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COPPER PRODUCTION AND TRADE IN THE NIARI BASIN (REPUBLIC OF CONGO) DURING THE 13TH TO 19TH CENTURIES CE: CHEMICAL AND LEAD ISOTOPE CHARACTERIZATION*

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In Central Africa, copper ore occurs in only a few locations and copper appears to have been a scarce commodity in the past—contrary to iron, which is attested more widely and earlier in the sub-Saharan archaeological record. This paper presents the first detailed characterization of an early copper-working region in Central Africa. Located along the southern border of the Republic of the Congo, the Niari Basin has revealed several copper production sites ranging from the 13th to the 19th century CE. *The evidence, specifically in the Mindouli, Mfouati and Boko-Songho areas, includes various production remains as well as different types of copper ingots and artefacts. In the context of a broader copper technology study, the chemical and lead isotope characteristics of the ore deposits in this region are presented. The results of the chemical and lead isotope analyses of copper objects and production remains from archaeological sites are then interpreted against this geological background data, with an emphasis on copper provenance features. Combining these results with archaeological and historical evidence for regional metallurgical activity, new and significant insights are given on the production of copper in the Niari Basin, emphasizing the potential of this research for forthcoming work on copper use and trade in a wider Central African context.*

KEYWORDS: ANCIENT METALLURGY, LEAD ISOTOPES, CHEMICAL ANALYSIS, PROVENANCING/SOURCING, AFRICA, CONGO

INTRODUCTION: COPPER IN CENTRAL AFRICA

For centuries, copper was considered a valuable metal and was used in ornaments or exchange objects in many parts of Central Africa (e.g., Martin 1972; Herbert 1984; de Maret 1985, 1992, 1995). In contrast to iron ores, which are widely and easily available (the average abundance of iron in the Earth's crust is ~5%, but that of copper is only 0.006%), copper mineralizations occur in only relatively few locations across Central Africa (Miller and van der Merwe 1994; Bisson 2000). The largest are the Central African Copperbelt deposits in southern Katanga (Democratic Republic of Congo: DRC) and northern Zambia, though smaller deposits occur, for example, in the Niari Basin (Republic of Congo), the Central Kongo province (DRC) and northern Angola. Most of these were exploited long before industrial exploitation in the 20th century

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and their copper was traded throughout the Congo Basin area and neighbouring regions. Herbert (1984: 161) also mentions copper from the Hufrat en-Nahas deposits (South Sudan) reaching the northern part of the Congo Basin.

Very little information is available, however, concerning the exchange networks of this copper before the late 19th century European colonization. Their identification would be very helpful for reconstructing ancient socio-economic networks in western Central Africa, particularly in establishing the origin of copper used in areas where several deposits may have been available. Of particular interest here is the possible involvement of Niari in networks linked to different polities, similar to those identified for the Copperbelt.

Indeed, the Copperbelt was integrated in large-scale exchange networks from the early second millennium AD onwards. Its copper reached (*inter alia*) the Indian and Atlantic coastal trade during the 18th century, possibly linked to the development of regional polities (Wilson 1972; de Maret 1992: 224). For recent periods (14th–19th centuries), the spatial distribution of the copper cross-shaped ingots ('*croisettes*')—used as currency—reflects sociopolitical spaces and boundaries linked to these polities (Nikis and Livingstone Smith 2017).

The Niari Basin, situated on the periphery of several polities in western Central Africa (Fig. 1), is mentioned in historical sources as having been a copper supplier since the 16th century (e.g., Pigafetta and Lopez 1591; Dapper 1686). These polities, which developed around the first centuries of the second millennium AD, participated in regional and, from the 15th century, international trade with Europe. The coastal trade involved mainly slaves but, equally, goods such as ivory, raffia cloth, redwood and copper were exchanged and most probably followed trade routes that were already in existence prior to European contact (Martin 1972).

During the 15th century, Mbanza Kongo, capital of the Kongo kingdom, had access to the Niari Basin deposits, in addition to Bembe and other minor outcrops in Angola. Moreover, control over the Niari copper deposits may have been a political issue, leading to conflict between the Nsundi province (the northern Kongo kingdom) and the Anzico kingdom, which controlled trade routes running through the Malebo Pool and the surrounding area (Hilton 1985: 55). In the 17th century, the Loango kingdom (on the Atlantic coast) sent smelters to get copper from the Niari area (Dapper 1686). In the same era, copper also seems to have reached the Ngoyo and Kakongo kingdoms, two coastal states strongly linked to the Kongo kingdoms. In the 19th century, Niari copper is reported as having reached the Inner Congo Basin (Coquilhat 1888: 160).

Unfortunately, the Niari Basin has until recently received very little archaeological attention (Lanfranchi and Manima-Moubouha 1984; Manima-Moubouha 1988), and studies of copper and its exchange networks are still mostly based on historical sources (limited to the 16th century) or on oral history (Martin 1972; Ndinga-Mbo 1984; Hilton 1985; Volavka 1998). The only attempt to provenance copper in the area (works of art from the Ngoyo kingdoms) by lead isotope and chemical analyses was based on very few surface ore samples from Boko-Songho and ethnographic objects (sometimes poorly contextualized), and was rather inconclusive (Farquhar 1998; Franklin and Volavka 1998).

Very few other African copper production regions have hitherto been analytically characterized in an archaeometallurgical framework. Although ancient copper samples from Kansanshi (Zambia) and Kipushi (DRC) were chemically analysed by Bisson *et al.* (1978), the only lead isotope (LI) data currently available for Central Africa relates to the geological deposits of the Central African Copperbelt (see the 'Results: ores' section), while no data is available for any Central African archaeological copper remains. More broadly, the limited existing LI data of archaeological African copper comes from West Africa (Craddock and Hook 1995; Goucher *et al.* 1976; Willet and Sayre 2006) and South Africa (Molofsky *et al.* 2014). The first data set covers West African

