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Calamine of the Bergamasque Alps as a possible source of zinc for Roman brass: Theoretical considerations and preliminary results

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ABSTRACT

The origins of brass are obscure and begin long before the Romans, however, it was the Romans who brought this gold-coloured alloy of copper and zinc from obscurity and placed it at the forefront of monetary policy and military power. Under the Romans, brass was produced on a scale never seen before, but until now there is little clarity on where brass was made and where they obtained zinc ore. Studies in the past focused on potential sources in Germany, but the lack of investigation on Italian sources represents a significant research gap. The major zinc source in Northern Italy (Gorno Pb-Zn district) could be the source mentioned by Pliny the Elder and may have played a major role in the Roman brass industry. Recent surveys around the area of Dossena, in the Gorno Pb-Zn district, have brought to light substantial traces of pre-modern calamine mining of unknown age. This study presents results from the mineralogical and geochemical characterisation of calamine ore from the Dossena area. A theoretical discussion on the sourcing of Roman brass through chemical and lead isotope analysis is provided along with a case study comparing analyses of brass with zinc ore sources. While the currently available lead isotope dataset on brass alloys is not ideal for exploring this issue, the lead isotope analyses confirm that the calamine from Dossena could have been used by the Romans for brass making and suggestions for future research are provided.

Keywords: archaeometallurgy; mining archaeology; aurichalcum; cementation; cadmea; sestertius.

INTRODUCTION

The origins of *aurichalcum* are obscure and begin long before the Romans, however, it was the Romans who brought this gold-coloured alloy of copper and zinc from obscurity and placed it at the forefront of monetary policy and military power. Under the Romans, brass was produced on a scale never seen before, and it is commonly accepted that the few suitable zinc ore sources known were closely controlled to create a state monopoly on brass and brass making (Beanlands, 1918; Grant, 1946; Craddock et al., 1980; Istenič and Šmit, 2007). The location of Roman zinc ore mines have been notoriously difficult to trace, as slag, a typical indicator of metal production, is absent. One of the only written sources available from the Roman period providing information is the Natural History of Pliny the Elder:

Vena quo dictum est modo foditur ignique perficitur. fit et e lapide aeroso, quem vocant cadmean, celebri trans maria et quondam in Campania, nunc et in Bergomatium agro extrema parte Italiae; ferunt nuper *etiam in Germania provincia repertum. fit et ex alio lapide, quem chalcitim appellant in Cypro, ubi prima aeris inventio, mox vilitas praecipua reperto in aliis terris praestantiore maximeque aurichalco, quod praecipuam bonitatem admirationemque diu optinuit nec reperitur longo iam tempore effeta tellure. proximum bonitate fuit Sallustianum in Ceutronum Alpino tractu, non longi et ipsum aevi, successitque ei Livianum in Gallia. utrumque a metallorum dominis appellatum, illud ab amico divi Augusti, hoc a coniuge. velocis defectus Livianum quoque; ... hoc a Liviano cadmean maxime sorbet et aurichalci bonitatem imitator in sestertiis dupondiarisque, Cyprio suo assibus contentis. et hactenus nobilitas in aere naturalis se habet. Book XXXIV, Chapter 2.*

In the passage, several important aspects should be emphasised. He describes an airy stone called *cadmea*, which can be used to combine with certain types of copper to produce a brass alloy similar to the *aurichalcum* of *sestertii* and *dupondii* coins. *Cadmea*, in this case, can be said to be synonymous with calamine, a historical term for secondary zinc ore consisting mainly of the minerals smithsonite, hemimorphite and hydrozincite. According to the account, *cadmea* was once found in Campania, but is now found in Germania and in the region of Bergamo in the extreme north of Italy. While his mention of an ancient source in Campania is deemed questionable due to a lack of significant or suitable zinc deposit, the latter two source locations are contemporary to Pliny's account, which could lend them greater credibility. Pliny was born around the year 24 AD in northern Italy at Como, (*Novum Comum*), a planned settlement built by order of Julius Caesar in 59 BC (Healy, 1999); the proximity of Novum Comum to the one and only nearby major zinc ore district means that it is plausible that Pliny knows of mining activity in the Bergamasque Alps from personal experience. Interestingly, in Biringuccio's metallurgical treatise, *Pirotechnia*, of the 16th century AD, calamine was said to be found in Germany and at a source near Como, the latter supplying the brass industry in Milan (Smith and Gnudi, 1959). Biringuccio was certainly in Milan, providing first hand descriptions of brass working there, but it is likely that the location of the calamine mines is a secondary account vaguely described to be in the direction of Como, and this is unlikely to be based on Pliny, who even less specifically stated the region of Bergamo. Thus Biringuccio's account gives a parallel to the main two calamine source regions given by Pliny 1,500 years earlier.

