

New evidence of metal exchange in Southeast Asia during the Iron Age: based on the scientific analysis of excavated bronze in Vilabouly, Laos

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New evidence of metal exchange in Southeast Asia during the Iron Age: based on the scientific analysis of excavated bronze in Vilabouly, Laos

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Abstract

This study combines several approaches to investigate bronze objects uncovered at the Thengkham East site in Vilabouly in 2018. According to SEM-EDS research, most copper wares are binary alloys of copper and tin. Metallography (OM) studies have demonstrated that generally, the forming technique of copper wares is casting, and the microstructure of extensive annealing is shown in some areas of isolated weapons. The experimental results for this set of samples were consistent with the previously published style of Vilabouly copperware. Comparative investigations suggest that multiple sites within Vilabouly had similar metallurgical industry technology and that there is a possibility of exchanging intermediate goods between mining/ore processing sites and ordinary consuming locations. Lead isotope studies have corroborated the prevailing opinion that during the early Iron Age in Ban Non Wat, central Laos superseded Thailand in supplying copper to various sections of the Indo-China Peninsula. Given Lead isotope analysis also indicate that there had been connections between Thailand, Laos, and Cambodia in the metallurgical industry during the same period, this study provides support for the hypothesis that incorporate northeastern Cambodia into the medium-range interaction zone where central Laos is located.

Keywords: Vilabouly, Iron Age, lead isotopes, copper wares, exchange, Indo-China Peninsula

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Introduction and background

Introduction

The significance of Vilabouly (Fig. 1) in the Southeast Asian metallurgical technology dissemination network has recently become a focus of Southeast Asian archaeology. Archaeologists have conducted numerous studies that have shown that mining and smelting sites in Vilabouly are extremely important for metallurgical research on the Indo-China Peninsula. During the early Iron Age of Ban Non Wat, between 420 BC and 100 AD, Vilabouly and Khao Wong Prachan Valley in central Thailand provided most of the copper for the Indo-China Pen (Cadet et al. 2019, Higham et al. 2021). The range of copper supply was quite broad and could extend as far as the Oakaie and Nyaung'gan sites in central Myanmar (Pryce et al. 2018) as well as the Khao Sek site in southern Thailand's Malay Peninsula (Pryce et al. 2018).

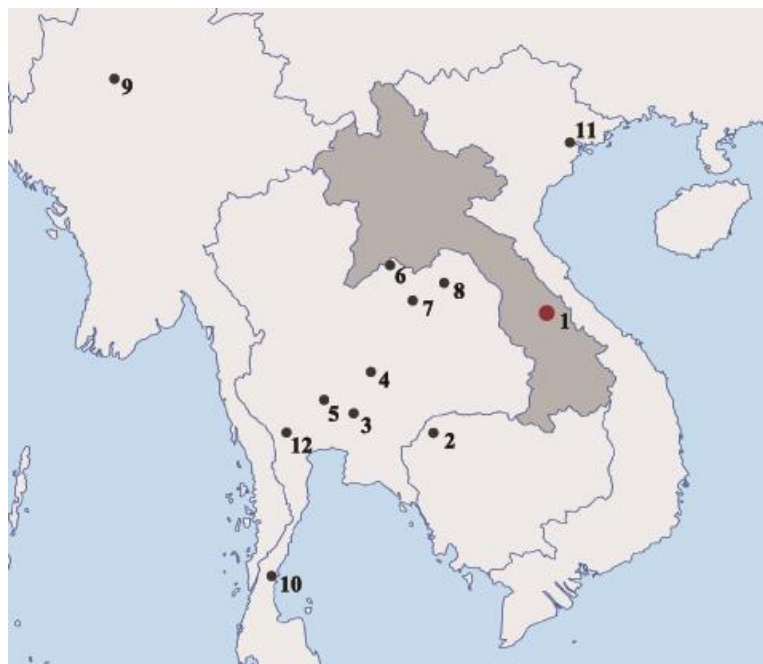


Fig. 1 The location of sites mentioned in the text: 1. Vilabouly (Sepon Mine), 2. Phum Snay, 3. Ban Pong Manao, 4. Ban Non Wat, 5. Khao Wong Phrachan Valley sites: Non Pa Wai, Nil Kham Haeng, 6. Phu Lon, 7. Non Nok Tha, 8. Ban Chiang and Ban Na Di, 9. Nyaung'gan and Oakaie, 10. Song Toh mine, 11. Dong Son

According to previous research, central Laos, central Thailand, and northeastern Thailand contained various metal-producing and metal-consuming sites from the Bronze Age to the Iron Age, with each region having short-range interaction zones. Furthermore, evidence exists that the three regions had metallurgical exchanges and

contacts and could be classified as the same medium-range interaction zone (Pryce et al. 2022). Though more research on short-range interaction zones in central and northeastern Thailand has emerged (Pryce et al. 2011, Higham et al. 2020), few scholars have discussed the interaction zone in central Laos. As a result, the full archaeological scope of Vilabouly is unknown, as are its technical characteristics; site connotations; and mining, smelting, casting, and living areas. The circulation path of metal products, mine developer affiliations, and management models are also unknown.