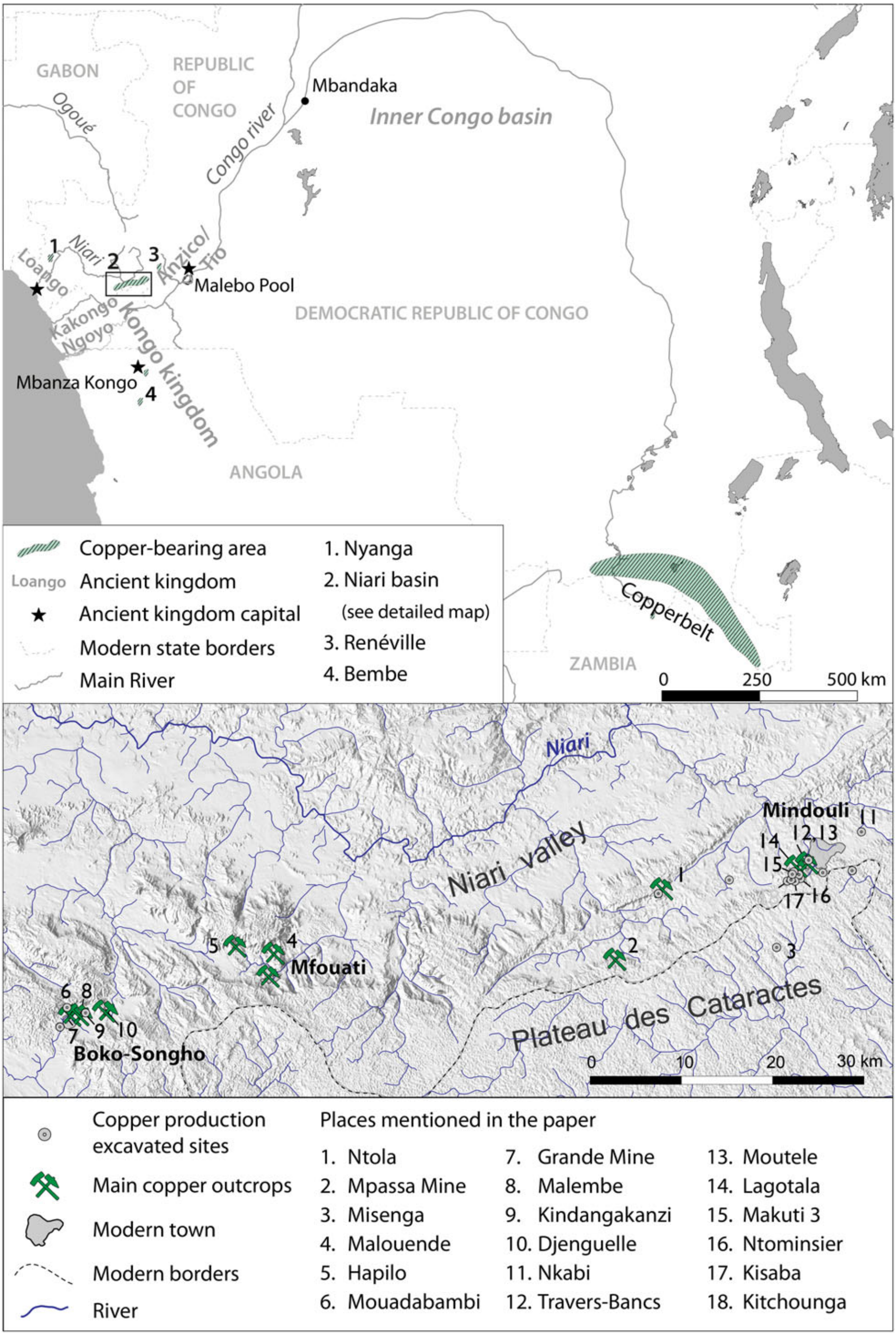

Figure 1 *An overview map highlighting the Niari Basin and surrounding area, with kingdoms mentioned in the text (top) and the different areas within the Niari Basin (bottom). A more detailed map is included in the OSI. [Colour figure can be viewed at wileyonlinelibrary.com]*

copper alloys (e.g., the famous Igbo Ukwu and Ife castings) and a selection of African ores (from Nigeria, Tunisia and Morocco). Perhaps the best understood ancient copper, and particularly tin and bronze, production is situated in South Africa, where the most extensive research has been carried out (e.g., Friede and Steel 1975; Grant 1999; Miller *et al*. 2001; Greenfield and Miller 2004; Chirikure *et al*. 2010; Heimann *et al*. 2010; Thondhlana *et al*. 2016).

Consequently, the data collected during recent archaeological and geological fieldwork in the Niari Basin (Nikis *et al*. 2013; Nikis and Champion 2014; Nikis and De Putter 2015; Nikis, in press a), with additional material from Maurits Bequaert's 1951 unpublished excavation in Misenga (currently conserved in the Royal Museum for Central Africa (RMCA), Belgium), provide a unique opportunity to realize an integrated analysis comprising both geological and archaeological materials. The surveys, geological observations and excavations mainly took place around Mindouli, Boko-Songho and, to a lesser extent, Mfouati (Fig. 1). Several periods of production, each one clearly distinct from the others in its manufacturing process (Nikis *et al.* in prep.) and associated ceramics (Nikis in press a), have been identified in the Mindouli and Boko-Songho areas. The basic characteristics of these production processes are summarized in Table 1, and examples of typical copper objects sampled in this study are illustrated in Figure 2. For a more detailed treatment of these metallurgical technologies and their products, see Nikis (in press a,b) and Nikis *et al*. (in prep.) and references therein.

Table 1 *An overview of metallurgical technology in Mindouli and Boko-Songho*

Area	*Period*	*Manufacturing process*	*Semi-finished products*	*Sites analysed (map number)*
Mindouli	*c*.13th–14th centuries	• Careful ore preparation • No furnace structures recovered • Smelting (?) or refining in large crucibles • Ingot casting in open moulds	Misenga-type ingots, transformed by hammering into elongated flat bars (*milambula*)	Misenga (3), Nkabi (11), Makuti 3 (15), Kisaba (17)
Mindouli	*c*.16th–18th centuries	• Smelting in partly buried furnaces • Slags with embedded copper prills • Refining in decorated ceramics (Moubiri type)	Unknown	Ntominsier (16)
Mindouli	19th century	• Rough preparation of ore • Smelting in partly buried furnace; large amounts of slag produced, some with gangue remains (+ lead or copper sulphides) • Refining in fairly standardized small crucibles	Unknown	Kitchounga (18)
Boko-Songho	*c*.15th–17th centuries	• Smelting in partly buried furnaces • Slags with embedded copper prills • Refining in decorated ceramics (Moubiri type)	Unknown	Kindangakanzi (9)
Boko-Songho	19th century	• Smelting in superstructure built furnace, with addition of lead (metal or sulphide) • Refining of the ingots in 'pierced crucible'	Mouadabambi-type ingots	Mouadabambi (6)