Up until now, most of the attention on Roman brass production has centred on the source in Germania, usually associated with the Aachen-Stolberg area (Davies, 1935; Craddock, 1995; Craddock and Eckstein, 2003; Nielen, 2006; Voigt, 1956) but also, to a lesser extent, Wiesloch (Hildebrandt and Kötz, 2009). While the discussion of Roman zinc sources in Germany has been grounded on physical mineral deposits, the dialogue concerning Cisalpine Gaul is of a theoretical and unspecific character. The possible significance of a zinc source in Cisalpine Gaul is discussed by several authors in context of the earliest uses of brass by the military and its first use in Roman coinage (Grant, 1946; Caley, 1964; Craddock et al., 1980; Istenič and Šmit, 2007). The beginnings of the use of brass by the Romans in the southern Alps is attested around 60 BC, and the first Roman brass coinage was minted around 45 BC by the dictator Julius Caesar, and it is thought by Grant (1946) that this minting of brass under Caesar began in Cisalpine Gaul at Milan. If this is correct, the choice of northern Italy for the production of the first brass coins could be related to the city's close geographic proximity to zinc sources in the Bergamasque Alps. It should be said that the only working mint in Italy during the Republican and the early Imperial period was in Rome, but Caesar had itinerant mints for his army usage, but they were not the official mint.

Despite the significance of the inception of the Roman brass industry, an industry that would ensue for many centuries, there has been little attention given to the location of potential sources of zinc ore in Cisalpine Gaul. Tizzoni (1996) was the first to suggest a connection between Roman zinc metallurgy in Milan and a potential zinc source in the Bergamasque Alps, ca. 36 km north of Bergamo in the Gorno Pb-Zn ore district. Consulting the geological literature, deposits of zinc ore can be found spread from the eastern branch of Lake Como eastwards to Gorno in the Val Seriana (Assereto et al., 1979; Rodeghiero et al., 1986). The Gorno district is a dense concentration of Triassic Mississippi-Valley-Type zinc-bearing mineralisations between the Val Brembana and Val Seriana, respectively from the villages of Dossena in the west and Gorno in the east. The Gorno district, encompassing the deposits of Dossena and Gorno, is the single major zinc ore source in the Bergamo region, and for that matter in continental Italy, and in the early 20th century it was named among some of the largest zinc mining regions in the world, supplying both calamine and sulphide ore (Moulden, 1916).

The zinc mineralisations of the Gorno Pb-Zn district were identified as the possible source mentioned by Pliny and Biringuccio. There are numerous traces of earlier mining throughout the Gorno Pb-Zn, but in the scope of recent preliminary investigations, traces near the village of Dossena were examined. Dossena, perched on the ledge of the Val Brembana is known for its zinc and fluorite mining tradition, which ended in 1981. Recent archaeological field surveys around Dossena have revealed extensive traces of mining, which include open-cast trenches, accessible galleries and collapsed mineshafts (*Pingen*) and working traces include fire-setting and pick-work (Maass et al., in press). The vast majority of mines was discovered in areas where secondary zinc and copper-zinc ores can be found. Due to the non-invasive nature of the surveys, it was not possible to date the mining activity more specifically than prior to the use of explosives. Nevertheless, evidence of Roman mining in the wider Gorno ore district is suggested by two denarii of the emperor Galba (68-69 AD) reported to have been found in an ancient mine gallery at Val del Riso (Alberti and Cessi, 1927).

More intensive archaeological surveys are currently being planned. It is expected that there will be little evidence for Roman zinc metallurgical processes near the mines because brass production was probably carried out in major urban workshops, such as Milan, where there is growing evidence for a Roman brass making industry (Tizzoni, 1996; Ceresa Mori and Cucini, 2012). Archaeometallurgy, more specifically lead isotope studies, may therefore play an important role in helping to substantiate the use of secondary zinc ore from the Dossena-Gorno area by the Romans and to identify when and to what extent it may have been used. The present archaeometallurgical study has two major goals: the first is the characterisation of the zinc ore, mineralogically, chemically and isotopically, and second, to compare the isotopic signature of the zinc ore with the lead isotope ratios of Roman brasses and bronzes to either confirm or refute possible relationships with the ore from the Bergamasque Alps and other potential zinc sources.

METHODS

In the scope of this preliminary study, zinc ore samples were provided by the Miniere di Dossena Cultural Association, and ten were selected for characterisation, ranging from secondary zinc ore, mixed secondary copper-zinc ore and a primary sulphide ore (Table 1). The ore samples are from old collections of the Museo di Dossena, most of which were surface collected, and it cannot be said for certain that the ore samples are representative of the ore used in the past, nor can it be said that the ore from the Dossena area are representative of the entire Gorno Pb-Zn district. The zinc ore fragments weighed between 100-1000 grams and the two secondary copper-zinc ores were around 20 grams each. At the laboratory of the Deutsches Bergbau-Museum Bochum, 20-100 grams of ore were crushed and pulverised for mineralogical characterisation by X-ray diffraction with a PANalytical X'Pert instrument (XRD). Ore powder was digested in a microwave (μPREP-A, MLS GmbH) in concentrated hydrofluoric acid and nitric acid followed by a second run with the addition of boric acid (50 g/l). Solutions were measured by inductively coupled plasma mass spectrometry (ICP-MS, Thermo Fisher Scientific, Element XR) for quantitative elemental analysis. The lead isotope ratios were measured by multicollector ICP-MS (Thermo Fisher Scientific, Neptune) at the Goethe-Universität Frankfurt am Main, and the methodology can be found in Klein et al. (2009).

