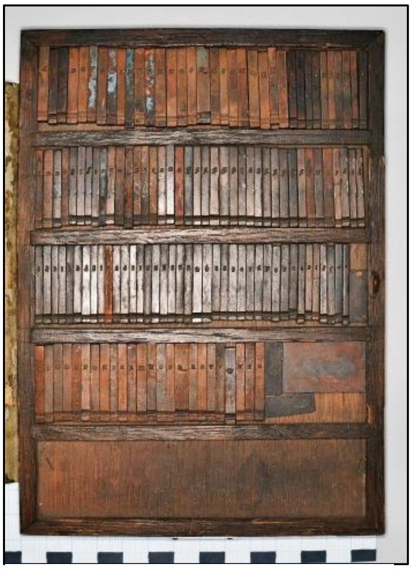


Fig. 100: Justified matrices MA 71

Matrices set MA 71, consisting of 'Upright caps, cursive caps, lc, lig, acc, nos, punct, 137 matrices[16], the numbers not being from ST 30. The upright capitals are the small capitals to Garamont's Bible Romaine [N° 77].' This set would have been already acquired in 1573, although it does not appear together with the 11 sets of strikes in the 1580 inventory. It does appear in the 1581 inventory as 'Cursiue de Robert Gronjon justifiée'. Perhaps it was not complete at that point and therefore was not listed. It was possibly without any capitals since he added a fount of small capitals from the 'Coronelle Romaine' to serve as the capitals (Ar. 153, p.155; Ar.42, f.2). This idea is supported by the texts which are attributed to this type, consisting only of the Jolie Cursive with upright capitals.[17]

Results of the measurement on strikes MA 120 etc. and matrices set MA 71

The measurements show that, a very few number of exceptions left alone, the matrices sets 120 etc. resemble the use of very similar copper alloys, being a main group (A) of very high purity copper (about 99.5 wt%) with mainly lead (average 0.26 wt%) and antimony (average 0.23 wt%) (Fig. 11). A second group with a much smaller number of matrices, shows average values for lead at 0.27 wt% average and elevated concentrations of antimony at 0.44 wt% average (group B).

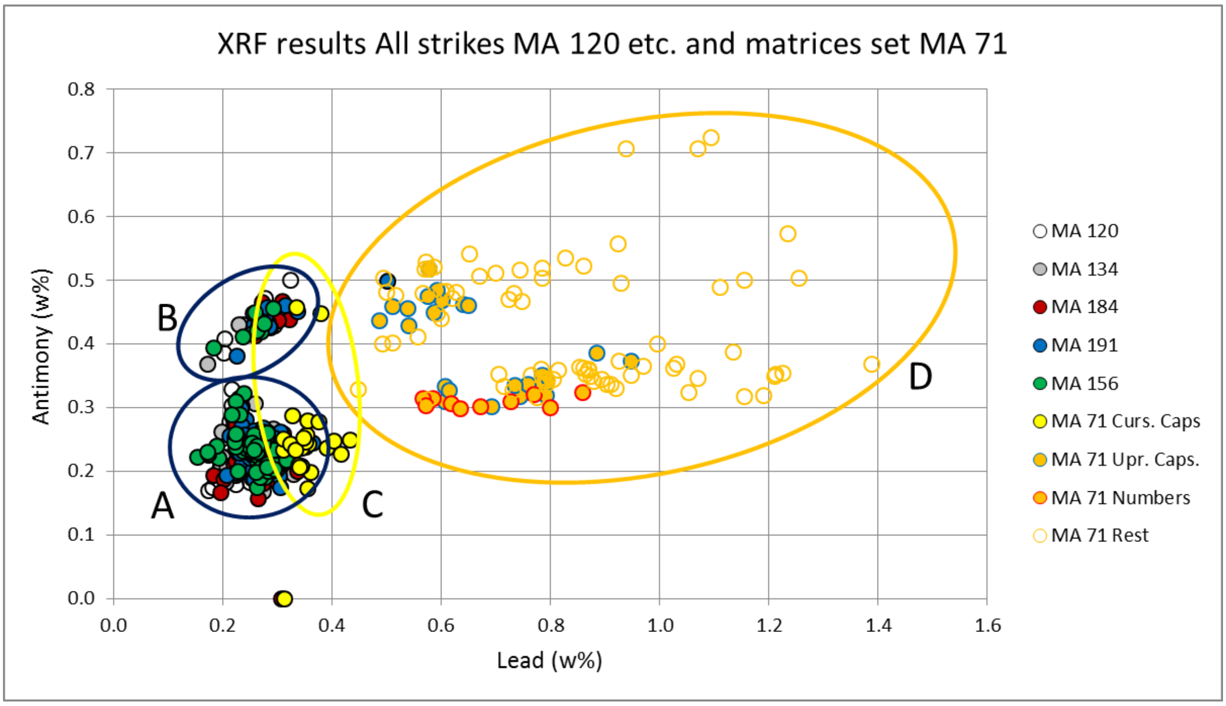


Fig. 11: The registration of all measurements shows the grouping of all sets of strikes (blue groups, A, B) and group D (orange) representing matrices set MA 71, wherein the cursive capitals form a separate group (yellow, C), resembling the alloy compositions of the strikes. The matrices in MA 71, other than the cursive capitals, show a large variation in composition and relative low copper purity.

