Laos' central role in Southeast Asian copper exchange networks: A multi-method study of bronzes from the Vilabouly Complex

Mélissa Cadet, Thongsa Sayavongkhamdy, Viengkeo Souksavatdy, Thonglith Luangkhoth, Philippe Dillmann, Christophe Cloquet, Justine Vernet, Paolo Piccardo, Nigel Chang, John Edgar, et al.

Lao's central role in Southeast Asian copper exchange networks: a multi-method study of bronzes from the Vilabouly Complex

Mélissa Cadet[1,4], Thongsa Sayavongkhamdy[3], Viengkeo Souksavatdy[3], Thonglith Luangkhoth[3], Philippe Dillmann[4], Christophe Cloquet[5], Justine Vernet[6], Paolo Piccardo[6], Nigel Chang[7] John Edgar[7], Eddy Foy[4], T. O. Pryce[1,4]

[1] Centre National de la Recherche Scientifique, Préhistoire et Technologie, UMR 7055, MAE, 92000 Nanterre Cedex, France

[3] Department of Museums, Historic Buildings and Archaeology, Ministry of information and Culture, Vientiane, Lao Democratic People's Republic

[4] LAPA : IRAMAT, UMR 5060, NIMBE, CEA/CNRS, CEA Saclay, 91190 Gif sur Yvette cedex, France

[5] SARM, CRPG, CNRS, 54500 Vandoeuvre les Nancy cedex, France

[6] Chemical and Industrial Department, Genova University, 16146 Genova, Italy

[7] College of Arts, Society & Education, James Cook University, Townsville QLD 4811, Australia

Abstract

The application of lead isotope-based provenance analysis in Southeast Asia over the last decade has strongly suggested a central role was played by the Lao PDR in regional copper production exchange networks for approximately 1500 years. The Vilabouly Complex, in central Lao's Savannakhet Province, has revealed major copper mining and smelting sites dated to the regional Iron Age (c. 400 BC – AD 500) and possibly Bronze Age (c. 1000 - 400 BC). Protohistoric metallurgical practices at the Vilabouly Complex, and indeed for all of the Lao PDR, are unknown, and the propose of this paper is to provide a comprehensive analytical study of the Vilabouly Complex metal assemblage, including 60 copper-base artefacts of multiple typologies. Cut samples of these were subjected to morpho-stylistic, metallographic (OM), elemental (XRF, SEM-EDS) and lead isotope analyses (MC-ICP-MS) in order to reconstruct the range of forms, metalworking materials, techniques (alloying, casting and post-casting treatments) used at the Vilabouly Complex. The results revealed an assemblage composed of copper, bronze and leaded bronze alloys, with a majority consistent with the lead isotopic signature for the Vilabouly Complex copper. The consistent geochemical and technological signature of the majority of artefacts strongly corroborates the extensive onsite production evidence, and fits with the burgeoning regional copper-base metals database for copper metal demand being sated in large part by Lao PDR supply.

Keywords: Lao PDR; Southeast Asia; copper-base metal; Bronze Age; Iron Age; Microstructure; Elemental and lead isotopic composition

1. Introduction

For many decades, archaeometallurgical research and debate in Mainland Southeast Asia (hereafter "MSEA") (Figure 1) was heavily focused on the origin/s of regional Bronze Age metallurgy: its timing, independence versus imported versus stimulated nature, and the mechanisms by which those technological transfers may have occurred – typically from present day China (White 1988, Higham 1996, Pigott & Ciarla 2007, Pryce et al. 2010, White & Hamilton 2009). It is now near universally understood that early mainland copper-base metallurgical traditions do not represent an independent invention (cf. Solheim 1968) but derive from North-South interaction networks. Two *ex Sina metallum* hypotheses have been proposed to explain this phenomenon, both sharing the prevalent Chinese metallurgical origins model of a Steppic "Seima-Turbino" derivation via the Gansu Corridor at the turn of the 3rd/2nd millennium BC (e.g. Mei Jianjun 2000) but dividing thereafter. The first, and most widely accepted, proposition is that MSEA copper-base metallurgy attested from late 2nd millennium BC regional contexts was diffused from the highly sophisticated bronze-using cultures of the mid-2nd millennium BC Central Plains via the border regions of Lingnan by down-the-line 'trade and exchange' relations (Ciarla, 2007; Higham, 1996; Higham et al., 2011; Pigott and Ciarla, 2007; Rispoli et al., 2013). In this model, the metallurgical tradition/s transmitted to MSEA lacked the technical and decorative elaboration of elite Chinese metal assemblages due to 'founder effect' but rather reflected the simpler metal traditions to be found in non-elite Chinese contexts. The second model (White and Hamilton, 2018a, 2018b, 2014, 2009) argued that MSEA metallurgy represented a direct transmission of Seima-Turbino metallurgy, by Seima-Turbino individuals and/or those trained by them, from the Gansu Corridor to northern Thailand at the aforementioned turn of the 3rd/2nd millennium BC.

Much interesting work remains to be done on the issue of MSEA metallurgical origins, namely as the border with present-day China is approximately 2500 km long and encompasses a range of environments and a huge diversity of ancient cultures, many of which remain poorly understood. There has, however, been very substantial convergence on the dating of the regional Bronze Age transition to the late 2nd to early 1st mill. BC, with some localised and likely historically significant variation (e.g. Higham & Higham 2009, Pryce et al 2018b, Rispoli et al., 2013). With this horizon if not fixed then certainly secure, we can turn our sights to the proceeding up to 1500 years of prehistoric metallurgical behavior, which may hold important clues as to Mainland Southeast Asia's diverse paths to early state formation.

To further tune our focus, there is now a considerable body of archaeometallurgical data for regional copper-base exchange systems, the majority of which pertain to the Iron Age (c. 400 BC to c. AD 500, e.g. Higham and Rispoli, 2014, Hirao et al. 2013, Pryce et al. 2011, 2014, 2018a). These data strongly suggest that copper production at the Vilabouly Complex (or "VC", previously known as Sepon or Xepon) in Savannakhet Province of central Laos (Figure 2) was of considerable importance to regional metal supply. Indeed, lead isotope signature consistency for MSEA copper-base metal consumption across MSEA could indicate that the VC, and/or a geologically-similar source, join the long known major prehistoric Thai copper production loci at Phu Lon (Natapintu 1988, Pigott & Weisgerber 1998) and the Khao Wong Prachan Valley (Pigott et al. 1997, Pryce et al. 2010, 2011). This supposition must, however, be tempered with the still partial nature of regional metals sampling and as yet unresolved recycling processes, i.e. there are strong hints that central Thai copper supplied parts of ancient Myanmar rather than much of eastern MSEA as previously assumed (Pryce et al. 2018b, Dussubieux & Pryce 2016). Nevertheless, the apparent predominance of prehistoric Lao PDR copper production is all the more remarkable given the relative dearth of protohistoric archaeological sites in the, admittedly highly mountainous and difficult-to-access, national territory. Indeed, despite the VC having been known as a major Iron Age copper producer since the mid-2000s, no prehistoric Lao copper/bronze artefacts (other than a single VC copper ingot, see Pryce et al.2011) have ever been subjected to full laboratory analysis. The entire VC copper production and consumption assemblage is currently the subject of a detailed technological and geochemical research programme for the lead author's doctorate. This production study will be published separately due to the space required but with regards to artefacts related to copper/bronze consumption, we pose the following questions: what sorts of artefacts were used at the VC, made with what alloys and with what techniques, and their relations to regional comparanda? What is the correspondence between heavy VC copper production and local copper/bronze consumption? Is all the copper produced locally, and, if not, what artefacts are imported and why? Retracing the Lao PDR's ancient metallurgical traditions, could also be of key interest in view of the copper metallurgy origin debate, sharing a ca. 400 km border with present day China to the North, and also with Vietnam, Thailand and Cambodia, with the Mekong River providing a significant means of long-distance mobility. With 60 VC metal samples, this present paper sets out to answer some of these questions.