RESULTS

The XRD of ore identified a range of zinc, lead and copper-bearing minerals together with gangue components (Table 2). The dominant ore phases are smithsonite, hydrozincite and hemimorphite. Macroscopically, smithsonite and hemimorphite specimens have botryoidal or vuggy textures and can range from orange, bluish-green, and shades of grey, while hydrozincite is typically white and appears chalky. Mixed secondary copper-zinc ore minerals, such as aurichalcite and rosasite, were found in two of the ore fragments intergrown with secondary zinc minerals. Aurichalcite and weathered oxidic copper minerals occur much less frequently than secondary zinc ore, though they can be found in the deposits at Dossena with little effort, mostly visible as thin infilled cracks both within ancient mines and in rock outcroppings and as tailings; pieces of mixed copper-zinc ore larger than a couple centimetres in thickness are rare. Lead minerals are commonly present in the ore pieces; in the calamine, lead is usually present as cerussite and occasionally as galena. The gangue material is calcite, quartz and fluorite.

The elemental composition of ore samples can be found in Table 3. The low totals of many of the ore samples is due to the presence of carbonate and hydroxide compounds. All the ore samples are silica poor except for DS1b, which is nearly pure hemimorphite. The two samples with aurichalcite clearly have elevated copper

Table 1. List of ore samples. Vaccareggio samples (VR) and Paglio Pignolino samples (DS).

Sample Nr.	Type	Mass (g)
VR5a	Calamine	147
DS4ac	Calamine	215
DS4bd	Calamine	108
VR1a	Primary Sulphide Ore	324
DS4aa	Calamine	930
VR2b	Calamine	175
VR3	Calamine	272
DS1b	Calamine	400
VR4a	Secondary Zinc-Copper Ore	24
VR4b	Secondary Zinc-Copper Ore	19

Table 2. XRD results of the ore samples from Vaccareggio and Paglio Pignolino.

Mineral	Formula	VR5a	DS4ac	DS4bd	VR1a	DS4aa	VR2b	VR3	DS1b	VR4a	VR4b
Smithsonite	$ZnCO_3$	X	X	.	X	.	X	X	.	X	X
Hemimorphite	$Zn_4Si_2O_7(OH)_2{\cdot}H_2O$	.	X	X	.	X	X	X	X	.	.
Hydrozincite	$Zn_5(CO_3)_2(OH)_6$	.	X	.	.	X	X	.	.	.	.
Sphalerite	(Zn,Fe)S	.	.	.	X	.	.	.	.	.	.
Aurichalcite	$(Zn,Cu)_5[(OH)_3\vert CO_3]_2$	.	.	.	.	.	.	.	.	X	X
Rosasite	$(Cu,Zn)_2(CO_3)(OH)_2$	.	.	.	.	.	.	.	.	.	X
Cerussite	$PbCO_3$	.	.	X	.	.	X	X	X	X	.
Galena	PbS	.	.	X	X	.	X	.	.	.	.
Vaterite	$CaCO_3$	.	.	.	.	X	.	.	.	.	.
Calcite	$CaCO_3$	X	.	.	X	.	.	.	X	.	.
Quartz	SiO_2	.	X	X	X	.	.	X	X	.	.
Fluorite	CaF_2	.	.	.	X	.	.	.	.	.	.

contents of around 6 wt% and are associated with smithsonite and minor amounts of cerussite, hydrozincite and hemimorphite. The lowest lead content is found in an exceptionally pure smithsonite (0.18 wt%), however this is unusually low; most calamines have more than half a percent and can be as much as 50 wt% lead, showing natural blends from zinc to lead-dominated parts. Regarding the trace elements, the significant elements present are arsenic, antimony, silver and strontium. The lead isotope ratios of the ore samples are presented in Table 4. The model ages of the ores are roughly between 200 and 320 million years, and the ores have U/Pb values that range from common terrestrial lead, ca. 9.75, to 10 based on the model of Stacy and Kramers (1975). One lead isotope analysis of galena from Val Vedra in the Gorno district has been published (Köppel and Schroll, 1985) and has comparable $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios but slightly higher $^{208}Pb/^{204}Pb$ ratios, but it is not certain if this is the result of differences in analytical methodology or actual heterogeneity within the wider ore district.

DISCUSSION

Secondary Zinc Ore and Aurichalcite

It is generally accepted that the Romans produced brass by direct cementation in the western provinces, meaning that they used calcined calamine as opposed to sublimated zinc oxide (Craddock and Eckstein, 2003). While by definition, calamine is a mixture of zinc carbonate and zinc silicates (i.e. hemimorphite), only non-silicates like smithsonite and hydrozincite can contribute to the zinc content of brass during cementation. The analyses show that seven of the nine secondary ore fragments are predominately smithsonite and/or hydrozincite with minor impurities from calcite and silicate intergrowths and contain some amounts of iron and lead. Iron and lead will, in part, enter into the brass alloys during cementation (Bourgarit and Thomas, 2015), while silicates and calcite, when present in minor amounts, are likely to react with charcoal ash to form a dross or slag. Thus, the presence of relatively clean zinc carbonate ore, easily found as pieces of hundreds of grams to kilograms in size, is a positive indication that the calamine found at Dossena could have been used for brass making in antiquity.