[16] 137 matrices minus the caps. curs. 25 + AE = 111, matching the number of strikes in MA 120 etc.
[17] (Parker and Melis, Inventory of the Plantin-Moretus Museum Punches and Matrices 1960)

In this group B with an antimony content exceeding 0.35 wt%, it is particular that the matrices for the lower cases *c* and *e* repeatedly are situated in this group. Also several numbers and some accents belong to this specific group, throughout the different matrices sets (Fig. 12). This could point towards the fact that Granjon had access to two batches of copper rods. A reason for the choice of making the matrices of *c*, *e*, some numbers and some accents in copper of a different batch is difficult to determine; it may also just be a coincidence. Nonetheless, it shows that the matrices were struck most probably with the respective punches consecutively in the same copper rods. Finally, the 0 (zero) from strike set MA 184 contains no antimony at all, which is an exception to all results. (Fig. 12) The external visual aspects are no different from the other strikes.

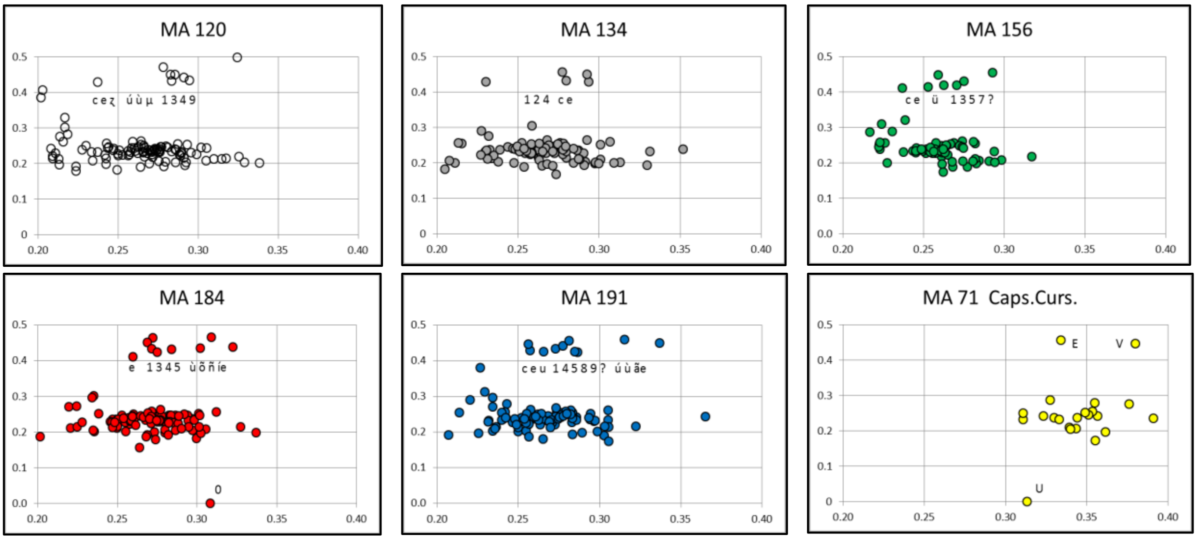


Fig. 12: Identification of the matrices which were struck in a deviant kind of copper compared to the majority of each set of strikes (MA 120 etc.) and the cursive capitals in MA 71. X-axis is lead (wt%), Y-axis is antimony (wt%).

Matrices set MA 71 does not resemble compositional similarity to any of the other measured sets, except for the cursive capitals (Fig. 11). These cursive capitals (25 matrices, top left in box,), are within MA 71 a separate group from the other alloys. The copper alloy is also relatively pure and homogeneous as a group with average levels of 0.35 wt% lead and 0.24 wt% antimony. Within this group there are three exceptions: the matrices E and V have a high level of Antimony (0.45 wt%) while the matrice U has no antimony. For the latter, it is most probable that this points towards a later addition of the matrice. (Fig. 12)

The close match of results between the cursive capitals in MA 71 with the other sets (MA 120, 134, 156, 184 or 191) and the fact that also two of the matrices have an elevated content of antimony, rises the probability that also Granjon produced these strikes. Since MA 156 does not contain (any more) the capitals, it puts the assumption forward that these were used to be justified and to complete the MA 71 set. The measurements tend to show this, including two deviated letters (E,V) with a similar higher content as seen in the other matrices sets. However, there are two remarks to be made. Firstly, there is a slight deviation in the measured Lead content which is probably due to the segregation effects. Since matrices have smaller dimensions because they are justified by filing down the strikes, the measurements are executed on a level more close to the centre of the copper piece. Therefore it is possible in this case as the results of group C indicate, that the Lead concentration is somewhat higher at the centre compared to the outer surface of the original strikes (alike MA 120 etc.). Secondly, as in the other strikes none of the capitals have an elevated concentration for antimony, it contests the probability that these were harvested from MA 156. But, since only 5 of the original 11 sets were measured, it leaves room for speculation.