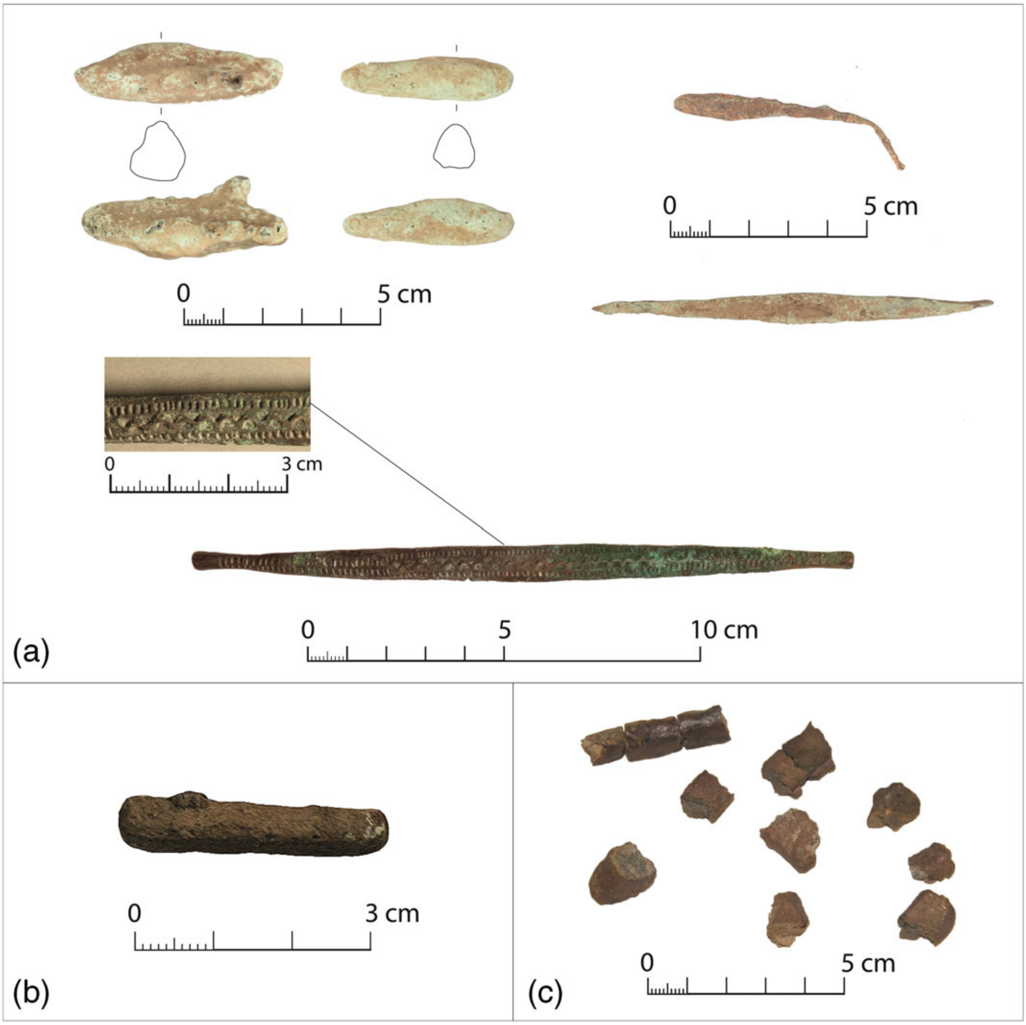

Figure 2 *Copper ingots. (a) Examples of the 13th–14th century production in Mindouli: Misenga-type ingots (top left), Misenga, DRC (KongoKing project 2014 excavation); ingots partially transformed into barrettes (top right), Nkabi, Republic of Congo (2015 excavation); a mulambula-type bar (middle right), Misenga, DRC (RMCA Inv. PO.0.0. 70475); and a decorated mulambula-type bar (bottom), Makuti, Republic of Congo (2013 excavation). (b) 19th century copper ingot from the Boko-Songho area, Mouadabambi, Republic of Congo (2014 excavation). (c) Fragmented copper ingots collected by C. Coquilhat between 1883 and 1891 in the Ubangi region, DRC (RMCA RGM 583). [Colour figure can be viewed at wileyonlinelibrary.com]*

This paper presents the results of elemental and lead isotope analyses of ore samples and well-contextualized archaeological copper artefacts, with an eye on copper provenance studies in the Niari Basin study area. It assesses whether the different zones within the Niari Basin have chemical or isotopic characteristics that may help future identification of changing exploitation of these zones throughout the past millennium, in relation to particular sociopolitical contexts. On a broader scale, the paper aims to assess how Niari Basin copper production may be distinguished from other production systems in Central Africa on the basis of such geochemical data, and sets a reference for future studies within the region (e.g., the forthcoming study of DRC *croisettes*: Rademakers *et al.* in prep.).

ANALYTICAL METHODS

An overview of all the samples discussed in this paper is presented in Table 2. Full lead isotope (LI) and chemical data are reported in the online supporting information (OSI), which includes a complete description of the sample preparation and the analytical methods.

LI ratios (Tables S1 and S2) were measured for all samples by multi-collector inductively coupled plasma – mass spectrometry (MC-ICP–MS), and are plotted in this paper with 95% confidence intervals (2σ): Figures 3 and 4 show selected LI ratio plots—additional presentations are included in the OSI.

Bulk chemical analysis of all ore and slag and of five metal samples was carried out by inductively coupled plasma – optical emission spectroscopy (ICP–OES). Ore, slag and three metal samples were further analysed by ICP–MS. Finally, all metal samples have been analysed by laser ablation ICP–MS (LA-ICP–MS). Elemental concentrations are reported either in wt% (as %) or $\mu g\ g^{-1}$ (full data are presented in Tables S5, S6 and S7).

RESULTS: ORES

The Niari Basin comprises several Cu–Pb–Zn deposits, notably at Mindouli and surroundings (M'passa Mine, Ntola), Boko-Songho and the Mfouati mines located to the west of Boko-Songho (Fig. 1). These deposits were exploited during the French colonial period for base metals and silver (Cosson 1955), but remain relatively poorly known and published (Buffet *et al*. 1987; Koud 1987). A reappraisal of these deposits is currently ongoing (De Putter and Nikis 2016; De Putter *et al.,* in prep.), and their detailed discussion is omitted here. A basic overview of the mineralization process is provided in the OSI.

Ore samples were taken from the three main deposit zones in the Niari Basin, and are discussed separately in detail in the OSI. Here, due to editorial constraints, discussion is limited to a broad overview.

Chemical and LI analysis has been performed for a total of 17 copper (malachite) and four lead (galena) ore samples (see Table 1). Further sulphide ores have been analysed and are presented in a separate paper focusing on the geological history of the Niari Basin (De Putter *et al.,* in prep.), together with complete trace element data for the ores presented here.