This dissertation used various methods to study the artefacts excavated in Vilabouly in 2018 and the Vilabouly Cultural Hall collections. It seeks to explain the production technology and raw material characteristics of these copper wares. Furthermore, this study examined related data pertaining to the Bronze Age and Iron Age in Vilabouly to determine whether there was any exchange of raw materials or technology within the mining area. Expanding the scope to the medium-range interaction zone where Vilabouly is located, this study investigated various issues related to copper metallurgy technology, copper metallurgy raw materials, and copperware-style circulation in Vilabouly using data from recent Iron Age sites in Thailand and Cambodia.

Background

Vilabouly is located in the central-southern Laos province of Savannakhet. As the first archaeological site in Laos to be discovered and one of the largest in Southeast Asia, Vilabouly has important significance in Laotian archaeology. Since the first archaeological survey of the Sepon mining area in 2001, many artefacts have been discovered, including bronze drums, spears, swords, pottery, crucibles, slag, and ingots.

Previously, the MMG Company, Lao Cultural Heritage Bureau, and James Cook University in Australia were mainly responsible for archaeological excavations in Vilabouly. Salvage excavations have taken place at Peun Baolo (2008–2012), Dragon Field (2008), Khanong A2 (2009), Malachite Cave (2011), and Tengkhamb South D. (2012) (Tucci et al. 2014). Modern mining activities severely damaged these sites, which were later salvaged. The most notable discoveries were wooden mining shafts, slags, crucibles, and copperware.

The Yunnan Provincial Institute of Cultural Relics and Sichuan University's School of Archaeology and Literature collaborated on the excavation of the Tengkhamb East site in 2018, which involved personnel from both sides. At the Tengkhamb East site,

excavations were conducted at three locations (sites #1, #2, and #3). Many crucibles of various sizes were unearthed, but no furnaces were discovered, and the slag forms were relatively uniform. This excavation yielded two significant discoveries. First, inclined shafts were discovered in Vilabouly. Previously discovered mining shafts were all vertical (Relics et al. 2018); second, stony molds used for casting copperwares were discovered at the Tengkham East site for the first time (Li 2018). These findings are extremely important for understanding ancient mining and metallurgical activities in Vilabouly.

Geology

Laos, located in the northern part of the Indo-China Peninsula is the only landlocked country in Southeast Asia. The country's terrain is long and narrow from north to south, with more plateaus than plains. It shares borders with China's Yunnan Plateau to the north, Cambodia to the south, the Mekong River Valley formed by the Mekong River and its tributaries along the basin and plains to the west, Myanmar to the northwest, Thailand to the southwest, and Vietnam to the east (Liu et al. 2016). Vilabouly is located in Thailand's Sepon Basin, which forms the southern boundary between the Longshan Mountains and the Hele High Plateau fold belt. The region is mountainous, rich in mineral resources, and a major copper and gold mine in Southeast Asia (Cromie 2010).

Brief description of samples

The samples were divided into two groups according to their sources (Table 1 and Fig. 2). Samples numbered TK1–TK8 were obtained from the 2018 excavations at the Tengkhram East site. The best-preserved bronze axes were found in TK1, TK3, TK4, TK5, and TK6, which were typically approximately 10 cm long and 6 cm wide, and the axes can be seen to have been used for cutting. There are no discernible surface patterns on the cross-section, which has a roughly oval or rectangular form. Based on their morphology, the axes can be loosely split into two categories: TK2 and TK4 differ from the others in that their overall shapes are axisymmetrical and resemble those of an adze, whereas the other axes are roughly trapezoidal in shape, with the blade being wider than the axe. TK7 is a flat, rounded bronze item with a diameter of approximately 3 cm. It is slightly bent on one side and covered with small holes and depressions of varying sizes. Conical ingots or slag ingots were used to describe this sort of artefact in earlier studies (Cadet et al. 2019), but in more current research, its nature has been modified to a multi-layered artefact (hereinafter ‘MLA’), probably as an intermediate byproduct of the metallurgical process (Cadet et al. 2022).

Numbers VL1–VL9 are samples from the Vilabouly Cultural Hall collections, collected in the same year. VL1 corresponds to a relatively well-preserved bronze drum that is 48 cm high and 64 cm wide. The tympanum is smaller than the barrel, which puffs up, whereas the waist gradually shrinks to a cylindrical shape, presumably a Don Son drum. The upper part of the barrel and the lower part of the waist are decorated with geometric patterns. There is a twelve-manifold sun motif in the middle of the tympanum, and various animals and geometric motifs are arranged around it. VL2 is a tubular bronze bangle with a braided cord pattern and hollow spherical ornaments on both ends. VL3–VL5 are all daggered axes, relatively intact, with elongated blades and wide anterior segments. VL9 is a round pie-shaped bronze artefact similar in nature to TK7.