None of the other alloys (besides the cursive capitals) in set MA 71 corresponds with the matrices sets of Jolie Cursive. The average values of the very dispersed measurements are 0.78 wt% lead (0.45-1.39 wt%) and 0.41 wt% antimony (0.30-0.72 wt%), see Fig. 11. The nos. which are struck from another set of punches, form a relative dense group at least at the level of Antimony content. These alloy compositions however appear equally in other matrices of MA 71. Also, the upright capitals resemble the copper composition of the other (non-cursive capitals) matrices. (Fig. 11)

As an intermediate conclusion, it is shown that XRF has the ability of measuring small differences in trace metals, present in copper. The most significant elements which seem to be present in the above measured matrices sets are lead and antimony. Other elements such as iron and silver may be present, but are very close to the detection limit of the instrument, making the readings uncertain and are therefore not considered primarily.

As a <u>first case study</u>, the selection of the matrices set MA 76 was made accordingly to the supposition in the 1960 Parker & Melis Inventory that the Roman capitals could have been moved from MA 71 to MA 76. The revised comments on this set, published in De Gulden Passer 1960 (p.62), abandoned already this idea because of the following arguments: “Plantin may have bought these matrices since they are not listed in the *post* 1581 De Vechter Inventory. However, the first inventory to list a second set of *Jolie Romaine* matrices seems to be the 1652 Inventory, which is somewhat confused in the smaller sizes. MA 76 is equipped with the small capitals to van den Keere’s *Coronelle* instead of the capitals of the *Jolie*. There are two plausible reasons for this: either van den Keere had not cut the *Jolie* capitals when he made this strike and so substituted the small capitals from the earlier *Coronelle* or else the set is a late one and the change in capitals is due to confusion.”

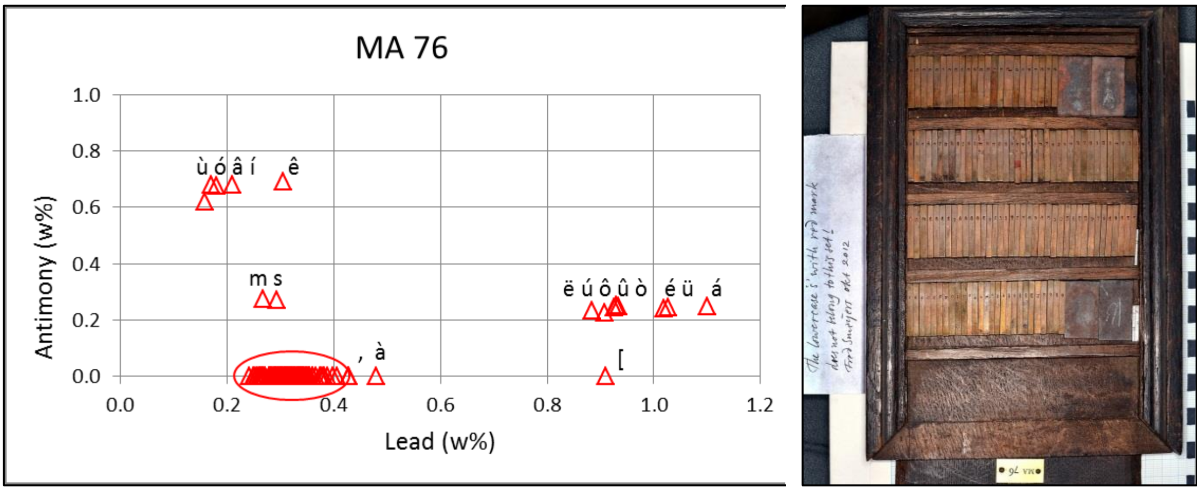
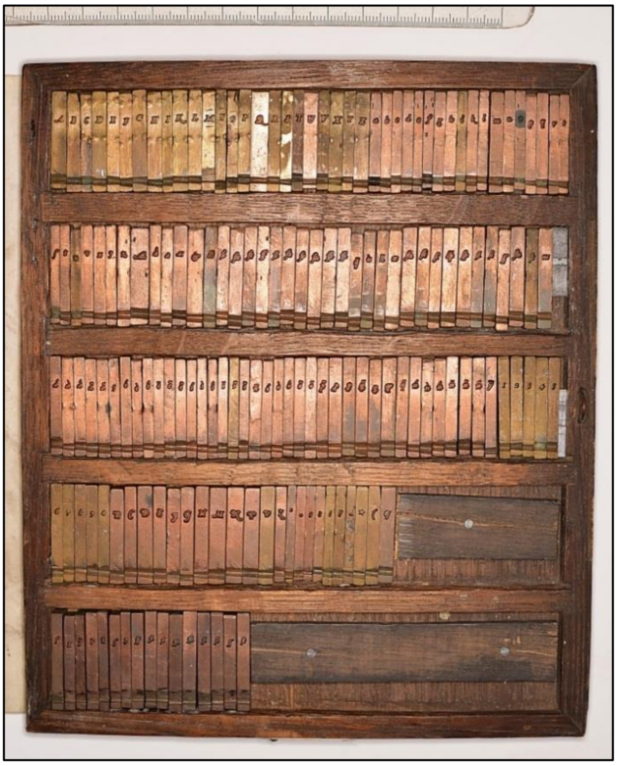


Fig. 13: MA 76 with 125 matrices showing 110 results without any antimony (majority in red circle, where the , à and [have a higher lead content), 8 results with a low antimony / high lead content and 5 matrices with a very high antimony / low lead concentration. All results which deviate from the main group are accents. The only two exceptions are the lower case letters m and s.