The LI ratios are presented in Figure 3. The ore results sections of the OSI highlight the most relevant elements towards understanding different ore formation conditions on the one hand, and distinguishing between different ore sources on the basis of trace elements that are less susceptible to heavy fractionation during metallurgical processes (see, e.g., Pernicka 1999, 2014; Ling *et al*. 2013, 2014; Rehren and Pernicka 2008) on the other. These two perspectives are considered through the complementary use of both chemical and LI data. Element to copper ratios (e.g., Ag–As–Pb/Cu) are particularly instructive for comparisons to metallurgical artefacts.

The geological age of these ores has been estimated (see the OSI). A rather consistent age of *c*.700 Ma is estimated for all Mindouli ores. The Mfouati and Boko-Songho galenas may be similarly dated, indicating a roughly contemporaneous formation of the base sulphides across the Niari Basin. The Mfouati and Boko-Songho copper minerals, however, were apparently formed at later hydrothermal fluid mineralization events than those at Mindouli, resulting in more radiogenic LI ratios. These variations are apparent from visual inspection of both the raw isotope ratio

Table 2 *An overview of the samples analysed in this study*

Geological Samples

Lab code	*Type*	*Site*	*Area*	*Description*
GPSNN 271	Malachite	Grande Mine	Boko-Songho	Laminar/botryoidal malachite coating in iron-rich laterite host-rock
GPSNN 271-3	Malachite	Grande Mine	Boko-Songho	Laminar/botryoidal malachite coating in iron-rich laterite host-rock
GPSNN272	Malachite	Malembe	Boko-Songho	Botroyoidal malachite + azurite
Malembe-1	Malachite	Malembe	Boko-Songho	Botroyoidal malachite in weathered argillaceous host-rock
Djen Geol	Malachite	Djenguele	Boko-Songho	Malachite and galena in highly weathered host-rock
Mpassa-1	Malachite	Mpassa Mine	Mindouli	Malachite and azurite in highly weathered porous host-rock
Mpassa-2	Malachite	Mpassa Mine	Mindouli	Malachite and azurite in highly weathered porous host-rock
MPA-3	Malachite	Mpassa Mine	Mindouli	Malachite and azurite in highly weathered porous host-rock
Ntola	Malachite	Ntola	Mindouli	Laminar/botryoidal malachite coating in weathered host-rock
NTO-3	Malachite	Ntola	Mindouli	Laminar/botryoidal malachite coating in weathered host-rock
GPSNN 259	Malachite	Lagotala	Mindouli	Thin malachite coating in highly weathered host-rock
GPSNN 259-6	Malachite	Lagotala	Mindouli	Malachite coating and later cuprite, dioptase
GPSNN 263	Malachite	Moutele 'faille'	Mindouli	Malachite coating (+ chrysocolla, dioptase) in weathered argillaceous host-rock
MOU-2	Malachite	Moutele	Mindouli	Botryoidal malachite in weathered porous host-rock
TRB-01	Malachite	Travers-Banc	Mindouli	Malachite and chrysocolla with quartz vein infilling
GPSNN 284	Malachite	Mfouati	Mfouati	Thin malachite coating in highly weathered, porous limestone host-rock
MAL-1	Malachite	Malouende	Mfouati	Laminar/botryoidal malachite in weathered argillaceous host-rock
GPSNN 289	Malachite	Mfouati/Hapilo	Mfouati	Malachite coating in silicified skeletal host-rock
BSD-01	Galena	Djenguele	Boko-Songho	Galena spots in weathered host-rock
NTO-02	Galena	Ntola	Mindouli	Galena cement in brecciated host-rock
MPA-01	Galena	Mpassa	Mindouli	Galena spots within massive sulfides
HAP-01	Galena	Hapilo	Mfouati	Galena spots within massive sulfides

Archaeological Samples

Lab code	*Type*	*Site*	*Area*	*Period - Context*
KNA 14-1	Cu fragment in slag	Kindangakanzi	Boko-Songho	15th-17th century CE - Excavation: Fill in of a copper smelting furnace
KNA 14-3	Malachite	Kindangakanzi	Boko-Songho	15th-17th century CE - Excavation (SVII -7cm): Metallurgical remains layer

(*Continues*)

Table 2 *(Continued)*

Archaeological Samples

Lab code	*Type*	*Site*	*Area*	*Period - Context*
KNA 14-2	Slag	Kindangakanzi	Boko-Songho	15th-17th century CE - Excavation (SVII -7cm): Metallurgical remains layer
MDB 14-1	Ingot fragment	Mouadabambi	Boko-Songho	19th century CE - Excavation (SIII -10cm): Metallurgical remains layer
MDB 14-3	Malachite	Mouadabambi	Boko-Songho	19th century CE - Excavation (SIII -10cm): Metallurgical remains layer
MDB 14-2	Slag (1)	Mouadabambi	Boko-Songho	19th century CE - Excavation (SIII -10cm): Metallurgical remains layer
MDB 14-2	Slag (2)	Mouadabambi	Boko-Songho	19th century CE - Excavation (SIII -10cm): Metallurgical remains layer
MSG70628-5	Barrette	Misenga	Mindouli	13th-14th century CE - M. Bequaert excavation, 1951
MSG70628-2	Ingot	Misenga	Mindouli	13th-14th century CE - M. Bequaert excavation, 1951
MSG70620	Bracelet	Misenga	Mindouli	13th-14th century CE - M. Bequaert excavation, 1951
KIS 14-1	Bar fragment	Kisaba	Mindouli	13th-14th century CE - Excavation (SIV -20cm): Metallurgical remains layer
KIS 14-4	Malachite	Kisaba	Mindouli	13th-14th century CE - Excavation (SIV -20cm): Metallurgical remains layer
KIS 14-2	Slag (aggregate)	Kisaba	Mindouli	13th-14th century CE - Excavation (SIV -10cm): Metallurgical remains layer
KIS 14-3	Slag (1)	Kisaba	Mindouli	13th-14th century CE - Excavation (SIV -20cm): Metallurgical remains layer
KIS 14-3	Slag (2)	Kisaba	Mindouli	13th-14th century CE - Excavation (SIV -20cm): Metallurgical remains layer
MKU3 14-1	Bar fragment	Makuti	Mindouli	13th-14th century CE - Excavation (SVI -5cm): Metallurgical remains layer
MKU 13-1	Barrette	Makuti	Mindouli	13th-14th century CE - Surface collection
MKU3 14-2	Mineral refuse	Makuti	Mindouli	13th-14th century CE - Excavation (SVI -25cm): Metallurgical remains layer
MKU3 14-3	Tuyere	Makuti	Mindouli	13th-14th century CE - Excavation (SVI -25cm): Metallurgical remains layer
GPSNN 294-1	Barrette	Nkabi	Mindouli	13th-14th century CE - Surface collection
GPSNN 294-2	Ingot	Nkabi	Mindouli	13th-14th century CE - Surface collection
KTG 14-3	Cu prill	Kitchounga	Mindouli	19th century CE - Excavation (SI -30cm): Metallurgical remains layer
KTG 14-1	Cu (+ Slag?)	Kitchounga	Mindouli	19th century CE - Excavation (SI -30cm): Metallurgical remains layer
KTG 14-2	Slag	Kitchounga	Mindouli	19th century CE - Excavation (SI -30cm): Metallurgical remains layer
NTM15-1	Copper extracted from slag	Ntominsie	Mindouli	16th-18th century CE - Excavation (SI): Metallurgical remains layer
NTM15-1	Slag	Ntominsie	Mindouli	16th-18th century CE - Excavation (SI): Metallurgical remains layer
GPSNN-290-1	Lead slag	Mfouati 'Songa-Melka'	Mfouati	19th-20th century CE - Surface collection
RGM583-1	Ingot fragment	Ubangi	Ubangi	19th century CE - Collected by Coquilhat in late 19th c. in Mbandaka region
RGM583-2	Ingot fragment	Ubangi	Ubangi	19th century CE - Collected by Coquilhat in late 19th c. in Mbandaka region