1.1 Geology

The Vilabouly Complex is situated in the Sepon Mineral District (SMD) of gold and copper deposits, under licence to Lane Xang Minerals Limited “LXML”, along the southern boundary of the NW-trending Truong Son fold belt in Savannakhet Province, south central Laos (Figure 1). The Sepon Basin is a small scale clastic-carbonate sedimentary basin approximately 20 km long by 8 km wide, belonging to a group of Palaeozoic successor basins. The prospect is located at an altitude of approximately 300 meters above sea level, with the central section of the district located longitude 105°59’E and latitude 16°58’N. Sepon Mineral District stratigraphy comprises Devonian to Carboniferous aged continental fluvial and shallow to deep marine sediments (Cromie, 2010; Manini et al, n.d.; Sillitoe, 1998). At least three mineralisation styles are recognised: sedimentary rock-hosted Au; Cu-Au skarn, and quartz stockwork porphyry Cu-Mo. Three types of intrusions occur: rhyodacite-porphyry, stocks and dykes.

The two main archaeological sites of the Vilabouly Complex are situated in the two main copper exploitation areas of the district: Khanong and Thengkham, even if it has been proved that the main paragenetic assemblage of the district occurs at both sites. For Khanong, the zone is at least 1.5 km long and 0.5 km wide and consists of a supergene-enriched leached blanket of chalcocite clay with associated malachite, azurite, cuprite, native copper and proximal exotic copper wad. At least three main groupings of copper mineralization types are reported to occur in the Khanong copper deposit area: supergene, exotic and hypogene. The geological setting for Thengkham (which includes the sites; Peun Baolo/Thengkham South C, Thengkham South D, Thengkham East & Tham Hin Kieow) is less well understood but copper occurs in massive sulphide deposits up to 100 m thick and containing between 0.5 to 1 wt.% Cu and is inferred to be a proto-ore type for the development of supergene enriched copper mineralisation (Cromie, 2010; Manini et al., n.d.; Sillitoe, 1998).

1.2 Archaeology

1.2.1 Vilabouly Complex

Rescue archaeology at LXML started in 2008 under the co-direction of Thongsa Sayavongkhamdy, Viengkeo Souksavatdy and Nigel Chang as a collaboration between the Lao Department of National Heritage, James Cook University and the mining concession. Archaeological finds in the Vilabouly Complex (Figure 2) show a long occupation period, thought to be from the Neolithic (c. 3000 BP), with a concentration of Iron Age copper mining sites with associated occupation and funerary activity, as well as isolated finds from the Lane Xang Kingdom (AD 1354-1707) and remains from the mid-20th century Indo-China wars (Tucci et al., 2014). Direct dates from ancient mining shaft structural elements suggest copper mining between about 1000 BC and AD 700 (Table 1).

1.2.2 Sepon mine

1.2.2.1 Thong Na Nguak

Thong Na Nguak or Dragon Field (“TNN”) was excavated for one season in 2008 with the aim of confirming that in-situ archaeological evidence was indeed present at the site. With this confirmed, all activity on the site ceased due to its spiritual importance for local communities. A single radiocarbon date places it in the early Iron Age (Table 1, 346-54 BC at 95% confidence from charcoal in a burial jar, WK-32284). The burial jar was associated with three others, within one of many rectangular stone arrangements, along with other finds indicating high-temperature ore processing with slags, crucibles, ores and metal artefacts.

1.2.2.2 Khanong A2

Khanong A2 is an extraction site that seems to be linked with Thong Na Nguak, where the latter was the processing area for the extracted minerals. Radiocarbon dates show an early-mid Iron Age activity period (Table 1).The site was first excavated as a 'rapid response' rescue project in 2009, with over 200 separate mining shafts identified, some of which containing intact wood and bamboo matting reinforcements (Tucci et al. 2014). A further series of excavations were completed when the mining pit was extended in 2015. The unique vertical mining shafts, some extending more than 30 m below the current surface, were first observed here.

1.2.2.3 Thengkham South D and Thengkham East

Thengkham South D, as with Khanong A2, was discovered during modern mining operations and was investigated as a 'rapid response' rescue archaeology project in 2012, with a further season in cooperation with modern mining in 2013. This revealed a further extensive field of over 200 vertical mining shafts with the deepest extending at least 40m from the surface. Thengkham East is another ancient extraction site that has been exposed by modern mining operations. Samples from exposed wood and bamboo elements of mining shafts have been collected, but further archaeological investigation has been delayed.

1.2.2.4 Puen Baolo and Thengkham South C

Puen Baolo or Crucible Terrace ("PBL") was the first site to be excavated, alongside Dragon Field. It has been the subject of major annual excavation seasons from 2008 to 2015 (Figure 3), with modern mining activity redirected to other parts of the mining tenement during these years (leading to the discovery of the other sites already mentioned). The nine excavation seasons exposed 1120 m², showing mining and production activities with shafts, slags, crucibles, ores and metal artefacts. Commercial mining activity restarted here in 2016 leading to the discovery of an adjacent, extensive, field of ancient mining shafts reflecting similar technology as seen at Khanong A2, Thengkham South D and Thengkham East. Now known as Thengkham South C, this new area was investigated over four short rescue archaeology projects in 2016. PBL appears to be the occupation and processing site, located on a small terrace, associated with an extensive field of vertical mining shafts cut into the steep hillside above (Thengkham South C).

On the PBL portion of the site we see a natural substrate of clay and soft siltstone, overlain by a layer of occupation and ore processing debris. Varying concentrations of metallurgical production debris (slag, technical ceramic and mineral) were found, some of which were quite dense (e.g. G15, see Figure 3) but no definite high temperature installation was identified in situ. To the northeast, deep deposits of at least three meters of ash and other sweep-out, presumably from processing activity, were identified in the final season of excavations as the site was being prepared for commercial mining. Burials and other features cut into the natural substrate are thought to date to the Bronze Age (regionally, c. 1000 BC to c. 400 BC) based on associated artefacts. The bulk of the complete bronze/copper alloy artefacts are associated with these deeper features – in particular bow-tie ingots – along with unique chalcedony beads, pottery & no iron; it should be noted that the acidic soils have dissolved any original bone in the burial features. A second type of burial or deliberate artefact cache has been identified. These lie within the upper layers of deposit and include iron axes, conical copper ingots and pottery. Surprisingly, no glass has been found at this site. One radiocarbon date from the upper edge of a pit or mining shaft places this later Puen Baolo activity to at least the regional early Iron Age, 2204 ± 20 BP (361-202 BC).

Crucially, the opening of the Thengkham South C pit allowed dating of the PBL associated mining shafts. As with all the mine shaft dates, we have relied on samples of structural rattan or bamboo that have been preserved in permanently wet clays 10 meters or more below the original ground surface, with the earliest determination of approximately 1000 BC (1071-922 calBC 95.4%; WK43470; 2843 +/- 20 BP). Other shaft dates confirm that mining activity continued at PBL/TKS-C to as late as 1309 +/- 20 BP (WK43467; 660-767 calAD 95.4%). As with the other sites noted above other ancient organic materials were also recovered including wooden ladders, parts of what appear to be pulleys, wooden mallets and baskets.