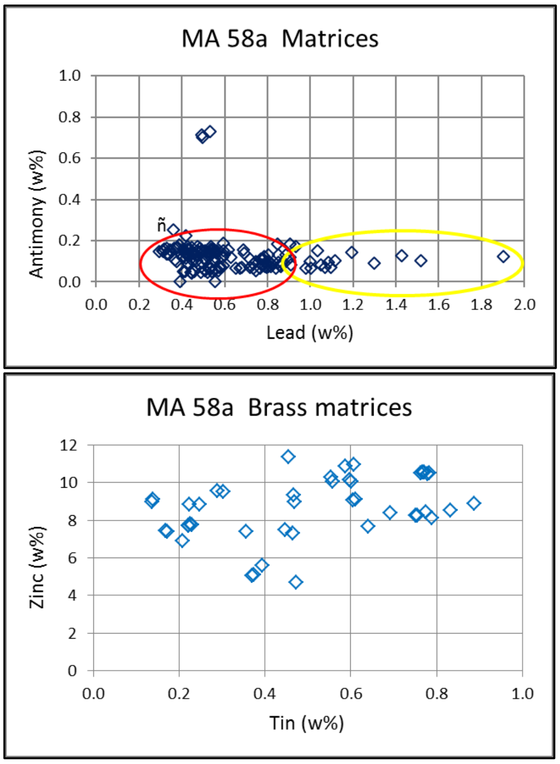
The small capitals, which are thought to be moved from this set to MA 76, do not resemble any composition found in MA 71. All capitals in MA 76 have a composition with a low concentration of lead (approx. 0.30 wt%) and no antimony, which is remarkable. The capitals also match with the rest of the set, i.e. lower cases (except for the m and the s), the ligatures, the numbers and punctuations (except for [) and most of the accents. The latter group however shows two other sub-groups but none of them with a composition that is to be related to MA 71. As a conclusion it appears that this set is consistent as a whole in its alloy compositions, besides the m and s and a number of accents which are struck in two deviant kinds of copper rods.

An interesting note can be made on the fact that Mr. Fred Smeijers added in 2012 a note in the matrices' box saying that the lower case s does not belong to this set. This is supported by the XRF result, but it also indicates that perhaps the letter m should be checked as well since its composition resembles that of the s.

As a <u>second case study</u>, set MA 58 drew attention because of the yellowish appearance of some of the matrices. The set consists of Caps, lc, lig, acc, nos, punct, 150 matrices (Parker-Melis Inventory, 1960). The Inventory further comments: 'In 1561, Plantin had matrices for a 'Breviaire Italique' [Inv 1561]. On January 1st 1562 he received from 'Francoijs le Fondeur' matrices for 'Italica Brevier N° 153' which he sent off to Martin le Jeune in Paris [N° 54; 36 p 17v°] probably as a precautionary measure before his own flight. The 1563, 1572, 1581 and Antwerp 1589 Inventories all list a set of Breviere or Bible Cursive matrices by Granjon, the latter giving the number as 157. The c 1612 Octavo Inventory and the 1652 Inventory lists 157 Bijbel Cursive. In October 1572 van den Keere revised all the matrices of the 'Cursive de breviere' and replaced a missing comma [153 p 150]. On January 4th 1576 he charged Plantin for adding an ñ in the 'bible Cursive' [153 p 183] which is in this set.'



Set of matrices MA 58a and b



XRF results indicating a mix of copper and brass matrices (brass: copper and zinc alloy).

Fig. 14: The yellowish and red appearance of the matrices proved to be a mix of brass and copper ones, as shown by the XRF results in the two graphs on the right. The brass also holds besides the zinc content (average 9 wt%) a very small and variating amount of tin (average 0.5 wt%) in its alloy composition (lower graph). The brass also has an elevated concentration of lead, indicated by the yellow circle (upper graph); the copper matrices are situated in the red circle, except for the U, ‘F and ;. The ñ which is added by van den Keere in1576 is situated at the outer edge of the main group’s composition.

The set shows to consist of a mix of copper and brass matrices. The copper matrices are all of a low antimony content, except for the capital U, the additional capital F (denoted ‘F) and the punctuation mark ;. All the capitals (except U) are struck in brass of about 9 wt% zinc and averages of 0.5 wt% tin and 1 wt% lead. Other matrices in brass are: i, u, ʃt, ĩ, the additional capital V, punctuations :, !, ?, -, * (ɑı and all the numbers 1, 2, 3,…, 0.

The comma, as mentioned to be replaced by van den Keere it in 1572, is not in the set today. The additional ñ by van den Keere in 1576 is still in the set, it’s composition slightly deviating from the group of other copper matrices in the set.