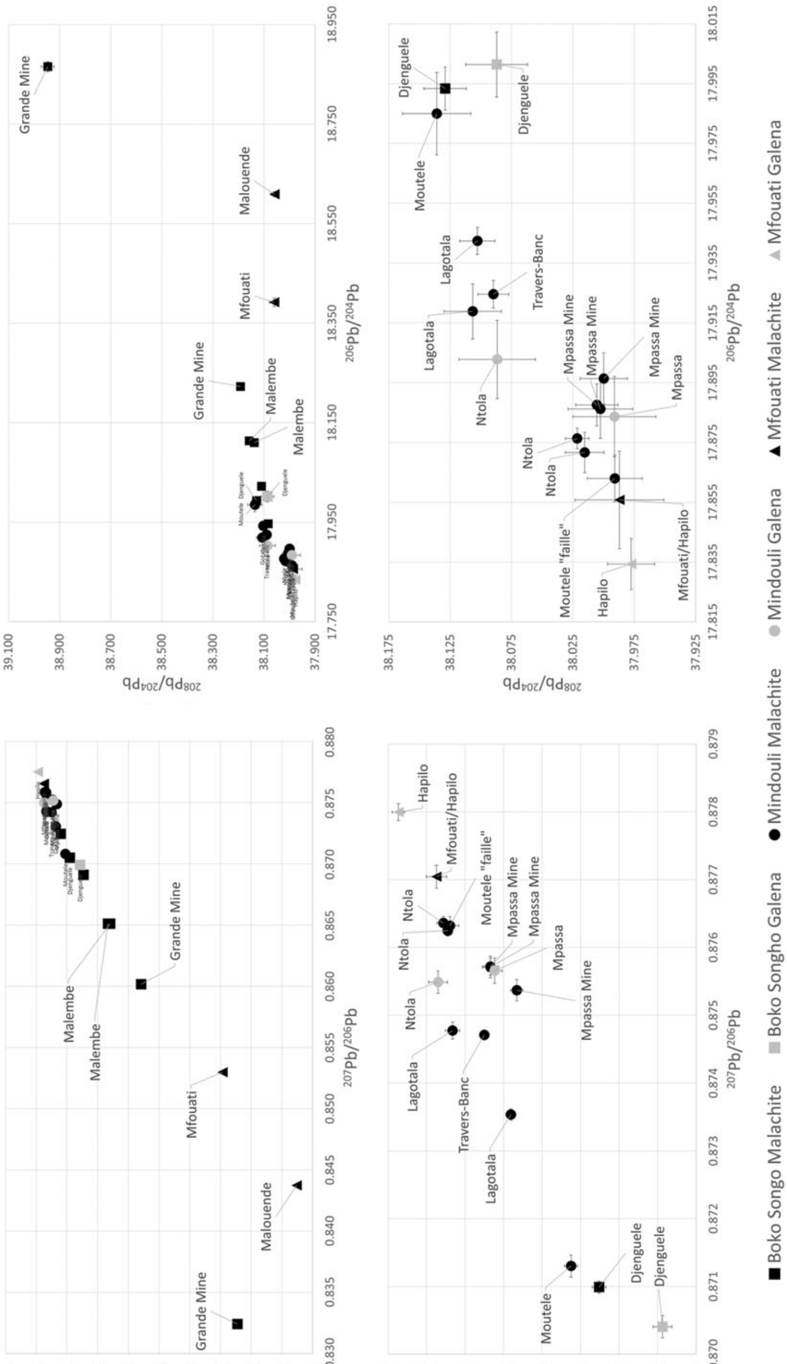

Figure 3 *LI ratios of the Niari Basin geological samples.*

Figure 4 *LI ratios of the Mindouli archaeological materials (top) and the Boko-Songho, Mfouati and Ubangi archaeological materials (bottom).*

data and the calculated T-μ-κ data, which in this case do not offer significant advantages for ore–artefact comparisons (Albarède *et al*. 2012). Therefore, $^{208-207}Pb/^{206}Pb$ plots (most familiar in the archaeological literature) are used throughout this publication, together with $^{208-206}Pb/^{204}Pb$ plots, which capture the largest thorogenic lead variation and offer better resolution in distinguishing the ore sources (e.g., Höppner *et al*. 2005; Baron *et al*. 2014).

Overall, the ore samples from different mining and production areas within the Niari Basin can be broadly distinguished in terms of their LI ratios, with the highest and more dispersed $^{208/207/206}Pb/^{204}Pb$ values occurring at Boko-Songho and Mfouati, in the low-lying eroded valley, while the Mindouli ores cluster more strongly. It is important to note, however, that these distinctions are not that well delineated, with some overlap occurring between samples from these areas. Furthermore, it is likely that more significant overlap exists between the orefields than is revealed by the presented sample. Nonetheless, the general trends still hold, and allow for a broad distinction between the different areas. Additionally, it appears possible to distinguish between ore sources within Mindouli as well—in particular, between the Mpassa Mine, Ntola and the eastern zones. The potential existence of more extensive overlap between these deposits than revealed by the current sample must be considered.