1.2.2.5 Malachite cave

Malachite Cave (Tham Hin Kieow), excavated in 2011 just 10 metres below the top of Thengkham Ridge, abovenear the Puen Baolo site, appears to be a mining area represent quite different mining activity. The archaeological team suspects a complex of tunnels may be contemporary to Puen Baolo activity extending into the hillside, perhaps similar to those now exposed at Phu Lon, but further investigation is necessary. A single radiocarbon date from a hearth within the mouth of the current cave/tunnel mouth suggests activity at 456 +/- 27 BP (WK 33831), however, it is not clear if this has a direct relationship to ancient mining at the site.

2. Assemblage

The entire metallurgical assemblage held by LXML, excavated and chance finds recovered by unexploded ordnance “UXO” teams, was assessed during a visit by the lead author and Pryce, in association with Chang, Souksavatdy and Luangkhot in November 2017. The total mass of slag stored from almost a decade of investigation was c. 145 kg, which is surprisingly little, compared to regional copper production sites like the Khao Wong Prachan Valley (Pigott et al., 1997), where individual test pits could contain several tonnes of slag. Being a manageable quantity, the entire assemblage was physically laid out by site (PBL for the great majority) to spatially represent the excavation squares, and in order of depth to represent chronology. Using this, a true hands-on ‘stratigraphic and sequential’ approach, 138 slag samples, 43 crucibles fragments, 14 ore samples, 8 scorched clays and 60 samples of copper-base artefacts were selected for analysis in order to reconstruct the production process. This article focuses on the metal artefacts. 45, come from PBL contexts, two artefacts from TNN and 13 chance finds from the Vilabouly Museum. The selection of samples was carefully made to ensure the representation of all VC artefact types - three types of ingots (bowtie, conical and bowl) and different kinds of copper-base objects: utilitarian objects (axes, adzes, knives), weapons (spearheads, *Ge*) and ornamental objects (drums, bells) (Table 2; Figure 4.a and b).

The predominant metal artefact type, with 20 examples, is the bowtie ingot, of 110 to 150 mm length, 60 to 80 mm width, and masses of 100 to 240 g (for complete examples, see Figure 4.a). The second most prevalent form, six conical ingots, have diameters of 40-50 mm and masses of up to c. 30g (Figure 4.a). The third ingot type, bowl-shaped, were stacked together and were chance finds. Aside from the ingots, the second major type is axes (10 samples, see Figure 4.a), of different shapes and dimensions, varying from 60-80 mm on their longest axis, and with masses of 20-80 g. One of the two excavated *ge*, ceremonial dagger-axes or halberds of distinctly Chinese-Han typology (Lorge, 2012) was sampled, as were three examples of similar form but without context. Differences can be observed between the excavated and chance find *ge*: the latter being simpler in form, with fewer fine details (Figure 4.a). *Ge* are occasionally found present in northern Vietnamese contexts showing links with China to the North but note that no other example of *ge* are recorded in Laos, also in Thailand. Finally, the presence of two chance finds of bronze drums of Dongson type (Figure 4.b), mainly known from northern Vietnam (Calo, 2014), may denote a link with southern China or northern Vietnam where drums and *ge* are identified.

3. Methodology

All the studied artefacts were cut on site in the Lao PDR using an ultra-fine jeweler's saw, taking into account areas of existing damage (preferred), corrosion (avoided) and presumed use (tools were cut on the working edge). The samples are in the order of millimeter size, cut from exterior surfaces, thus they might not be representative of the object heterogeneity in terms of poor alloy mixing, segregation during cooling, or any unidentified joints or welds. The cut samples were then halved, for elemental/microstructural and isotopic analyses, respectively.

Cut samples were mounted in epoxy resin and ground using silicon carbide wet-dry paper (80-4000 grit), before final polishing with diamond pastes (3, 1 and 0.25 µm). The microstructure was then studied by optical microscopy before (for the corroded samples) and after etching with a ferric chloride solution to reveal the crystalline structures.

X-ray fluorescence was used for the OM samples' global elemental composition of major, minor and (some) trace elements conducted at the Laboratoire Archéomatériaux et Prévision de l'Altération (LAPA-IRAMAT/CEA) in Saclay, France. XRF data were obtained using a NITON XL 3t GOLDD+ portable X Ray Fluorescence analyser in 'laboratory mode' (fixed stand) with a max 40 kV accelerating voltage and by using the 'alloys' mode. Certified Reference Materials (CRMs) were used to check accuracy and precision, with good results for majors and minor components (Table 3). Eleven different CRMs were used: B10, B12, B31, UZ 52-3, B21 and L 20-1 from the *Centre de Development des Industries de Mise en Forme des Matériaux*, France, 71.32-4 and 51.13-4 from the *Bureau of Analyzed Samples Ltd*, England and 500, C1123 and 1275 from *National Institute for Standards and Technology* (NIST). Note that light elements like phosphorous, silicon, aluminium, magnesium and also sulphur at low content were not reliably detected due to non-vacuum conditions. The analyses were performed on the mounted and polished sections, as per OM, using a 3 mm beam diameter, which allowed for reliable results as long as the sample was larger than this. Three such spot analyses were made for each sample to account for heterogeneity: corrosion and inclusions.

µRaman spectroscopy was also used for the identification of non-metallic phases present in one sample, PBL/11, with an Invia Reflex spectrometer at room temperature. Spectra were acquired with an excitation wavelength of 532 nm, a laser power of 0.5 mW and a laser beam diameter of 1µm with the use of Wire 3.4 software. The acquisition time was of 10-20 s.

Lead isotope analysis was conducted at the *Service d'Analyse des Roches et des Minéraux* of the Centre for Petrographic and Geochemical Research (SARM-CRPG) in Nancy, France, using a Multi Collector – Inductively Coupled Plasma – Mass Spectrometre (MC-ICP-MS) after lead extraction (Manhes et al., 1980) and using Thallium NIST SRM 997 to correct for instrumental mass bias (Thirlwall, 2002). All parameters were adjusted to obtain the closest values relative to NIST SRM 981 as determined by DSTIMS (Thirlwall, 2002). More details about the instrumentation and its performance for lead isotope analysis are available in (Cloquet et al., 2006; Pienitz et al., 2015).

The OM/XRF samples were then carbon coated for Energy Dispersive Spectrometry (EDS) coupled with a Scanning Electron Microscope (SEM, JEOL 7001F), in order to establish the global composition of samples too small for pXRF (<3 mm), those with intergranular corrosion, and also to study any inclusions. The SEM-EDS was operated in both secondary electron (SE) and backscattered electron (BSE) modes, using a 20 kV accelerating voltage, a 10 mm working distance with an Oxford Silicon Drift Detector, and processed using Oxford Instruments Aztec software. Detection limit was fixed at 0.5 wt% with a count rate of 4000/s (detection time of 40 s) used for a good resolution of pertinent peaks with respect to background noise. We consider that the relative quantification error (2 σ) is about 10% of the measured value. SEM-EDS accuracy was evaluated using the same mounted and polished CRMs as used for the pXRF analysis, and we obtained good results for the major elements (Table 3). Global composition for each sample was obtained by a mean of 3 to 4 areas scan (0.4 mm) per sample.

4. Results

4.1. Ingots

The ingots exhibit microstructures, in most cases, with non-dendritic equi-axed grains and no twinning, associated with as-cast objects (Figure 6.a). Dendrites can be observed for some of the ingots, along with equi-axed grains (for metallographic results see Table 4). They are all composed of predominantly copper (Table 5). Six samples exhibited iron content up to 3 wt% (by SEM-EDS analyses, avoiding corrosion products; PBL/11, PBL/13, PBL/14, PBL/19, PBL/20, PBL/21). One sample, PBL/11, is different from the others. Indeed this sample was remarkably different in appearance and mass as well (Table 2, Figure 4.a). Moreover, embedded in a metallic matrix made of copper with 2.5 wt% of iron, goethite (α-FeOOH), an iron oxyhydroxide, was identified by Raman spectroscopy with characteristic spikes at 299, 387 and 685 cm^{-1} (Figure 5). The spheroidal form, very comparable to that identified by Cooke and Aschenbrenner (Cooke and Aschenbrenner, 1975), strongly suggests that this goethite is the corrosion product of an α-Fe phase, formed after the cooling of an alloy containing elevated quantities of Fe.