Fig. 15: The brass capitals and copper lower cases, except for the U which is also copper and most probably a later addition.

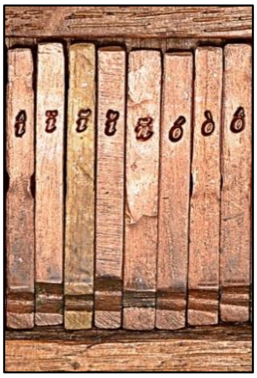


Fig. 16: Detail of the brass ĩ amongst other accents in copper, as also the added ñ.

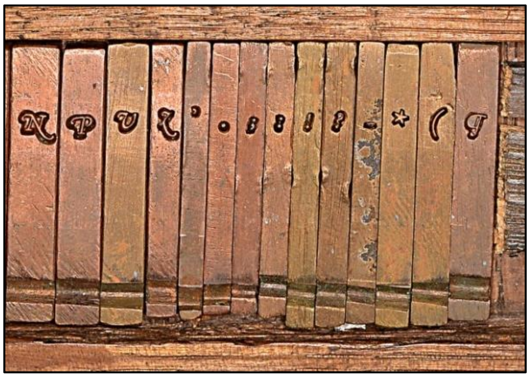


Fig. 17: The additional capital V in brass, followed by the punctuations in copper (‘ . ; : ɑı) and in brass (! ? - * (), lacking the comma that would have been replaced in 1572.

This set of matrices shows that brass was used for striking matrices, certainly for the smaller typefaces which punches require not too much force to be driven into the metal. Brass is only slightly harder than pure copper[18] but has the advantage to harden less locally at the face of the punch. From a metal worker’s point of view, it is surprising to see that brass was used not more frequently for the production of matrices since hardness cannot have been an argument. The lesser corrosion resistance or the wear and tear by casting type could perhaps be reasons for matrice makers to persist in using ‘pure’ copper.

[18] (ASM 1990), p.779-782: Brinell hardness value of Cu is 44 while brasses may vary upon specific composition between 33 and 60. All values are for annealed (softened) conditions.

Multivariate data analyses

In addition to the previous analyses, where groups of matrices were observed by plotting metal compositions in a pairwise manner (Pb/Sb and Zn/Sn), multivariate data analyses were performed. These techniques allow to explore patterns in the alloy compositions using the concentration of all 8 measured metals besides copper simultaneously (Pb, Sb, Zn, Sn, Ag, Fe, Ni, As), showing to what extent the overall metal composition differs between and within sets of matrices. Some multivariate analysis techniques use the information on group membership to optimally split the observations (here: the matrices) between groups (here: the sets), whereas other techniques ignore the set membership and merely use the observed metal concentrations.

Cluster analysis belongs to this latter group of techniques. In brief, it explores which of the individual matrices are similar across all 8 metals, grouping the similar matrices together in clusters. The actual group (here: set) membership is not taken into account – matrices are assigned to clusters regardless of the set they belong to. In a first stage, we explored how many clusters would give an optimal separation of the clusters. We pre-specified the number of clusters to all values from 2 to 15 clusters, and plotted the within-cluster versus the number of clusters (Fig. 18). This showed that any solution between 5-9 clusters would be acceptable. Table 2 shows the results for 7 cluster centres. This was found to correlate optimal with the historical information.

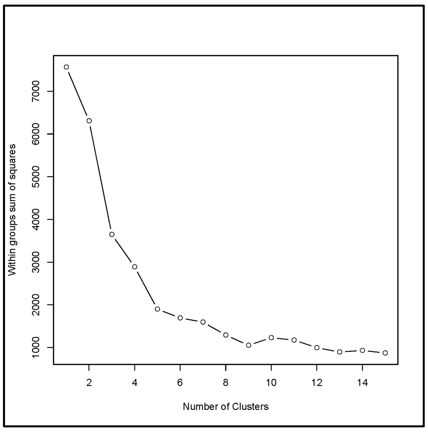


Fig. 18: Variability within clusters (Y-axis)
Versus Number of clusters (X-axis).

Subsequently, we applied the Partitioning Around Medoids (PAM) algorithm to assign the matrices to the 7 clusters. This is an iterative algorithm, whereby 7 initial cluster centres were defined (in 8 dimensions, for the 8 initial variables). Each observation was assigned to the nearest of the 7 initial cluster centres. Therefore, 7 new cluster centres were calculated based upon the observations belonging to one cluster. Typically, cluster centres shift with regard to the initial cluster centre. Using the novel cluster centres, individuals observations were again assigned to the nearest cluster centre. This algorithm was repeated iteratively until the cluster centres no longer change. The final solution has assigned each observation to one of the 7 clusters.