It is important to emphasize that the detailed discussion of Niari Basin ores (see the OSI) focuses on a relatively small range of LI ratios. Many orefields characterized in archaeological provenance studies—for example, the Sinai Desert (Abdel-Motelib *et al*. 2012) and Oman (Weeks 1999; Begemann *et al*. 2010)—range widely in their LI ratios, while the Niari Basin ores are better constrained. With regard to regional provenance studies, it is useful to compare these ore data to the published Central African ore LI data from the DRC and Zambia (plots in the OSI). It must be emphasized that most of these comparative LI data represent primary (sulphide) mineralizations, which might differ from their associated supergene copper ores.

The Niari Basin ores can be well distinguished from the other main copper sources currently known (and characterized) in Central Africa, and thus offer an excellent potential for provenance studies of traded copper objects in the wider region. Only the most radiogenic Boko-Songho and Mfouati ores are similar to some Copperbelt ores (the Damaran–Lufilian Fold Belt), but the majority of Copperbelt ores have more extreme radiogenic LI ratios. Furthermore, many of the Copperbelt ores are from mixed copper–cobalt mineralization (Key *et al*. 2001; Dewaele *et al*. 2006), whereas the Niari Basin ores (and metals; see the 'Results: archaeological materials' section) have very low trace element levels. Therefore, the majority of Copperbelt ores, and copper smelted from them, may have different characteristics. It is, however, difficult to say which Copperbelt ores were used in precolonial times, and ancient copper and its production remains from that region should be studied for better comparisons.

RESULTS: ARCHAEOLOGICAL MATERIALS

Introduction

Details of the 30 archaeological materials analysed in this study, which comprise copper ingots, a bracelet, copper minerals and various production waste (mainly copper and lead slag), are presented in Table 2. Their LI ratios are illustrated in Figure 4. As Mfouati has only been surveyed (no test pit), no contextualized archaeological material is available for the area, but some sites yielded very similar ceramics and crucibles to those of the 15th–17th century production in Boko-Songho.

The selection of these samples aims to capture the variability of copper witnessed in the Niari Basin at the different periods currently identified, and to establish tentative links between the ores (see the 'Results: ores' section), production waste and copper (alloys) from production and consumption contexts. Comparisons are made between archaeological and ore materials to discuss chemical and LI similarity. Chemical similarity is implied by the presence/absence of specific elements, as well as their ratios to each other: elemental fractionation between metal and slag typically distorts absolute concentrations with respect to ore compositions. As noted in the 'Results:

ores' section, this is different for specific elements, due to their varying sensitivity to changing redox conditions and mobility during the smelting process. Certain ratios remain (more or less) constant, while others may change irregularly (e.g., loss of volatile elements: Mödlinger *et al*. 2017). Detailed slag analysis can clarify this further (elemental distributions in slag matrix vs. embedded prills), although chemical heterogeneity is inevitable in (ancient) metallurgical waste (Humphris *et al*. 2009; Rademakers and Rehren 2016). LI consistency is reported when materials fall within the range noted for ores discussed in the 'Results: ores' section. It is emphasized again here that the LI ratios for these outcrops may overlap to an important extent. Furthermore, the LI ratios of slag may sometimes differ significantly from those of the ores used or the resulting metal (Baron *et al*. 2014), as may those from crucible slag and charge (Rademakers *et al*. 2017). For metallurgical operations involving the addition of lead at some stage, which may be the case for some Niari Basin contexts (see the Introduction), this must be carefully evaluated. Finally, the use of small samples for acid digestion and small analysis areas for laser ablation analysis increases the sensitivity to chemical heterogeneities. Therefore, the LI ratios and chemical compositions are discussed in light of the available evidence and the above-mentioned considerations.

Given the current absence of known ore deposits similar to those of the Niari Basin in terms of their LI ratios, the archaeological materials from the Niari Basin are compared to these local ores without reference to other ore deposits (from Africa or further away). Indeed, most of these materials are highly consistent with the various local ores: if we take a regional provenance perspective, most metallurgical samples can be seen to overlap the Niari Basin ores as a small cluster, and it is most likely that early metallurgists used copper from these nearby deposits. This section makes use of ore LI ratios and geochemistry (see the 'Results: ores' section) to distinguish between the different mining areas wherever possible, as a micro-regional provenance study. It must be kept in mind, however, that this subdivision into mining zones located only a few miles apart (Mindouli area) is at the resolution limit of geochemical analysis.

The sections below highlight the most important observations, while a more extended interpretation of the results is provided in the OSI.

Mindouli

The three 13th–14th century CE *barrettes* (known as *mulambula*) from Nkabi, Misenga and Makuti are consistent with the Mindouli copper ores. These three *barrettes* clearly represent different production events, given their variation in chemical and LI compositions. They were probably produced using copper from Ntola and perhaps Lagotala, although their elevated silver contents are difficult to explain.

Two fragments of contemporary *barrettes* from Makuti and Kisaba have different compositions, consistent with Lagotala ores: they are identical within analytical error in terms of their LI ratios, and chemically very similar. This suggests that these two fragments were made from the same ore, possibly as part of a single production batch: with the high analytical resolution currently available, this is as close as we could expect to identify smelting (and casting) events. The two fragments may thus represent two pieces of a single (broken) *mulambula*, excavated at two sites ~1 km apart. However, it is equally possible that they represent the use of highly similar ores and metallurgical techniques at the two production sites.

The composition of production waste from Kisaba and Makuti is mostly consistent with metallurgical processing of Mindouli ores, as detailed in the OSI.

The two contemporary Misenga-type ingots from Nkabi and Misenga isotopically fall on the edge of the Mindouli range, and may correspond to Moutele–Lagotala (Mindouli) as well as

Djenguele (Boko-Songho) ores. For copper production remains with LI ratios falling within this range, it is thus difficult to distinguish the origin, though eastern Mindouli ores were most probably used in this context.

The bracelet from Misenga has a very different LI composition from the Mindouli ingots. It corresponds more closely to the low-lying eroded valley ores, and particularly the Grande Mine (Boko-Songho) ones. Its trace element pattern indeed does not suggest the mixing of Mindouli copper with another (copper or lead) source, although its silver content is notably high. Unfortunately, this bracelet comes from Maurits Bequaert's 1951 excavation and its find context is not sufficiently precise to explain these dissimilarities with other Misenga objects.

Copper prills and slag samples were physically extracted from a 16th–18th century CE Ntominsier slag fragment for analysis. As expected (but nonetheless reassuringly), the LI ratios of copper and slag are identical within analytical error and very similar to those of the 13th–14th century CE Misenga-type ingots. Chemically, however, an important fractionation is observed between the copper and slag matrix, suggesting poorly reducing smelting conditions. This illustrates the possible discrepancy to be expected when comparing (trace) element patterns between ore, slag and copper.