Small sulphide inclusions are present in all the ingots at the grain boundaries but are more abundant for the bowl and conical ingots, which display massive sulphide inclusions and even a Cu-Cu_2S eutectic for TNN/4 (Figure 6.b). Conical and bowl ingots have a S content around 1 wt% whereas the bowtie ingots have less than 1 wt%, or below SEM-EDS detection limit. A different eutectic can be observed for one other sample: Cu-Cu_2O for PBL/10, a bowtie ingot (Figure 6.c). Impurities are slightly soluble and may form eutectics with a low melting point, they are the last to solidify and are situated in interdendritic space. The eutectic Cu-Cu_2O makes the copper more brittle and decreases the workability. Oxygen is quite soluble in liquid copper and can cause the precipitation of Cu-Cu_2O (Chen et al., 2009; Hauptmann et al., 2015; Vander Voort, 2004).

Elemental data (Table 5) for the conical ingots (PBL/42, PBL/48, PBL/49, PBL/50, PBL/51 and PBL/52), except the TNN example indicate a different composition of matte and slag. Suggesting these samples represent an intermediate stage of the production process, potentially matte smelting, rather than raw or final products. As such these samples will be further discussed in a subsequent paper on the VC copper production assemblage.

4.2 Axes

Different types of microstructure are visible for the axes: 7 samples have been submitted to a more or less complete cycle of hammering and annealing (PBL/24, PBL/25, PBL/29, PBL/30, PBL/38, PBL/44, PBL/45), some samples don't have a completely homogenized structure. One sample is as-cast (PBL/26) and two (PBL/9 and PBL/22) are just slightly cold-worked with dendrites present alongside strain lines from hammering (Figure 6.d). Sulphide inclusions are visible in all axes (Table 4). All the axes are composed of a binary alloy of copper and tin, with a wide range of tin content but some samples have intergranular corrosion, which can distort the original tin content. To avoid this corrosion problem, SEM-EDS spot analyses were also done on the residual metal phases, though these too risks not being entirely representative of the original alloy.

4.3 *Ge*

The typological differences seen between the heavily stylised *ge* excavated from Puen Baolo and those with simpler forms from the Vilabouly Museum are supported by their microstructures and composition. The excavated *ge* (PBL/8) is hammered and annealed showing recrystallized grains and annealing twins (Figure 7.e) and is a bronze with 11 wt% Sn, while the three other *ge* (VC/1,2 and 3) have a slightly granular structure, neither twins nor preserved dendrites (Figure 7.f) and are bronzes as well but with a range of tin between 1.6 and 6 wt% (SEM-EDS). These three *ge* exhibit also a higher porosity, which may be linked to a low concentration of alloying components, as well as production processes.

4.4 Bells, drums and others

For the other typologies, different microstructures can be seen: the bells are as-cast, showing an alpha solid solution matrix of clear dendritic segregation, with one sample (VC/10) exhibiting an $\alpha + \delta$ phase. The drum is also as-cast, with an $\alpha + \delta$ phase and dendritic segregation present, as well as sulphide inclusions (Figure 7.g). The bells and the drum are the only objects of the assemblage with a high enough lead content to identify their alloys as ternary Cu-Sn-Pb. Note that two of the bell and drum samples were almost completely corroded (VC/11 and VC/13). In the case of VC/13, SEM results cannot be used reliably because of excessive corrosion but for VC/11 the ternary alloy can still be deduced. The hair pin (PBL/23) is as-cast, whereas the chisel (PBL/17), the adze (PBL/28) and the spearhead (PBL/31) have been subjected to cycles of hammering and annealing. They all are bronzes with variable Sn content. Sulphide inclusions can be observed in all (Table 4).

4.5 Inclusions

SEM-EDS analyses are useful in the characterisation of inclusions in back-scattered electron (BSE) mode. In addition to the sulphide inclusions present in most of the samples (Table 4), the BSE mode permitted the identification of another type of inclusion, composed of copper with lead and bismuth; less present than the sulphides, though sometimes in close association. These inclusions are mostly observed in the ingots (bowtie, bowl and conical) as well as one bell (VC/10), the chisel axe (PBL/17), three axes (PBL/24, PBL/25, PBL/45) and the adze (PBL/28). For the bowl ingots differences are to be noted in these inclusions, lead is not present and arsenic is detected. The SEM-EDS analyses give the major composition of the sulphide inclusions, with respect to copper, sulphur and sometimes iron, but also identifies unusual components of selenium or tellurium at concentrations of up to 6 wt. %. Selenium and tellurium regularly are found in association in copper ore mineralizations so it not so surprising to find these elements in the inclusions. These elements can be seen as relict of the original smelting charge not fully eliminated during the process.

4.6 Lead isotopes

The lead isotope signatures for the VC metals samples were plotted with those previously obtained by the Southeast Asian Lead Isotope Project (SEALIP) (Pryce et al., 2011) for seven slags from Puen Baolo and two slags and one ingot from Thong Na Nguak on three biplot of $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios (Figure 8). Overall the new data (Table 6) fall within or close to the Vilabouly Complex signature but with some notable outliers: PBL/9 an axe, PBL/10 a bowtie ingot, PBL/28 an adze, PBL/17 a chisel, PBL/41 and PBL/43 unidentified fragments, PBL/45 an axe, PBL/46 a knife, PBL/47 a bowtie ingot, VC/1 and VC/2 two *ge*, VC/9, 10 and 11 the three bells and VC/12 and VC/13 the two drums, that cannot be said to match under our subjective but conservative criteria. Pryce et al. 2011 tested Kernel Density Estimates on the available dataset but found the attributions overly optimistic given that the majority of SEALIP data plot relatively close to the VC signature, when the mining complex lies on the Truong Son belt, likely to contain as yet unknown early production sites with potentially similar signatures, and when a good proportion of the signatures derive from leaded alloys and thus relate to lead rather than copper exchange networks (Pryce et al., 2014). The primary copper production signatures for Phu Lon and the Khao Wong Prachan Valley (Nil Kham Haeng and Non Pa Wai; Figure 8) (Thai production) were also added to the plots, both of which were highly inconsistent with the new VC data.

5-Discussion

5.1- Technical characteristics

Given the microstructural and compositional data the objects can be interpreted as not all being of the same 'quality'. A correspondence seems to exist between the artefact type and the alloys chosen to make them, and no significant alloy variation is noted within an object category: ingots are made of copper; the axes, the adze, the spearhead, the *ge* and the chisel are made of bronze; and bells and drums of leaded bronze. Correspondences can also be seen between the casting and post-casting treatments and the object types. Ingots are as cast because they are intermediate products and presumably didn't need other treatment. The axes are mainly annealed and hammered even if three exceptions are recorded (PBL/9, PBL/22 and PBL/26). Thermo-mechanical treatments can improve mechanical performance, which might be sought for utilitarian objects such as axes, adzes, spearheads and chisels. The bells and the drum are generally as-cast because they imply more complex molds. As discussed in 4.1, the ingots have a equi-axed granular structure, mixed with dendrites in some cases. Equi-axed grains indicate a slow cooling rate leading to the absence of dendrites (Scott, 1991), which excludes quenching. This reconstruction seems feasible: VC ingots were poured and left to cool.