Regarding the mineral aurichalcite, in the Roman and later periods, oxidic copper-zinc ores are likely to have played no role in brass technology, but it has been postulated that mixed copper-zinc ores may have been the earliest ores used to make brass alloys and belong to a technology that long predates cementation (Craddock, 1978). Although the process of making brass from such ore is evidently difficult to control, experiments have shown that it is possible to produce brass directly from mixed secondary ore (Moulden, 1916; Zwicker et al., 1985). In Italy, only two examples of brass are known that pre-date the Romans; these are Etruscan brasses with 6-12 wt% zinc and are thus thought to have been made in this fashion (Craddock, 1978; 1980).

Lead Isotopes and Provenance of Roman Brass

Copper, Brass and Bronze

In order to interpret the lead isotope ratios of Roman brass it is first necessary to deconstruct the alloy into its parts. It is known that lead found in brass can have three origins: 1. lead in the copper, 2. lead associated with the zinc source and 3. mixing with lead-bearing alloys whether it be contamination or intentional alloying. It is

Table 3. Quantitative ICP-MS analysis of Dossena ore.

Wt%	VR5a	DS4ac	DS4bd	VR1a	DS4aa	VR2b	VR3	DS1b	VR4a	VR4b
Na_2O	0.01	0.02	0.01	0.004	0.03	0.01	0.003	0.02	0.01	0.01
MgO	0.08	0.04	0.04	0.05	0.05	0.08	0.05	0.07	0.03	0.01
Al_2O_3	0.07	0.07	0.26	0.08	0.05	0.25	0.07	0.40	0.04	0.14
SiO_2	0.58	1.32	5.19	0.58	5.87	1.02	3.84	42.0	0.84	1.14
P_2O_5	0.01	0.02	0.06	0.01	0.02	0.01	0.05	0.02	0.01	0.03
K_2O	0.03	0.04	0.07	0.06	0.05	0.08	0.05	0.11	0.03	0.05
CaO	0.06	0.68	0.20	3.23	0.63	0.55	0.12	0.90	0.19	0.64
TiO_2	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	0.01	<0.001	0.004
MnO	0.03	0.10	0.003	0.02	0.20	0.05	0.001	0.09	0.04	0.05
FeO	0.32	0.48	1.17	1.79	0.08	1.06	1.15	0.84	0.04	0.20
ZnO	65.8	67.0	13.5	61.7	75.7	50.4	71.3	50.4	62.3	66.4
BaO	<0.005	<0.005	0.006	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
S	0.07	0.14	0.76	24.4	0.34	0.93	0.07	0.10	0.17	0.08
Cu	0.05	0.35	0.02	0.05	0.46	0.24	0.01	0.07	6.39	5.80
Pb	0.18	0.65	53.4	6.63	0.53	18.2	3.15	1.40	1.19	0.86
Total	67.29	70.94	74.70	98.63	84.00	72.91	79.84	96.40	71.31	75.40
ppm										
V	2	3	5	7	3	6	5	15	3	30
Cr	<1	3	6	2	2	3	5	5	1	2
Co	<1	<1	<1	4	5	<1	<1	<1	<1	20
Ni	10	<3	<3	<3	<3	<3	<3	<3	3	45
As	<5	25	65	<5	10	440	45	60	35	240
Se	<5	<5	20	25	<5	<5	<5	6	<5	<5
Sr	2	10	350	8	10	860	45	20	3	15
Ag	<5	50	35	80	<5	65	50	170	<5	<5
Sn	2	1	1	2	<1	<1	1	6	<1	<1
Sb	210	140	310	160	100	480	520	940	350	460
Te	6	4	3	3	3	2	2	2	2	2
Bi	<1	<1	5	<1	<1	2	<1	<1	<1	<1
U	1	5	5	1	350	2	1	1	0.2	20

therefore important to identify which contributes the most significant portion of lead to the brass. In the following discussion of copper alloys, the definitions of Bayley and Butcher (2004) will be used, and strictly speaking, 'leaded' alloys (>4 wt% Pb) are excluded from the discussion.

Large datasets exist that allow something to be said about the source of the lead found in Roman brass, at least for the early Imperial Roman period. Roman copper is typically lead-poor when compared to brass (Figure 1). In fact, the *aes* coinage under Augustus (n=120) is so poor in lead that 94% of copper coins contain less than 0.05 wt% lead (Klein and von Kaenel, 2000; Klein et al., 2012). Imperial Roman copper ingots (n=116) found in shipwrecks in the Western Mediterranean and at the Gallo-Roman settlement of Alesia have slightly more lead, but still, 80 % of ingots had under 0.1 wt% lead and 93 % under 0.2 wt% lead (Klein et al., 2007; Rabeisen and Menu, 1985; Rico et al., 2006). If these values are

Table 4. Lead isotope ratios of Dossena ore samples.