The Kitchounga slag and copper prills were excavated separately, with the latter representing loose debris (probably unretrieved prills formed during smelting, given the high sulphur content and their occurrence amidst crushed slag). The results from Ntominsier and Kitchounga suggest a continuity in the exploitation of Mindouli ores through time—possible interruptions being beyond the resolution of this study. Increased lead contents in later periods may be related to the addition of Ntola-consistent lead or the use of a lead-rich copper ore from this mining area in the smelting process.

Boko-Songho and Mfouati

The Kindangakanzi copper production waste is isotopically consistent with the Djenguele copper ores. The slag composition suggests that either more lead-rich ore was used in the smelting process or that lead was added during the smelting process (in the latter case, lead smelted from Djenguele galena may have been used). Detailed slag analysis may shed further light on this issue.

Both the Mouadabambi smelting slag and the small Mouadabambi bar ingot have LI ratios that are identical (within analytical error) to those of Mfouati/Hapilo copper ore, but the ingot contains ~25% lead, while the slag contains ~30% lead. This suggests that either a lead-rich Mfouati copper ore was used for the smelting of copper used in this ingot, or that Hapilo lead was added to copper during the ingot production process.

The lead (and copper) smelting slag bear testimony to the production of lead at Songa-Melka (near Mfouati) during the 19th and early 20th centuries CE. The LI ratios of this lead slag are fully consistent with Hapilo galena, and identical to the LI ratios of smelting slags from Mouadabambi. This supports the suggestion that Hapilo lead was used to produce lead-rich copper ingots, although further copper slag microanalysis is needed to test this hypothesis.

Interestingly, the two analysed ingot fragments from the Ubangi region, ~800 km north-east of the Niari Basin, were found to have LI ratios very similar to those of the Mouadabambi ingot fragment and similarly high lead contents (~15%). This strongly suggests that these ingots were made following a similar production process, in which the same Hapilo lead was employed.

The addition of lead during production processes obscures the analysis of copper: LI ratios are reflective of the added lead, as this contribution usually well exceeds the lead content present in copper ores from the Niari Basin. However, the high arsenic content of the Mouadabambi bar ingot (~1.5%) may point to the use of a particular copper ore. Although trace elements could theoretically be used to discriminate different copper batches produced in this lead-adding smelting procedure, the differences within the Niari Basin may be too weak to offer clear distinctions (see the 'Results: ores' section).

As for the Mindouli ingots, these high-resolution data again offer the closest analytical view we may expect in order to achieve understanding of this *chaîne opératoire*, where it is possible to trace a piece of galena as it was smelted to obtain lead at Mfouati, then travel to other sites in the Niari Basin to produce lead-rich copper ingots, which subsequently spread both within the region (e.g., Boko-Songho) and far beyond it (e.g., Ubangi).

DISCUSSION

This presentation of the Niari Basin copper production industry sets a reference for future provenance studies within the region (e.g., DRC *croisette* ingots: Rademakers *et al.,* in prep.). It aims to establish the potential for discriminating these production regions on the basis of geochemical analysis and outlining the extent to which the produced ingots were traded, to better understand ancient socio-economic networks in western Central Africa. This paper shows that the majority of Copperbelt ores, and by extension copper smelted from these ores, should be clearly distinguishable from the Niari Basin in terms of their LI ratios.

For Igbo Ukwu and Ife, Willet and Sayre (2006) conclude that some of the early West African castings may have been produced using local copper ores, although many made use of copper from northern Africa obtained through trans-Saharan trade, and of copper imported from Europe. However, arguments from chemical composition and chronology suggest that the Igbo Ukwu copper was local, reflecting ores hitherto not characterized (Goucher *et al*. 1976; Killick 2016). A comparison of the Niari Basin copper ores with North and West African copper ores discussed by Willet and Sayre (2006) shows that these differ significantly in their lead isotopes, and should be easily distinguishable in provenance studies (see the OSI).

Analysis of several bronzes from South Africa revealed relatively low lead contents: Molofsky *et al*. (2014) convincingly showed their highly radiogenic LI composition to be dominated by the lead isotopic signature of the bronze's tin (smelted from Rooiberg cassiterite), rather than that of the copper alloyed with it. As such, these bronzes offer no insight into the LI composition of copper used in that region: an important issue that may affect future provenance studies of African bronze and copper artefacts. The three Rooiberg copper (ore) samples analysed by Molofsky and co-authors are the only known published LI data for Southern African copper. As illustrated in the OSI, these may be easily distinguished from the Niari Basin ores.

The results obtained for the Niari archaeological material have all been discussed with reference to local ores. This appears to be appropriate given the high proximity of these ores to the various production sites, but it must be stressed that geochemical data should be used to discriminate between possible sources, and to reject sources that do not match archaeological remains, rather than to provide 'positive answers' (of which the 'Results: archaeological materials' section appears to be guilty). The main reason for presenting such positive identifications above, however, is the lack of alternative comparisons, leaving such interpretations as the only available option. Even so, it is hard to imagine that a better ore–slag–copper match could be found and that ores from outside Central Africa would be more likely candidates for the production of copper in the Niari Basin.

However, ore deposits within comparable geological formations (along parallel fault systems) stretch discontinuously over the south-east (Renéville) and south-west (Nyanga, Niari loop area) Republic of Congo, the Kongo Central province (DRC) and northern Angola, and may present the only relevant geochemical overlap. The DRC ore is fairly similar to the Niari one (although with more vanadium) but is, however, located at depth, with few traces visible at ground level (Noël 1983). As a result, there is no evidence of indigenous copper exploitation in this area. In contrast, active mining has been attested until the mid-19th century in the Bembe region in Angola (Bisson 2000), and ancient workings were reported near minor outcrops such as Renéville (Dupré and Pinçon 1997) and in the Nyanga area (Nicolini 1959), but never studied. A detailed characterization of this production area would be helpful in further refining provenance studies for the region.