The bowl and conical ingots from TNN have a higher sulphur content at 1-2 wt%. which is linked to the composition of the original smelting charge. The bowl and conical ingots from TNN are different to the PBL ones in their sulphur content. Differences are also present in the bowl ingots' inclusions, which contain no bismuth as opposed to the bowties, and have arsenic and antimony contents. However, taking into account that the presence of equi-axed grains in ingots are a sign of a slow cooling rates, with up to 3 wt% Fe, Fe-rich precipitates should be visible in the samples according to the Cu-Fe binary system, yet none are present. The iron is present in oxide form for one sample (PBL/11) identified as goethite (α-FeOOH), which may indicate that α-iron was originally present in the sample and linked to an iron content higher than 3 wt%. Furthermore, the goethite's spheroidal shape may be indicate a liquid state was achieved. An estimation of the original Fe content was attempted by OM and image treatment by calculating the surface occupied by the goethite and by the copper matrix, assuming that goethite was originally composed of nearly 100 wt.% Fe. Therefore, Fe metal content estimation for PBL/11 is around 20 wt. % (assuming that the millimeter scale of the sample is not representative of the full object composition). The fact that six of the twenty bowtie ingots have significant iron contents may suggest variations in the production process or at least poor mastery of the procedure. It has also been suggested in some cultural contexts that iron reduction was encouraged in the case of metals used for currency where density has an importance and thus an economic advantage is gained, albeit at the cost of dishonesty (Craddock and Meeks. 1987). There are a few examples of deliberate copper-iron alloys in the literature for the Mediterranean world, i.e. Roman *ramo secco* bars, as well as Indian coins between 20-40 wt. % Fe (VII century AD) and also a Chinese ingot from the Eastern Zhou Period (8^{th}-3^{rd} c. BC) with 34 wt. % Fe. Other than PBL/11, the VC iron contents are lower so it cannot be concluded that copper-iron alloys were deliberately produced. However, in terms of ingot debasement, they can be compared to the conical ingot from Thong Na Nguak (TNN/3) that had ceramic fragments at its core, seemingly to reduce the quantity of copper therein but at the risk of its lower density being identified by potential customers. Too also note, that bowtie and conical ingots are never found associated in context, the first one correspond to a Bronze Age activity and the second one to Iron Age. Bowtie may, in this way, be associated to a Bronze Age process. The iron content of the bowtie ingots could also arise from the lack of a refining step after smelting but the ingots may have been used for export with low risk of retribution or the technique was simply not known. In any event, refining was not systematically fully accomplished.

Only two ingots from TNN have been sampled for this study, which prevents reliable conclusions being drawn. Potential explanations could be that the two production sites were not using the same raw materials (minerals from nearby but different mineralisations) or production processes. The bowl ingots, with no formal excavation context, may also be from a later period, which might imply a different process though this would not of course affect their lead isotope signature if the same mineralization was used. Indeed, this ingot type is also recorded for the Bang Krachai II shipwreck in the Gulf of Thailand, dated to the late first millennium AD and with a comparably high sulphur content (Pira Venunan pers.comm., BROGLASEA data forthcoming).

Concerning iron contents in the copper ingots, most of which probably represent post-depositional corrosion but six samples with up to 3wt% of iron stand out and may indicate that (at least occasionally) VC copper smelting furnaces got too hot and their atmospheres too reducing with the use of iron oxides in the smelting charge, which can lead to the formation of metallic iron (Cooke and Aschenbrenner, 1975; Craddock and Meeks, 1987; Rostoker et al., 1989). Metallic iron's presence can also be explained by the use of iron tools during the smelting and/or alloy making that can also lead to incorporation of iron in copper (Cooke and Aschenbrenner, 1975).

5.2-VC bronze production?

Assuming for the time being that VC copper mining and smelting activity is a given, we can here begin to address whether alloying was taking place at the Vilabouly Complex? All the non-ingot artefacts are alloyed with tin, and occasionally with lead. Plutonic and placer tin sources are abundant in well-defined geological zones within Southeast Asia (Hutchison, 2005; Hutchison and Taylor, 1978; Schwartz et al., 1995) and are amongst the largest in the Old World. None, however, are known to exist in the direct vicinity of Vilabouly. Tin deposits are known in northern Laos and especially in Phathen Valley, Khammuane Province (Lahiri-Dutt and Invouvanh, 2010) close to the Savannakhet Province, with a possible transport route down the Mekong. Ongoing analyses on a crucible fragment assemblage from the VC may permit the confirmation of bronze and leaded bronze manufacture on site, with the identification of tin or/and lead traces. Looking at the other production sites in Southeast Asia, tin traces were recorded in the Khao Wong Prachan Valley production in Cu smelting slags of Nil Kham Haeng (Pryce et al., 2010) where at least two bronze artefacts were also discovered. At Phu Lon some adhering interior crucible slags were also found to contain prills of tin mixed with copper (Pryce et al., 2011; Vernon, 1996) showing that tin was processed on site (Pigott and Natapintu. 1988; Pigott and Ciarla. 2007; Pryce et al. 2011). Further evidence for casting at Vilabouly does exist but is surprisingly sparse, consisting of only two possible sandstone mold fragments in context at Puen Baolo but apparently for conical ingots casting. One explanation may be the loss of the molds over time. Observations made by (Ottaway and Wang, 2004) have shown that clay molds, tempered with chaff, fired at 700-900°C and used for casting bronzes, disintegrate into unrecognizable shapes after being left in open air for less than a year. Bivalve molds without organic temper have been recorded with some regularity at protohistoric Southeast Asian settlement sites. Discovered in graves or/and possible casting sites, like Ban Non Wat, which has a number of small bivalve molds and a set of molds for mass-producing bangles, and Non Pa Wai in the Khao Wong Prachan Valley, where ceramic molds for socketed-axes and arrow heads, mostly fragmentary, have been identified (Higham, 2014; Pigott et al., 1997). One considered hypothesis for Vilabouly is that smelting and casting activities do not take place in the same area, which could explain why so little evidence of casting was recovered at Puen Baolo. One of the very rare ethnoarchaeological studies of copper-bronze production, among castes in Nepal (Anfinset, 2011), shows inter and even intra village specialisation, with some doing the mining and smelting, and others the casting.

5.3-Metal exchange networks

One of the main objectives was to place Vilabouly Complex copper production in the context of Southeast Asian metal exchange networks using lead isotope analysis. The Vilabouly Complex has furnished a large corpus of different ingot shapes and it remains to determine how Vilabouly metals were exchanged: in which form are the objects transported, ingot or final object? Both? Do they have different productions for local use and exportation? The dimensions observed for the bowtie ingots are quite uniform in shape (for the whole ones); but their mass is quite variable, suggesting that the standardization was only superficial, or unimportant to ancient producers and consumers. No other ingots of this type have been recorded in Southeast Asia. However, this form is similar to that used for Angkorian Period (IX century AD-XV century AD) iron crampons used in temple masonry (Leroy et al., 2017), which though much later could indicate some regional preference for this shape – especially as iron ingots were known to serve as currency as late as the 19th century Cambodia (Harmand, 1876).

The lead isotope results permit us to know if the objects discovered at the Vilabouly Complex were produced using copper locally smelted or otherwise – we cannot answer where the actual casting took place in the absence of secondary production assemblages. This question is most pertinent in the case of the objects with a "foreign" typology, as per the *ge* and the drums. As seen in part 4.3, with the structural and compositional analysis, differences are present between the *ge*. The three *ge* from the museum (VC/1, VC/2 and VC/3) are typologically simpler and have an as-cast structure with a lower tin content as compared to the Puen Baolo example (PBL/8), which has been worked and annealed and has a higher tin content (11 wt. %). The obvious expectation was that the one from Puen Baolo would be an import whereas the three others were local imitations, but this transpired not to be the case. The production of classically Han material culture with VC copper (whether at the VC or elsewhere) has naturally major implications for the local population history but we feel that a single sample is insufficient to make any reliable interpretation. An initial typological study of the VC *ge* has identified, perhaps unsurprisingly, an association with Vietnamese *ge* (Livingston. C, 2014).