In table 2, cluster membership is plotted versus set membership. Almost all matrices from set MA 120, 134, 156, 184 and 191 belong to one and the same cluster, indicating their composition is similar. Also the cursive capitals of MA 71 are attributed to this cluster, supporting the previous observations. On the other hand, MA 58A consist of matrices that are mainly assigned to 3 other clusters, indicating that this set was not put together at the same time or from the same batch of copper. In detail, the lower case letters belong to cluster 2 while the ligatures are assigned to cluster 5 and the brass matrices are in cluster 6. It is also interesting to see that the letter ñ, added at a later time by van den Keere is separated from the others by its position in cluster 1. MA 58B on the contrary is totally assigned to cluster 7, that consists almost exclusively of matrices from this set, and conversed all matrices from set MA 58B are assigned to cluster 7, highlighting that these matrice set is distinct from the other sets. Finally, the matrices except from the cursive capitals in MA 71 and MA 76 are all distinguishable from the measured sets.

Table 2: Cluster analysis giving 7 distinct groups for each set and its individual matrices.

	Cluster assignment						
	1	**2**	**3**	**4**	**5**	**6**	**7**
MA120	110	0	1	0	0	0	0
MA134	111	0	0	0	0	0	0
MA156	77	0	0	0	0	0	0
MA184	110	0	0	1	0	0	0
MA191	110	0	1	0	0	0	0
MA58	1	79	3	0	22	44	1
MA58B	0	0	0	0	0	0	16
MA71	28	2	105	0	0	0	0
MA76	2	4	1	110	8	0	0

Discussion on the measuring technique

With this measuring technique we were able to show that strikes and matrices were made from different groups of copper alloys, including also a minority of brass matrices. These observations also show the level of measuring accuracy since the formation of the groups cannot be considered coincidental. Simultaneously, a certain deviation of results within each group is not to be avoided due to the metal alloy itself, segregation effects, possible corrosion layers of other substances on the surface, the fact that XRF is a surface measuring technique and that the peaks for the accompanying elements besides copper in the spectrum are relatively weak as a signal to noise ratio. The maximum overall deviation on identical materials (i.e. MA 120 etc.) with the measurement settings as described lies around 0.1 wt% (+/-) (Fig. 21). From these first measurements, we can give preliminary assumptions concerning the average levels of lead and antimony contents in 'pure' copper in the 16th century. This is supported by measurements on '*insculpatieplaten'* or copper plates carrying the hallmarks of goldsmiths from the 16th century.[19] Very similar levels of lead (average of 0.6 wt%) and antimony (average of 0.3 wt%) were found in these plates, which were also used as pure copper for a similar goal, being the striking of punches. As for the composition of matrices, many more measurements are needed to increase the critical mass for statistical evidence. Nevertheless, it is to be considered that this measuring technique can act as a useful tool for determining groups in historical metal objects.

[19] Copper plates from the collection of STAM, Ghent (not published)

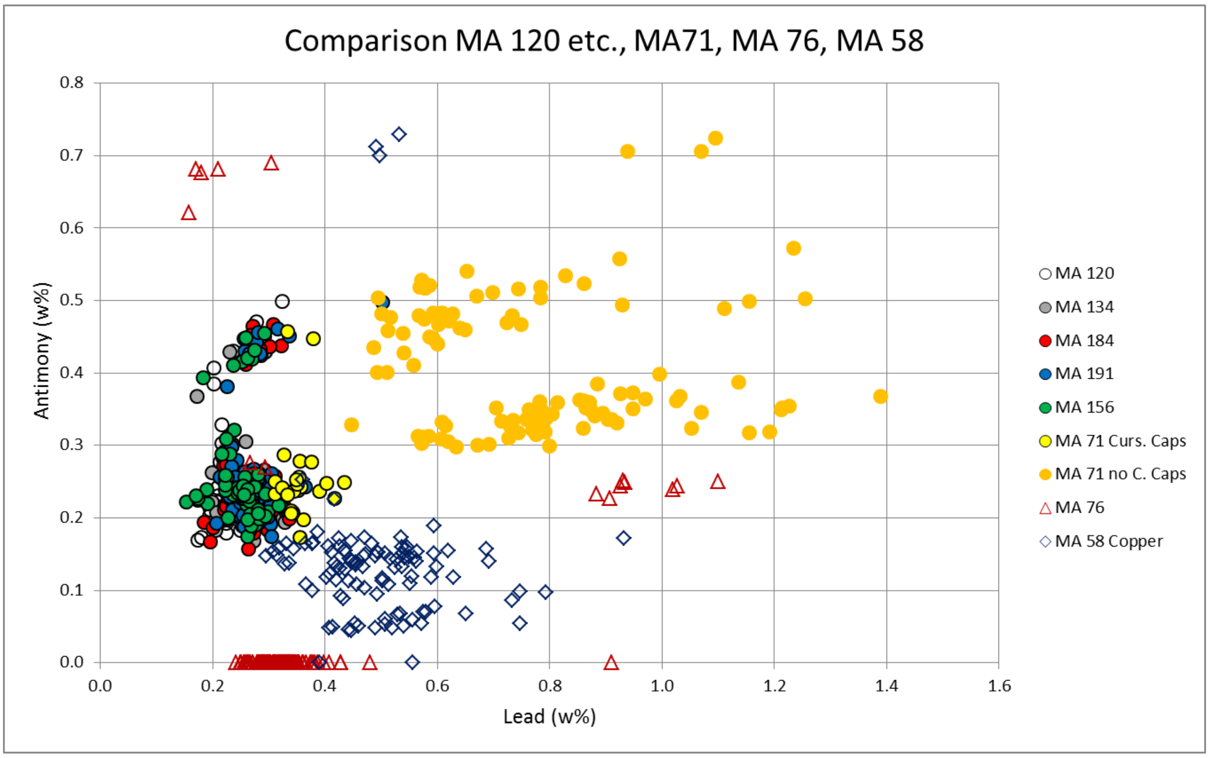


Fig. 19 Graph with the comparison of all measurements of the main elements Pb/Sb, showing the grouping per set and the parting of non-matching sets and/or matrices. This is coherent with the multivariate analyses.