Sample	206/204	2σ	207/204	2σ	208/204	2σ	207/206	2σ	208/206	2σ
VR5a	18.395	0.004	15.665	0.004	38.518	0.008	0.8516	0.0001	2.0940	0.0001
DS4ac	18.396	0.006	15.665	0.006	38.519	0.019	0.8515	0.0001	2.0939	0.0004
DS4bd	18.385	0.007	15.620	0.006	38.472	0.017	0.8496	0.0002	2.0926	0.0004
VR1a	18.394	0.006	15.661	0.007	38.513	0.017	0.8514	0.0002	2.0936	0.0004
DS4aa	18.394	0.008	15.680	0.014	38.546	0.025	0.8524	0.0006	2.0955	0.0008
VR2b	18.408	0.010	15.625	0.010	38.491	0.023	0.8488	0.0002	2.0910	0.0004
VR3	18.382	0.008	15.676	0.007	38.536	0.017	0.8527	0.0002	2.0963	0.0004
DS1b	18.369	0.005	15.654	0.008	38.501	0.018	0.8522	0.0003	2.0959	0.0004
VR4a	18.396	0.007	15.642	0.006	38.533	0.017	0.8503	0.0002	2.0947	0.0004
VR4b	18.382	0.006	15.652	0.007	38.540	0.015	0.8515	0.0002	2.0965	0.0004

compared to the brasses from the Augustan *sestertii* and finds from the Augustan-period Roman fortress at Haltern (n=180), it is apparent that none of the brasses have less than 0.05 wt% lead and are more often in the range of 0.2-0.35 wt% (Riederer, 1995; 2002a). Riederer (2002a) also shows that several of the unalloyed copper objects from Haltern have lead in the low hundreds of ppm range, consistent with the copper analyses of Klein et al., so we can be confident that the discrepancy between the lead contents of copper and brass is real and not likely to be attributed to the applied analytical methods. The earliest Roman brass military brooches dating to the middle of the 1st century BC are much less standardised than Augustan brasses and often contain higher amounts of lead (compare Istenič and Šmit, 2007).

Under close examination, the analyses of Riederer (2002a) provide further information regarding the alloying of brass with tin-bearing alloys during Augustan times. While more than half of the brasses from Haltern have tin contents lower than the detection limit, it is not unusual for brasses to contain tin in the low percentage range. Above about 1.25 wt% tin, the data show that the lead content of the brasses becomes much more variable and frequently elevated; this means that an impact from mixing with leaded tin-bearing alloys may begin to be visible above this point (Figure 2). Below this point, the brasses almost consistently have between 0.1-0.5 wt% lead. The presence of small amounts of tin in the brass is

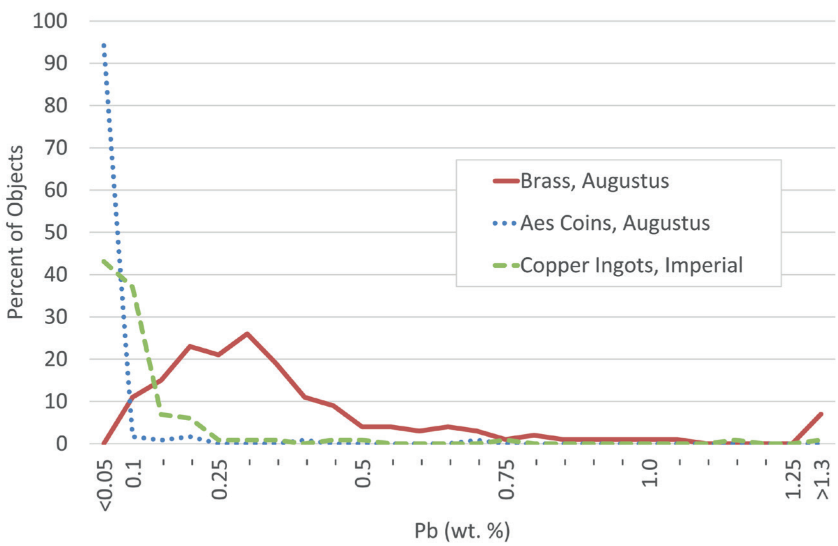

Figure 1. Comparison of lead contents of Augustan copper *aes* (n=120) and Imperial copper ingots (n=116) and Augustan brasses (n=180) (after Rabeisen and Menu, 1985; Riederer, 1995; Klein and von Kaenel, 2000; Riederer, 2002a; Rico et al., 2006; Klein et al., 2007; Klein et al., 2012).

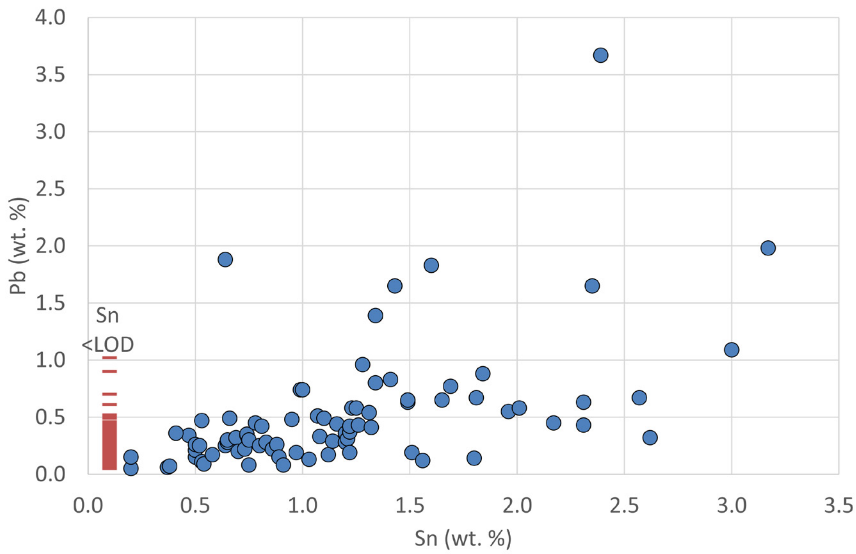

Figure 2. The lead and tin contents of Augustan brasses after the analyses of Riederer (1995, 2002a). A large number of the analyses show tin contents less than 0.25 wt%, below detection (n=100), while 80 objects had detectable tin, 0.4-3.2 wt%. All brasses had detectable quantities of lead.