The lead isotope signatures for the majority of the artefacts in this study are highly consistent with the previously defined Vilabouly Complex copper signature (Pryce et al., 2014, 2011). The ingots plot in the Vilabouly field, confirming ingot primary production on site, with the two exceptions (PBL/10 and PBL/47) indicating the possible recycling of non-local metal and/or the use of this ingot form at other production sites. Plotting all the ingots' signatures according to their typology we can see minor but clear distinctions (Figure 9). The bowtie ingots from Puen Baolo cluster apart from the chance find bowl ingots and Thong Na Nguak conical ingots, which may indicate different, but still local, ore mineralizations were used, perhaps varying through time as resources were consumed or small-scale political boundaries determined access. This isotopic observation supports differences seen in elemental data for the ingot types; the bowtie ingots containing less sulphur than the conical and bowl ingots. These isotopic and elemental differences may relate to the association of the Puen Baolo and Thong Na Nguak production sites to the Khanong and Thengkham South mineralizations mentioned above, but unfortunately we cannot confirm this as the LXML mineral data are proprietary. Of the 21 tin-alloyed objects, 12 plot well within the Vilabouly copper signature, which would seems to reinforce an interpretation of VC bronze production, even if we recognize that VC copper could have been exported to cast bronzes elsewhere and the final products re-imported, or indeed that there is another primary copper producer with near identical lead isotope ratios.

The artefacts inconsistent with the VC copper signature are two bowtie ingots (PBL/10, PBL/47), the chisel (PBL/17), the adze (PBL/28), two fragments (PBL/41 and PBL/43), two axes (PBL/9 and PBL/45), the knife (PBL/46), two *ge* (VC/1, VC/2), the three bells (VC/9, VC/10 and VC/11) and the two drums (VC/12, VC/13). The sixteen outliers observed included seven from the Vilabouly Museum with poor context and 2 from PBL WEST (adjacent to the main excavation). As it should always be considered, mixing, alloying and recycling practices can also explain the signature differences observed for these sixteen samples. There is as yet no evidence for VC recycling or alloying practices but it is a strong likelihood given the scale of the locality. The three bells and the drums from the museum are alloyed with lead so the signature pertains to lead rather than copper exchange networks, and not much can be said about these in the absence of regional lead production studies (Pryce, 2012). However, it can be noted that the bells' and drums' signatures are different, which suggests multiple lead sources and associated producing populations. Besides, the bells come from a separate context and are of mid-late Iron Age pan-Southeast Asian type.

6. Conclusion

This paper presents the first archaeometallurgical study for Lao PDR copper-base objects with metallographic, compositional and lead isotopes data permiting us to address copper-bronze working practices in the protohistoric Vilabouly Complex. The results revealed objects types conformed with the alloy and the working treatments, with copper (ingots), bronze (axes, adzes, knifes, *ge*, hair pins, spearheads, chisels) and leaded-bronze (drums and bells). Some variation can be seen in the bowtie ingots, with six samples bearing iron contents up to 3 wt%, which may suggest that fluctuations were present in the production process with a lack or a poor mastery of smelting conditions and/or refining steps. Or that these represent an earlier Bronze Age technology in the area, overtaken by Iron Age conical ingot processes

The lead isotopic data show that most of the objects are consistent with the Vilabouly Complex signature, suggesting that primary copper production, and most probably secondary production, were taking place on site. Lead isotopic and elemental data also permit the identification of differences between bowtie ingots from Puen Baolo and conical ingots from Thong Na Nguak (as well as bowl ingots with poor context), the two production sites currently known in Vilabouly. The isotopic data for bowtie and conical/bowl ingots suggests that different copper mineralizations were used for each smelting site, possibly varying through time. Some of the observations made on the copper-base objects may then be connected to results of the full reconstitution study of the copper production at VC with analyses of artefacts from the entire *chaine operatoire*: ores, slags and crucibles, which aims to access different considerations like the composition of the smelting charge even more the type of ore used (oxidic, carbonate and/or sulphidic) and the different production steps. For instance, will the complete technological reconstruction confirm production differences between Puen Baolo and Thong Na Nguak?

More investigation is needed to place convincingly the Lao PDR in the long-standing regional "origins of metallurgy" debate as the Vilabouly Complex is located to the south, away from the Chinese border regions critical for resolving the technological transmission issues. The identification of *ge* and drums of Dongson type with matching VC lead isotope signatures may denote at a minimum contact and local appropriation of Vietnamese and/or Chinese cultural tradition, if not the presence of non-local artisans but far more sites and metal assemblages representing production and consumption behaviours need to be studied before any reliable reconstruction of these potentialities can be made. We hope, nevertheless, this paper will be an important step in that direction.

Acknowledgments

This work is financed by the French National Research ANR BROGLASEA "Bronze and Glass as Cultural Catalysts and Tracers in Early Southeast Asia" started in 2016 and directed by T.O. Pryce. We also offer our thanks to the Lao archaeologists and the mining concession operator MMG-LXML who provided us help and permits in Laos for the export of the artefacts.

Bibliography

Anfinset, N., 2011. Social and Technological Aspects of Mining, Smelting and Casting Copper: an Ethnoarchaeological study from Nepal. Deutsche Bergbau-Museum Bochum.

Calo, A., 2014. Trails of Bronze Age Drums Across Early Southeast Asia: Exchange Routes and Connected Cultural Spheres. Institute of Southeast Asian Studies, Singapore.

Chen, K., Rehren, T., Mei, J., Zhao, C., 2009. Special alloys from remote frontiers of the Shang Kingdom: scientific study of the Hanzhong bronzes from southwest Shaanxi, China. J. Archaeol. Sci. https://doi.org/10.1016/j.jas.2009.04.016

Ciarla, R., 2007. Rethinking Yuanlongpo: The Case for Technological Links between the Lingnan (PRC) and Central Thailand in the Bronze Age. East West 57, 305–328.

https://doi.org/10.2307/29757731

Cloquet, C., Carignan, J., Libourel, G., 2006. Atmospheric pollutant dispersion around an urban area using trace metal concentrations and Pb isotopic compositions in epiphytic lichens. Atmos. Environ. 40, 574–587.

Cooke, S.R.B., Aschenbrenner, S., 1975. The Occurrence of Metallic Iron in Ancient Copper. J. F. Archaeol. 2, 251–266. https://doi.org/10.1179/009346975791491015

Craddock, P.T., Meeks, N.., 1987. Iron in Ancient Copper. Archaeometry 29, 187–204. https://doi.org/10.1111/j.1475-4754.1987.tb00411.x

Cromie, P.W., 2010. Geological setting, geochemistry and genesis of the Sepon gold and copper deposits, Laos. Centre of Excellence in Ore Deposits University of Tasmania.

Dussubieux, L., Pryce, T.O., 2016. Myanmar's role in Iron Age interaction networks linking Southeast Asia and India: Recent glass and copper-base metal exchange research from the Mission Archéologique Française au Myanmar. J. Archaeol. Sci. Reports. https://doi.org/10.1016/j.jasrep.2016.01.005

Harmand, D., 1876. Voyage au Cambodge. Bull. la Société Géographie 337–367.