<u>Discussion on the matrices composition results</u>

Comparing the measured sets of matrices, the grouping of identical sets and the parting of non-matching sets is evident (Fig. 18- Fig. 20 and Table 2). The very close concentration of the strikes MA 120 etc. with its two groups of copper as described above, including the very close match with the cursive capitals from MA 71 can be observed. The results of the other matrices in MA 71 show relative large spreading of lead concentration results, but gives also two levels of antimony concentrations. Set MA 58 is made of copper with noticeable lower concentration of lead and antimony as far as the copper matrices are concerned whilst set MA 76 isolates its position from the other sets because of the absence of antimony for 110 on 125 matrices. The few matrices with deviated concentrations of lead and with antimony also gather in two groups, separated from the other results.

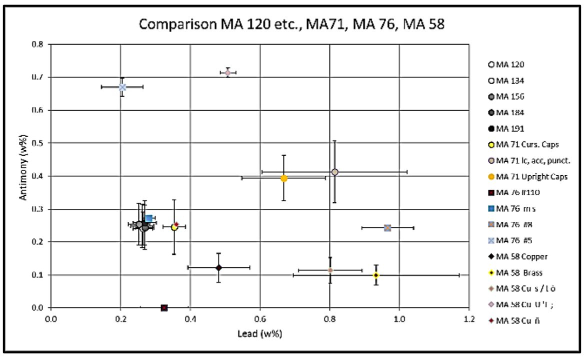


Fig. 20: Graph with the comparison of all averaged results and standard deviations per set and the parting of non-matching sets and/or matrices

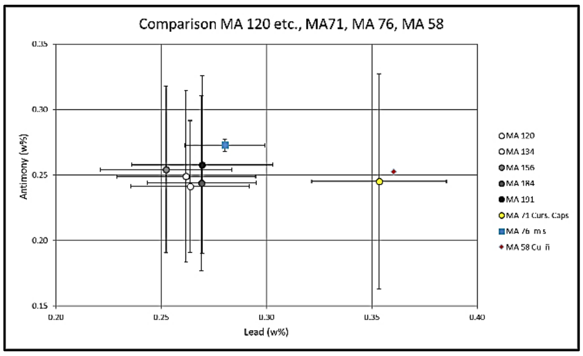


Fig. 21: Detail of Fig. 20, with the group op MA 120 etc. compared to the Cursive Capitals of MA 71. Also the later additions of MA 76 (m, s) and MA 58 (ñ) seem to be within the same composition ranges, 0.25-0.35 wt% Pb and 0.25 wt% Sb.

From the archives and the inventories, it is unclear when exactly MA 71 was made or on what moments of time it was changed or completed. When this set would be '*Les matrices de granjon cursive nonpareille'*, it was bought by Plantin in 1573 from Granjon [Inv 116 p 285]. From the XRF results it is only clear that the cursive capitals are a separate group in the set and that the compositions very closely match these of the sets of strikes MA 120 etc. The other matrices in the set do not show distinct groups apart from the two levels of antimony content. Therefore, it is likely that all the matrices in MA 71 (except for the cursive capitals) were produced from the same copper supply. Additionally it can be noted that the upright capitals' compositions are part of both groups of antimony content whilst the nos. are exclusively and well grouped at the lower antimony level (Fig. 11).

MA 76 is attributed to van den Keere, 1575. This set would have been bought by Plantin with van den Keere's death in 1580. The only other mention of a second set of Jolie Romaine matrices seems to be in the 1652 Inventory which lists '129 Jolie Romaine, ghejusteert' (Parker & Melis, 1960). They comment further on this: 'It seems reasonable to suppose that van den Keere's set of matrices might have been struck before he had finished, or started, the correct capitals, and so been equipped with the small capitals from his earlier Coronelle. Alternately, perhaps it postdates 1612 and the capitals are those of MA 71.' The measurement results give no indication of the completion of the set with capitals from another set, certainly not from MA 71. When the small capitals would have been transferred from van den Keere's earlier Coronelle, it cannot be excluded that the same batches of copper present in his studio were also used. This would give no different analytical readings. In any case, it seems correct that the assumption of a transfer from MA 71 to MA 76 was abandoned (Parker, Melis & Vervliet, 1960). It would however be interesting to measure van den Keere's earlier Coronelle (MA 161, MA 62, MA 148 strikes) to look at possible material similarities. The complete absence of antimony in the copper is certainly particular and points towards 'rosette' or 'rose' copper, as described by Biringuccio and Moxon.