not necessarily proof that the alloy was made of recycled metal; Albertus Magnus wrote in the 13th century AD that small amounts of tin could be added to brass to make the colour appear more like gold (Wyckoff, 1967), and this was evidently practiced by the Romans for certain types of brass objects, such as the standardised 1-2 wt% tin found in a sizable number of Aucissa and eye brooches and the uniform 3-4 wt% tin found in the Augsburger brass eye brooches (see analyses of Riederer, 2002b).

The studies show a clear tendency that brasses contain more lead than contemporary copper. If a rather generous arbitrary value of 0.02 wt% is given for all copper where lead is below the detection limit, then the average lead contents of the ingots is 0.10 wt% (n=116) and for the *aes* coins, 0.03 wt% (n=120). In contrast, the average lead content of brasses (with under 1.25% tin) is 0.29 wt% (n=151) and is thus three to ten times the amount in unalloyed copper. Naturally, it must be considered whether the copper ingots and *aes* coinage analysed are representative of the copper used for the purpose of brass making, however there is no evidence at the moment suggesting the contrary. The most rational explanation of the elevated lead contents of low-tin brasses is a contribution of lead from calamine, which has implications for interpretation of lead isotope ratios of brass.

Lead Isotope Ratios and Zinc Sources

It must be first stated that the lead isotope analysis of Roman brass is in a state of infancy, and four studies have been found on which comparisons can be made. The first study is of Beck et al. (1985) in which two brass objects from the site of Alesia were analysed (Cat. No. 11279 and 11280). The second study is of the brass ingots from the Aléria shipwreck off of Corsica dated to the end of the 2nd century AD (Hanel and Bode, 2016). The brass of these 21 ingots is exceptionally fine for the period and must have been newly produced, though the ingots have a notably higher lead content than early Imperial brasses. The study includes seven lead isotope analyses. The third study is of Durali-Müller (2005), which has elemental compositions and lead isotope ratios of twelve brasses and four bronzes from Roman Mainz. The final two studies are more recent: Reichmann et al. (2019) contributes three lead isotope analyses from Roman finds from the harbour at Duisburg/Krefeld-Gellep and Merkel (in press) offers 20 analyses of Roman brass and copper/brass alloys from the Germanic goldsmithing workshop at Elsfleth-Hogenkamp dated primarily to the 2nd-3rd century AD. In the latter study, ten 2nd century brass and copper/brass *sestertii* coins are among the objects analysed as well as two eye brooches of the 1st century AD and a crossbow brooch with onion-head terminals, ca. 3rd-4th century AD.

As mentioned with the Aléria brass ingots, mid-to-late Imperial brasses tend to have higher lead contents than Augustan brasses, and, with exception of the Aléria brass ingots, the later brasses tend to have lower zinc contents and more irregular tin contents probably as a result of intentional dilution and alloying. Additionally, concerning the higher lead contents of mid-to-late Imperial brass, particularly visible in the case of Aléria ingots, the calamine used during cementation may have been richer in lead and this lead may carry over into all manners of copper-zinc alloys.

All of the above studies, with exception of the Aléria ingot study, also contain lead isotope analyses of bronzes, and a comparison between Roman brasses and bronzes can be found in Figure 3. There are two notable tendencies:

1. that there are many bronzes and brasses that cluster together at around 0.847 ($^{207}Pb/^{206}Pb$) and 2.088 ($^{208}Pb/^{206}Pb$), which appears to represent the "melting pot" of Roman copper alloys, leaded copper-based alloys alike (compare data from Beck et al., 1985; Brill and Shields, 1972; Merkel, in press); however,

2. many of the brasses plot on a line above the bronzes and this line contains nearly all of the sestertii coins and several of the brass ingots from Aléria. Within the dataset, no relationships among the tin and lead contents and the lead isotope ratios could be found in the brasses and copper/brasses except that all with $^{207}Pb/^{206}Pb$ ratios greater than 0.851 have low quantities of lead, between 0.07 and 0.25 wt%.