Hauptmann, A., Schmitt-Strecker, S., Levy, T.E., Begemann, F., 2015. On Early Bronze Age Copper Bar Ingots from the Southern Levant. Am. Sch. Orient. Res.

Higham, C., 2014. Early Mainland Southeast Asia : from first Humans to Angkor. River Book, Bangkok.

Higham, C., 1996. The Bronze Age of Southeast Asia. Cambridge University Press, New York.

Higham, C., Higham, T., Kijngam, A., 2011. Cutting a Gordian Knot: The bronze age of Southeast Asia: Origins, timing and impact. Antiquity. https://doi.org/10.1017/S0003598X00067971

Higham, C.F., Rispoli, F., 2014. The Mun Valley and Central Thailand in prehistory: Integrating two cultural sequences. Open Archaeol. 1, 2–28. https://doi.org/10.2478/opar-2014-0002

Higham, C.F.W., Higham, T., 2009. A new chronological framework for prehistoric Southeast Asia, based on a Bayesian model from Ban Non Wat. Antiquity 82, 1–20.

Hirao, Y., Ro, J.-H., 2013. Chemical composition and lead isotope ratios of bronze artifacts excavated in Cambodia and Thailand, in: Yasuda (Ed.), Water Civilisation: From Yangtze to Khmer Civilizations. Springer.

Hutchison, C.., 2005. The geological Framework, in: Gupta, A. (Ed.), The Physical Geography of Southeast Asia. New York, pp. 3–23.

Hutchison, C.., Taylor, D., 1978. Metallogenesis in SE Asia. J. Geol. Soc. London. 135, 407–428.

Lahiri-Dutt, K., Invouvanh, C., 2010. Gender in Artisanal Tin Mining in Lao PDR A Case Study in Ban Moua Khay and Ban Nanhi, Khammuane Province.

Leroy, S., Hendrickson, M., Bauvais, S., Vega, E., Blanchet, T., Disser, A., Delque-Kolic, E., 2017. The ties that bind: archaeometallurgical typology of architectural crampons as a method for reconstructing the iron economy of Angkor, Cambodia (tenth to thirteenth c.). https://doi.org/10.1007/s12520-017-0524-3

Livingston. C, 2014. Who were the people of ancient Vilabouly? Exploring origins and relationships through the study of Ge. James Cook University, Townsville.

Lorge, P.A., 2012. Chinese Martial Arts: From Antiquity to the Twenty-First Century. Cambridge

University Press, New York.

Manhes, G., Allègre, C.J., Dupré, B., Hamelin, B., 1980. Lead isotope study of basic-ultrabasic layered complexes: Speculations about the age of the earth and primitive mantle characteristics. Earth Planet. Sci. Lett. 47, 370–382. https://doi.org/10.1016/0012-821X(80)90024-2

Manini, T., Aquino, J., Gregory, C., Aneka, S., n.d. Discovery of the Sepon District gold and copper deposits, Laos.

Mei, J., 2000. Copper and bronze metallurgy in late prehistoric Xinjiang: its cultural context and relationship with neighbouring regions. Cambridge.

Natapintu, S., 1988. Current research on ancient copper-base metallurgy in Thailand, in: Charoenwongs, P., Bronson, B. (Eds.), Prehistoric Studies: The Stone and Metal Ages in Thailand. Amarin Printing Group, Bangkok, pp. 107–124.

Ottaway, B.S., Wang, Q., 2004. Casting Experiments and Microstructure of Archaeologically Relevant Bronzes. British Archaeological Reports (BAR) International Serie 1331.

Pienitz, R., Aebischer, S., Cloquet, C., Carignan, J., Maurice, C., 2015. Disruption of the geochemical metal cycle during mining: Multiple isotope studies of lake sediments from Schefferville, subarctic Québec. Chem. Geol. 412, 167–178. https://doi.org/10.1016/j.chemgeo.2015.07.028

Pigott, V., Natapintu, S., 1988. Archaeological investigations into prehistoric copper production: the Thailand Archaeometallurgy Project, 1984-86, in: Maddin, R. (Ed.), The Beginning of the Use of Metals and Alloys. M.I.T Press, Cambridge, pp. 156–162.

Pigott, V.C., Ciarla, R., 2007. On the origins of metallurgy in prehistoric Southeast Asia: the view from Thailand, in: La Niece, S., Hook, D.R., Craddock, P.T. (Eds.), Metals and Mines – Studies in Archaeometallurgy. London Archetype Publication.

Pigott, V.C., Weisgerber, G., 1998. Mining archaeology in geological context. The prehistoric copper mining complex at Phu Lon, Nong Khai Province, northeast Thailand, in: Rehren, T., Hauptmann, A., Mulhy, J.. (Eds.), Metallurgica Antiqua: In Honour of Hans-Gert Bachmann and Robert Maddin. Deutsche Bergbau-Museum Bochum, pp. 135–161.

Pigott, V.C., Weiss, A., Natapintu, S., 1997. Archaeology of Copper Production: Excavations in the Khao Wong Prachan Valley, Central Thailand, in: Ciarla, R., Rispoli, F. (Eds.), South-East Asian Archaeology 1992. Proceedings of the Fourth International Conference of the European Association of South-East Asian Archaeologists. Rome, 28th September-4th October 1992. Rome, Instituto Italiano per l'Africa e l'Oriente, pp. 119–157.

Pryce, T.O., 2012. A flux that binds? The Southeast Asian Lead Isotope Project, in: Jett, P., Douglas, J. (Eds.), Proceedings of the 5th Forbes Symposium on Ancient Asian Bronzes. Smithsonian, Washington, DC, pp. 113–121.

Pryce, T.O., Baron, S., Bellina, B.H.M., Bellwood, P.S., Chang, N., Chattopadhyay, P., Dizon, E., Glover, I.C., Hamilton, E., Higham, C.F.W., Kyaw, A.A., Laychour, V., Natapintu, S., Nguyen, V., Pautreau, J.P., Pernicka, E., Pigott, V.C., Pollard, M., Pottier, C., Reinecke, A., Sayavongkhamdy, T., Souksavatdy, V., White, J., 2014. More questions than answers: The Southeast Asian Lead Isotope Project 2009-2012. J. Archaeol. Sci. https://doi.org/10.1016/j.jas.2013.08.024

Pryce, T.O., Brauns, M., Chang, N., Pernicka, E., Pollard, A.M., Ramsey, C., Rehren, T., Souksavatdy, V., Sayavongkhamdy, T., 2011. Isotopic and technological variation in prehistoric Southeast Asian primary copper production. J. Archaeol. Sci. https://doi.org/10.1016/j.jas.2011.07.016

Pryce, T.O., Calo, A., Prasetyo, B., Bellwood, P., 2018a. Copper-base Metallurgy in Metal-Age Bali:

Evidence from Gilimanuk, Manikliyu, Pacung, Pangkung Paruk and Sembiran: Copper-base metallurgy in Metal-Age Bali. Archaeometry 60.

Pryce, T.O., Kyaw, A.A., Kyaw, M.M., Win, T.T., 2018b. A first absolute chronology for Late Neolithic to Early Bronze Age Myanmar: New AMS 14C dates from Nyaung'gan and Oakaie. Antiquity 92.

Pryce, T.O., Pigott, V.C., Martinón-Torres, M., Rehren, T., 2010. Prehistoric copper production and technological reproduction in the Khao Wong Prachan Valley of Central Thailand. Archaeol. Anthropol. Sci. https://doi.org/10.1007/s12520-010-0043-y

Rispoli, F., Ciarla, R., Pigott, V.C., 2013. Establishing the Prehistoric Cultural Sequence for the Lopburi Region, Central Thailand. J. World Prehistory. https://doi.org/10.1007/s10963-013-9064-7

Rostoker, W., Piggott, V.C., Dvorak, J.R., 1989. Direct Reduction to Copper Metal by Oxide-Sulfide Mineral Interaction. Archaeomaterials 69–87.