Concerning the set MA 58a, it is assumed that this set is the 'Breviaire Italique' [Inv 1561], attributed to Granjon and meaning it would predate this year. In those years, Granjon worked in Lyon.[20] The copper matrices in this set are all of a low lead (average of 0.5 wt%) and an very low antimony content (average of 0.1 wt%). Particular are the brass matrices of which it cannot be said if they were struck at the same time or added to the set to replace missing items, or vice versa for the copper matrices. The character ñ which is added by van den Keere in 1576 is situated at the outer edge of the copper groups composition (Fig. 14, top right), showing a low lead (0.36 wt%) and a higher antimony content (0.25 wt%). This is quite similar to the composition of the Cursive Capitals from MA 71 (Fig. 21), although this does not prove that it was made by the same person or from the same batch of copper. It does show that it is different of composition compared to the other matrices in the set.

The 16 matrices of MA 58b appear to form a separate group because of the presence of arsenic in the copper alloy (average values of Pb 0.35 wt% and Sb 0.22 wt%), which is unlike any other set composition measured until now. The MPM inventory describes this set as being 17th or 18th century.[21]

Considering that the copper used by van den Keere in 1576 is of a low lead and medium antimony content, we can come back to the MA 76 matrices composition. From there, it is open to discussion whether MA 76 is indeed from his hand. Perhaps MA 76 does postdate 1612 (see above), although the

[20] (Lane 2004), p.39
[21] (Parker and Melis, Inventory of the Plantin-Moretus Museum Punches and Matrices 1960) [N° 119]

capitals seem not to be from MA 71 as suggested.[22] A change in capitals, as later put forward[23], is also unlikely because the majority of matrices in the set (110 on a total of 125) do match closely with each other, suggesting that they were all struck from the same batch of copper. The two small distinctive groups of accents, respectively 8 and 5 matrices, do not match any other measured group either. The matrices for the lower case letters m and s have a similar composition as MA 120 etc., although at this stage of the investigation it cannot be excluded that it is merely a coincidence.

Conclusions

The results from the above measurements can act as a benchmark for further measurements on the MPM matrices collection since it was known from archival evidence and it has been confirmed here that the matrices MA 120, 134, 156, 184 and 191 were purchased by Plantin in a total order of 11 sets of strikes with the accompanying punches from Granjon in October 1574. The cursive capitals in the MA 71 set may be by Granjon, whilst the other matrices in this set show divergent results. Nevertheless there are similarities and arguments to put forward that the cursive capital matrices for MA 71 were harvested from MA 156, although the findings offer no exclusive answers.

As a case study, MA 76 was measured upon the supposition that the small capitals were moved from MA 71 to MA 76 or a change due to confusion. Analysis has shown that both hypotheses are unlikely since at the one hand no match in composition with MA 71 was found and on the other hand the capitals do match with the other matrices in set MA 76 proving that this set is an integrated whole.

As a second case study, MA 58A was measured for its yellowish appearance. It proved to consist of a mix of copper and brass matrices, all struck with steel punches. MA 58B proved to be of a completely different origin, containing arsenic and could confirm the later date attribution of being from the 17th or 18th century.

The results gathered in this research offer insights in the kind of copper the punch cutter and/or matrice maker has used. Nonetheless they used 'pure' copper, the lead/antimony concentrations varied for each batch of copper they acquired. These results will not lead directly to dating the copper, although multiple measurements on the MPM collection could provide more specifications on the kinds of copper used by certain punch cutters / matrice makers at a certain location or a certain point in time. It is also shown that the multivariate analyses can help in determining groups and outliers from the results.

The applied XRF technique could provide further proof for earlier research, draw some question marks upon former findings or bring new correlations at light in the origin and provenance of the matrices. It also can provide an insight in the use of copper throughout the 16-19th century and the evolution of refining techniques in combination with the sources and trade of the used copper.

[22] (Parker and Melis, Inventory of the Plantin-Moretus Museum Punches and Matrices 1960) [N° 82]
[23] (Parker, Melis and Vervliet, Early Inventories of Punches, Matrices and Moulds 1960), p.62

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Abbreviations

Groups of typefaces: Caps (Capitals); lc (lower case); lig (ligatures); acc (accents); nos (numbers); punct (punctuations)

Analytical measuring technique: XRF (X-Ray Fluorescence), keV (Kilo Electron Volt), wt% (weight percent), Metallography (the optical study of microstructures in metal alloys), SEM-EDX (Scanning Electron Microscope with an Energy Dispersive X-Ray Detector)

Metals: Cu (copper), Pb (Lead), Sb (Antimony), Sn (Tin), Zn (Zinc), Ag (Silver), Fe (Iron), Ni (Nickel), As (Arsenic)

Glossary

Annealing: heating a metal or alloy to a point of recrystallization, which softens the metal or alloy after cooling down.

Forging, sinking, hammering: plastic deformation methods on metals/alloys, normally executed at room temperature with non-ferrous metals. Due to the deformation, a hardening effect is obtained which allows at some point no further deformation without cracking or breaking the metal/alloy. At this point, annealing is needed.

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