On the 'local' scale, the similarity of early (13th–14th century CE) Mindouli copper in terms of chemical purity must be noted: all trace elements are below 10–30 $\mu g\ g^{-1}$, with the exception of iron (mostly $< 50\ \mu g\ g^{-1}$, two $< 0.05\%$), arsenic ($< 0.1\%$), silver (0.2–0.7%) and lead ($< 0.1\%$). Such a pattern of low iron, nickel, cobalt and antimony and higher arsenic and silver relative to levels seen in the ore samples (ratios to copper) is remarkable. Low iron contents are generally considered an indication of poorly reducing smelting conditions and/or the use of very pure ores resulting in an almost slag-free process whereby little iron is reduced with the copper (Craddock and Meeks 1987). The archaeological data further suggest a careful crushing and concentration of ore at production sites (Nikis and De Putter 2015), which could have diminished the iron in the smelting system. Poorly reducing smelting conditions, however, could lead to significant loss of arsenic, which appears unlikely as the copper is arsenic-rich with respect to the Niari Basin ores. Thus, the ores used at that time may have been more arsenic- and silver-rich than those found in outcrops today. However, another possible hypothesis is the use of native copper in these early production contexts. Although native copper is very difficult to recognize analytically, low nickel–cobalt–antimony and increased arsenic–silver contents are some of its few tentative indicators (Pernicka 1999). Native copper has not been encountered during recent field surveys, but may have been present at the top of the oxidized ore sequence and has been attested in the study area (Koud 1987). Regardless of the origin of this silver and arsenic, they may serve as important chemical markers for Niari Basin copper, in conjunction with their characteristic LI ratios.

Another important factor to bear in mind is the possible recycling, reuse and mixing of metals in the Niari Basin at any time. This could lead to a mixed chemical and LI ratio signature of the resulting metal. In the case of extensive mixing, a homogenized 'average' composition (chemical and LI ratios) would be expected to emerge over time. However, the notable smelting evidence at these different sites suggests regular input of fresh metal from different regions in the Niari Basin. While extensive mixing is not apparent from the presented data, mixing between the most different sources in the Mindouli area, for example, would result in a 'typical Mindouli signature', making it very hard to detect. As such, mixing, both at the level of primary production (different ores in a single smelt) and secondary production (different metals in a crucible), may go undetected. While the homogeneity in terms of chemical composition (e.g., silver) in early Mindouli copper is remarkable, it may suggest the use of a particular ore type rather than important mixing practices, as noted above.

Whatever the sources of copper were, a clear change in production technology is evident in the later Boko-Songho, Mindouli and Ubangi material: lead is introduced in the production process certainly at Kitchounga and Mouadabambi (but not Ntominsier), as described by historical sources (Pleigneur 1888; Dupont 1889; Barrat 1895: 234; Reibell 1903: 234). At Kindangakanzi, it is less clear if the lead addition is intentional or from lead-rich copper ores. This lead dominates the LI ratios of the resulting alloys, impeding copper provenancing. However, lower silver and sometimes increased iron, arsenic and antimony contents are noted. This may indicate a shift in exploited ore types and/or deposits in conjunction with smelting technology.

Interestingly, no evidence for the exploitation of Mpassa Mine ores could be noted in the LI data, which matches the absence of mining evidence in the field. This is in line with the wider trend of deeper-lying ores not being unnecessarily exploited elsewhere in Africa where an abundance of surface deposits is available (Miller and van der Merwe 1994).

In addition to the chemical and isotopic markers increasingly available to distinguish between African copper sources, typological differences between ingots from the Niari Basin such as the *mulambula* and Misenga-type ingots or the 19th century ingots, various bar and cross-shaped (*croisette*) ingots from the Copperbelt region (de Maret 1995; Swan 2007) or the characteristic *musuku* (Stayt 1931) and *lerale* in Southern Africa (e.g., van der Merwe and Scully 1971) can further help us to understand copper in its trade patterns or currency format (homomorphic South African tin *lerale* ingots are known as well: Killick 1991). However, while these distinctions offer a much quicker and cheaper guide towards distinguishing the provenance of such ingots, the validity of their provenance often remains to be tested. For example, HXR-type *croisettes* have been produced both in the southern Copperbelt, in the region of Lubumbashi, and in northern Zimbabwe between the 15th and 17th centuries. In regions more remote from the aforementioned production areas, the provenance of copper (ingots) may yield surprising results. The possible replication of widely traded ingot shapes using local copper sources could help reveal the myriad of sociocultural interpretations of these ingots. For example, during the 19th century, *handa croisettes* were recast from copper objects in areas hundreds of kilometres away from Copperbelt (Sundström 1974, 221). In our sample, variability in ingot shapes, particularly from the Mindouli area, has shown no strong correlation with find contexts. It could be noted that the *barrette*-type ingots appear to have been made from Ntola and/or Lagotala ores, while the sampled Misenga-type ingots conform to other deposits. However, this pattern is difficult to interpret due to possible sample bias, geochemical overlap and temporal changes across the different production areas, which are not yet fully understood. Furthermore, the question whether (and where) these ingots were transformed into other objects still needs to be explored. For example, it appears that the *barrette*-type ingots are hammered versions of the Misenga-type ingots (Nikis, in press b), while the analysed Misenga bracelet (no other copper 'consumption objects' from the area were available for study) does not match any of the sampled ingot types, but appears to be made using 'non-local' Boko-Songho ore.

Nonetheless, the discussion of the new data presented in this paper shows how it may strengthen the developing understanding of copper production in the Niari Basin and beyond. The example of Niari Basin ingots travelling over 800 km, probably along the Congo River, and reaching the Ubangi area is certainly the tip of the iceberg: the copper trade routes and the uses of this sought-after metal in Central Africa form a research field in need of more attention than it has hitherto received.

CONCLUSION

This paper has presented new analytical data on early copper production in the Niari Basin, Republic of Congo. It is the first time in Central Africa that both geological samples from various mines and outcrops in the region and archaeological materials from controlled excavation and survey have been characterized. These analyses reveal that the majority of Niari Basin ores are well constrained in terms of their geochemical characteristics, and that all copper produced in the region is compatible with these local ore sources. While micro-provenance may be examined within the production zone in very high detail, some variability is beyond currently available analytical resolution. On this local scale, this paper has documented homogeneity in copper production during the 13th–14th centuries CE, resulting in very pure copper with a remarkable silver content. A clear break in smelting technology is noted with respect to the 19th century CE, where the addition of lead to the smelting process changes both the chemical and LI compositions. On a wider scale, these results will be useful as a first reference for provenance studies in the wider Central African region. In particular, the future study of areas without direct access to such rich copper deposits, where copper was often imported over long distances, may determine whether the copper came from Copperbelt, Niari or other deposits, and thus reveal the distances over which Niari Basin ingots were exchanged. Geochemical and isotopic data on ore–artefact–slag material from reliable archaeological contexts, as presented in this work, combined with ethnographic and historical written evidence, offer a powerful methodology to gain a more integrated understanding of ancient metal production and exchange in sub-Saharan Africa.

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