In order to attempt to make sense of the differing distribution of brasses and bronzes, it is necessary to compare the lead isotope ratios to zinc ore deposits. The locations of the most important non-sulphide zinc deposits in Europe are described by Boni and Large (2003). Concerning the Roman world, lead isotope data exists for many of the potential areas where large-scale zinc ore deposits occur: the Aachen-Kelmis area (geology, see Coppola et al., 2008; data, see Cauet, 1983; Bielicki and Tischendorf, 1991; Krahn and Baumann 1996; Durali-Müller, 2005; Bode, 2008; Chatziliadou, 2009; Merkel, 2016) and Wiesloch (Ströbele et al., 2012), Spanish deposits, for example at Cartagena (Graeser and Friedrich, 1970; Arribas and Tosdal, 1994; Stos-Gale et al., 1995) and the Reocin deposit on the south coast of the Bay of Biscay (archaeology, see Davies 1935, 97; data, see Velasco et al., 1996; 2003; see), Laurion (geology, see Skarpelis and Argyraki 2009; data, see Oxalid:Greece) and Thassos in the Aegean area (Oxalid:Greece), Iglesiente in southwest Sardinia (geology, see Boni et al., 2003; data, see Boni and Koeppel, 1985; Ludwig et al., 1989; Stos-Gale et al., 1995; Valera et al., 2005) and the Gorno district north of Bergamo (this study; Köppel and Schroll, 1985). Furthermore, lead-zinc deposits occur in the south of France, but little is known as to extent they were used in antiquity; importantly, there is a report of Imperial Roman brass cementation vessels being found in Lyon (Picon et al., 1995). The Massif Central has a widely diverse lead isotope ratio distribution, and since the source of zinc used in Roman Lyon has not been characterised, the Massif Central must be left out of the discussion until more information is gathered.

Comparing the Roman brasses to potential calamine sources, no relationships can be found with sources in the Aegean, Sardinia, Wiesloch and Reocin. Calamine sources that might contribute to the lead contents of the brasses are the Gorno ore district (Dossena), Aachen-Kelmis and Cartagena (Figure 4). It must be born in mind that the large cluster of brasses bordering on the Aachen field are also

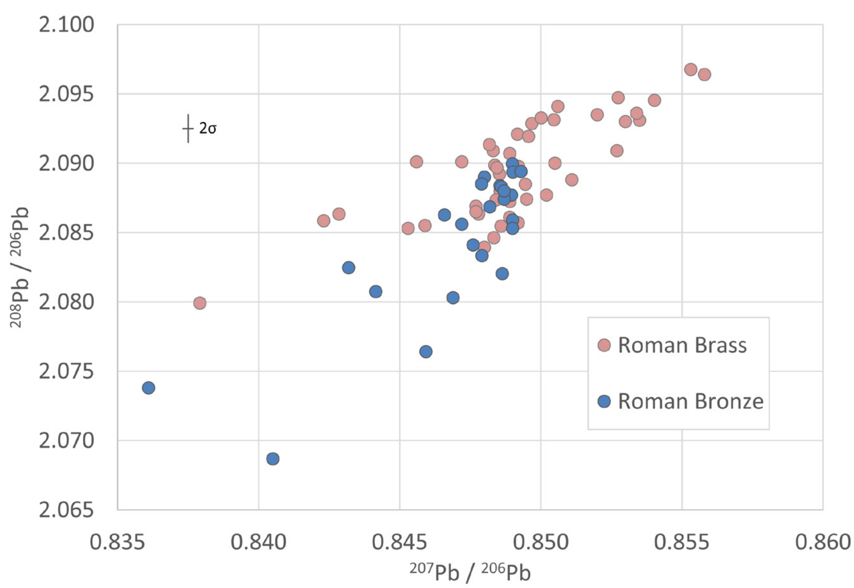

Figure 3. Comparison of the lead isotope ratios of Roman bronzes and brasses (Beck et al., 1985; Durali-Müller, 2005; Hanel and Bode, 2016; Reichmann et al., 2019; Merkel, in press).