Schwartz, M.O., Rajah, S.S., Askury, A.K., Putthapiban, P., Djaswadi, S., 1995. The Southeast Asian tin belt. Earth Sci. Rev. 38, 95–293. https://doi.org/10.1016/0012-8252(95)00004-T

Scott, D.A., 1991. Metallography and microstructure of ancient and historic metals. The Getty Conservation Institute.

Sillitoe, R.H., 1998. Comments on geology and exploration potential for copper and gold deposits at Sepon, Laos. A Rep. Prep. Lane Xang Miner. Ltd.

Solheim, W., 1968. Early Bronze in Northeastern Thailand. Curr. Anthropol. 9, 59–62.

Thirlwall, M.F., 2002. Multicollector ICP-MS analysis of Pb isotopes using a 207Pb - 204Pb double spike demonstrates up to 400 ppm/amu systematic errors in Tl-normalization. Chem. Geol. 184, 255–279.

Tucci, A., Sayavongkhamdy, T., Chang, N., Souksavatdy, V., 2014. Ancient Copper Mining in Laos: Heterarchies, Incipient States or Post-State Anarchists? J. Anthropol. Archaeol. https://doi.org/10.15640/jaa.v2n2a1

Vander Voort, G.F. (Ed.), 2004. Metallography and Microstructures, in: ASM Handbook. pp. 1844–1850.

Vernon, W.W., 1996. The crucible in copper-bronze production at prehistoric Phu Lon, northeast Thailand: analyses and interpretation., in: Bulbeck, F.D., Barnard, N. (Eds.), Ancient Chinese and Southern Asian Bronze Cultures 1988. SMC publishing, Taipei, pp. 809–820.

White, J.C., 1988. Early east asian metallurgy: the southern tradition, in: Maddin, R. (Ed.), The Beginning of the Use of Metals and Alloys: Papers from the Second International Conference on the Begining of the Use of Metals and Alloys, Zengzhou, China. MIT Press, Cambridge, pp. 175–181.

White, J.C., Hamilton, E., 2018a. Ban Chiang, Northeast Thailand, Volume 2B: Metals and related evidence from Ban Chiang, Ban Tong, Ban Phak Top and Don Kland. University of Pennsylvania Museum of Archaeology and Anthropology, Philadelphia.

White, J.C., Hamilton, E.G., 2018b. Ban Chiang, Northeast Thailand, Volume 2A: background to the study of the metal remains. University of Pennsylvania Museum of Archaeology and Anthropology, Philadelphia.

White, J.C., Hamilton, E.G., 2014. The transmission of early Bronze Age technology to Thailand: New perspectives, in: Roberts, B.W., Thornton, C.P. (Eds.), Archaeometallurgy in Global Perspective. pp. 805–852. https://doi.org/https://doi.org/10.1007/978-1-4614-9017-3_28

White, J.C., Hamilton, E.G., 2009. The Transmission of Early Bronze Technology to Thailand: New Perspectives. J Wolrd Prehist 22. https://doi.org/10.1007/s10963-009-9029-z

Table 1. APAP and KPF doses/animal weight/day (50 mg/kg/day and 5 mg/kg/day respectively)

Animal No.	**Day**	**Weight (g)**	**Dose (mg)**	**Weight (g)**	**Dose (mg)**
		APAP		**KPF**	
1	I	28	1.4	27	0.135
	II	29	1.45	29	0.145
	III	32	1.6	31	0.155
2	I	34	1.7	36	0.18
	II	33	1.65	37	0.185
	III	37	1.85	38	0.19
3	I	29	1.45	29	0.145
	II	33	1.65	31	0.155
	III	36	1.8	34	0.17
4	I	35	1.75	33	0.165
	II	36	1.8	36	0.18
	III	39	1.95	38	0.19
5	I	34	1.7	34	0.17
	II	34	1.7	35	0.175
	III	36	1.8	38	0.19
6	I	37	1.85	36	0.18
	II	38	1.9	38	0.19
	III	39	1.95	41	0.205
7	I	32	1.6	32	0.16
	II	35	1.75	35	0.175
	III	37	1.85	37	0.185
8	I	29	1.45	29	0.145
	II	33	1.65	33	0.165
	III	35	1.75	35	0.175
9	I	32	1.6	32	0.16
	II	35	1.75	35	0.175
	III	37	1.85	37	0.185
10	I	29	1.45	29	0.145
	II	32	1.6	32	0.16
	III	34	1.7	34	0.17

Table 2. The main tendency and dispersion parameters for the mean values (the average values computed for the 3 tested days) for the mice weight and APAP/KPF doses

Statistics	Mean Weight Group 1	Mean APAP dose	Mean Weight Group 2	Mean KPF dose
Mean	33.97	1.70	34.23	17.83
Standard Error	0.77	0.04	0.87	0.41
Median	34.67	1.73	34.67	18.20
Mode	34.67	1.73	35.67	18.20
Standard Deviation	2.44	0.12	2.75	1.28
Sample Variance	5.96	0.01	7.56	1.64
Kurtosis	-0.05	-0.05	0.16	-0.05
Skewness	-0.11	-0.11	-0.51	-0.11
Range	8.33	0.42	9.33	4.38
Minimum	29.67	1.48	29.00	15.58
Maximum	38	1.90	38.33	19.95
Sum	339.67	16.98	342.33	178.33
Count	10	10	10	10

Table 3. The ANOVA Two-Factor Without Replication test applied on the APAP injected mice group for weight and for drug dose.

Source of variation	SS	df	MS	F	F crit.
Weight					
The analysis on mice	160.9667	9	17.88519	16.82578	2.456281149
The analysis on the 3 studied days	92.86667	2	46.43333	43.68293	3.554557146
Error	19.13333	18	1.062963		
Total	272.9667	29			
Drug dose					
The analysis on mice	0.402417	9	0.044713	16.82578	2.456281
The analysis on the 3 studied days	0.232167	2	0.116083	43.68293	3.554557
Error	0.047833	18	0.002657		
Total	0.682417	29			

Table 4. The ANOVA Two-Factor Without Replication test applied on the KPF injected mice group for weight and for drug dose.

Source of variation	SS	df	MS	F	F crit.
Weight					
The analysis on mice	223.6333	9	24.84815	59.90179	2.456281
The analysis on the 3 studied days	105.8667	2	52.93333	127.6071	3.554557
Error	7.466667	18	0.414815		
Total	336.9667	29			
Drug dose					
The analysis on mice	0.005591	9	0.000621	59.90179	2.456281
The analysis on the 3 studied days	0.002647	2	0.001323	127.6071	3.554557
Error	0.000187	18	1.04E-05		
Total	0.008424	29			

Table 1: Radiocarbon dates with sample number and calibrated dates (at 95% confidence) for VC sites, Puen Baolo, Thong Na Nguak, Khanong A2, Thenkgham East and Thenkgham South D.

Table 2: VC copper-alloy samples with associated context (square, layer, feature and catalogue number) and attribute (mass, length, piece).

Table 3: CRM results with pXRF and SEM-EDS analysis to test accuracy (wt%).

Table 4: Microstructural observation for all the samples along with the type of inclusions present. H/A = Hammered/Annealed; S = Sulphide inclusion with indication of selenium-tellurium (Se-Te) presence for some samples.

Table 5: Compositional results for 60 samples obtained by pXRF and SEM-EDS depending on the corrosion products (wt%).

Table 6: LI ratios for the lead isotopic results in Figure 8.