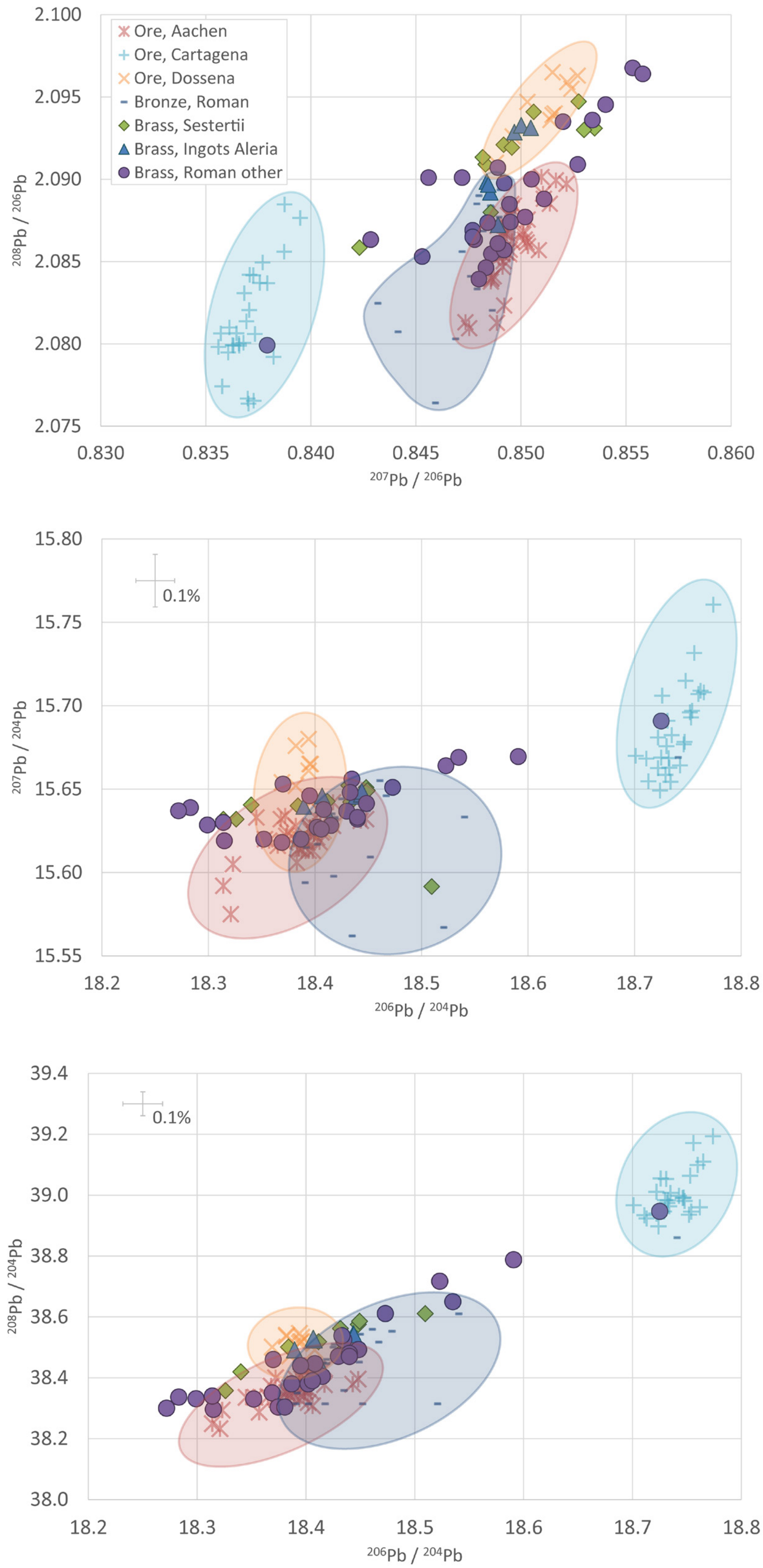

Figure 4. Comparison of Roman brasses to zinc ore from Dossena, Gorno ore district, northern Italy (this study) and published ore data from the Aachen area (Germany/Belgium) and Cartagena (Spain), see references in text.

consistent with Roman bronzes, as previously mentioned, possibly representing the 'melting pot' of Roman copper-based alloys; since nearly all these objects have between 1 and 3% lead, it is not possible with the present dataset to distinguish brasses made with calamine from Aachen from recycled metal. The Dossena ore is consistent with 5 of the 10 *sestertii* coins minted in Rome (2nd century AD), 2-3 of the brass ingots of Aléria and a brass casting sprue from Elsfleth-Hogenkamp that has the composition comparable to a 2nd century *sestertius*, thus probably a melted *sestertius*. One piece of brass, a sheet (10 wt% Zn, 0.56 wt% Sn and 0.10 wt% Pb) from the 2nd-3rd century AD workshop at Elsfleth-Hogenkamp, has lead isotope ratios consistent with Cartagena in southeast Spain and is the first indication that some Roman brass may have a Spanish source. A number of brasses fall on lines between Cartagena and Dossena or Dossena and Aachen (or mixed copper-based alloys) and could constitute mixtures. One group of brasses, however, fall outside this framework, and these are the low-lead brasses with $^{207}Pb/^{206}Pb$ ratios greater than 0.851. In this group are two 2nd century *sestertii*, a possible *sestertius* fragment (meant for recycling) and a 1st century eye brooch. The low lead contents of these brasses could mean that the lead isotope ratios are noticeably influenced by lead coming from the copper, copper of an older geological age, for example, from the central and southwest Spain (see Klein et al., 2004, 2009).

CONCLUSIONS

Pliny and Biringuccio suggested that zinc ore was mined in the Bergamasque Alps in antiquity and later periods, and regarding regional geography and geology, the zinc deposits of the Gorno ore district is the only major source that can match the references. Though there are extensive and diverse traces of pre-modern mining in the calamine deposits at Dossena, hinting at multi-phase exploitation, there is still very little information on the chronology. Through archaeometallurgical analysis of ore and brass, this study has shown that the Gorno ore district has the potential of being an important source of zinc ore, for example, for the production of 2nd century brass *sestertii* minted in Rome. It is important to reiterate that the lead isotope analysis does not conclusively prove that the calamine of Dossena, Gorno district, was used by the Romans; it does confirm the possibility that it could have been used.

Furthermore, this study provides a theoretical foundation for the interpretation of the lead isotope ratios of Roman brasses, but two suggestions can be made to strengthen this basis:

1. the lead isotope analysis of Roman cementation vessels would be especially useful to characterise brass production centres such as at Milan, Lyon and the Aachen/Lower Rhine area (for example Xanten, Rehren, 1996), as the lead residues in the vessels should mirror the exact mixture of lead found in the brass produced, and

2. a sampling strategy should be developed aimed at specific types of non-mixed brass alloys that can be expected to provide indications of the zinc source, such as Roman military brooches and earliest *sestertii* coins, for example, of Julius Caesar and Augustus. If the inception of early Roman brass production can be proven to be connected to the mining activity in the Bergamo region, this would solve a standing problem for Roman archaeology and archaeometallurgy alike.

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