Table 1 Basic information of samples

Lab Num	Catalogue	artifact type	provenance	sampling part	location
TK1	2018SST	axe	Thengkhram East	blade	excavation area east cross-section
TK2	2018SST TN3E1④:7	axe	Thengkhram East	stubble	2018SST TN3E1④
TK3	2018SST TN3E1④:8	axe	Thengkhram East	blade	2018SST TN3E1④
TK4	2018SST TN3E1④:4	axe	Thengkhram East	socket edge	2018SST TN3E1④
TK5	2018SST TN3E1④:5	axe	Thengkhram East	socket edge	2018SST TN3E1④

TK6	2018SST TN3E1④:6	axe	Thengkham East	blade	2018SST TN3E1④
TK7	2018SST TN1E1④:1	'mla'	Thengkham East	edge	red gravel layer
TK8	2018SST	unknown	Thengkham East	stubble	collection
VL1	AF48-10	drum	Pha Pilang	tympanum, foot	Vilabouly Cultural Hall
VL2	AF0081	bangle	--	stubble	Vilabouly Cultural Hall
VL3	AF0030 13-10	dagger- axe	Pha Pilang	blade	Vilabouly Cultural Hall
VL4	AF0030 15-10	dagger- axe	Pha Pilang	hu	Vilabouly Cultural Hall
VL5	AF0030 3-10	dagger- axe	Pha Pilang	hu	Vilabouly Cultural Hall
VL9	37 36	'mla'	Pha Vat	center	Vilabouly Cultural Hall



Fig. 2 Four types of bronze samples (1. axe; 2. MLA; 3. dagger-axe; 4. drum)

Methodology

All samples in this study were collected using special tools and numbered at the excavation site and at Vilabouly Cultural Hall. Heavily corroded areas were avoided in the sampling process. Each sample was split into two pieces: one for metallographic analysis and the other for primary, trace element, and lead isotope analyses.

Samples for metallographic and principal element analyses were inlaid with epoxy resin and their surfaces were prepared using a grinding and polishing machine (model: Struers Tegramin-20). Using SEM-EDS (model: PHENOM Pro XL; experimental parameters: accelerating voltage: 15kv, beam intensity: energy spectrum linear/mapping, high vacuum: 1 Pa, backscattering mode), the maximum area was selected for mapping to avoid rust and holes in the sample. Each sample was measured numerous times, and the average value was taken as the final outcome. The microstructure and inclusions were observed using optical microscopy (Model: Leica DM2700M) after the surface of the sample was etched with an alcoholic solution of ferric chloride hydrochloride.

After removing the surface corrosion, the remaining portion of the sample was dissolved in a specific amount of aqua regia, and the solution was then brought to a predetermined volume with deionised water so that the lead element concentration of the solution was close to 1000 ppb. The samples' trace element concentrations and lead isotope ratios were measured using the aforementioned solutions. Lead isotopes were determined using a multi-receiver inductively coupled plasma mass spectrometer (MC-ICP-MS) (model: VG-ELEMENTAL) at the Key Laboratory of Orogenic Belts and Earth Evolution, School of Earth and Space Science, Peking University. The relative errors of the $^{207}\text{Pb}/^{206}\text{Pb}$, $^{208}\text{Pb}/^{206}\text{Pb}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios were $< 0.01\%$, 0.01% , and 0.1% , respectively.

Sample preparation was completed by adding a thallium (Tl) standard solution (SRM997) to the solution so that the thallium content was approximately two-thirds of the lead content. The instrument was calibrated using the American National Bureau of Standards standard solution for lead isotope testing (SRM981); the SRM981 was tested for every three to five samples.

Trace elements were examined using a full-spectrum direct-read inductively coupled plasma emission spectrometer (ICP-AES) (model: Leeman Prodigy7) at Sichuan University's Experimental Teaching Center of Archaeology's Cultural Relics Analysis Laboratory. Before the experiment, the instrument was calibrated using

standard solutions to ensure that the relative standard deviation (RSD) of each element was less than 0.1% and that the limit of quantitation (LOQ) value of ICP-AES was maintained throughout the determination of the samples. The instrument analysis conditions were as follows: RF (high-frequency generator) power: 1.1kw, argon flow rate: 20 L/min, nebuliser pressure: 30 psig (imperial units, approximately 20MPa), peristaltic pump (sample lift) rate: 1.2 ml/min, and integration time: 30sec/time.

Results

SEM-EDS analysis

According to the SEM-EDS analysis (data visualisation is shown in Fig. 3), the copper content of the test samples ranged from 97.93% to 60.81%, and the tin content ranged from 17.98% to 1.48% (Table 2). The oxygen content of the samples reflected the degree of corrosion; the average oxygen content of the tested samples was 4.20%. Corrosion affected 80% of the samples, and their lead and tin contents were generally higher than average in the SEM-EDS analysis. Therefore, these data were not used as a basis for quantitative analysis, but only to make a qualitative judgment of the material of these samples.

The criterion for artificial addition in this case is greater than or equal to 2% (Sun et al. 2011). Only VL1 (drum) contained intentionally added lead, indicating that it was a Cu-Pb-Sn ternary alloy, whereas the remaining samples were all Cu-Sn binary alloys.

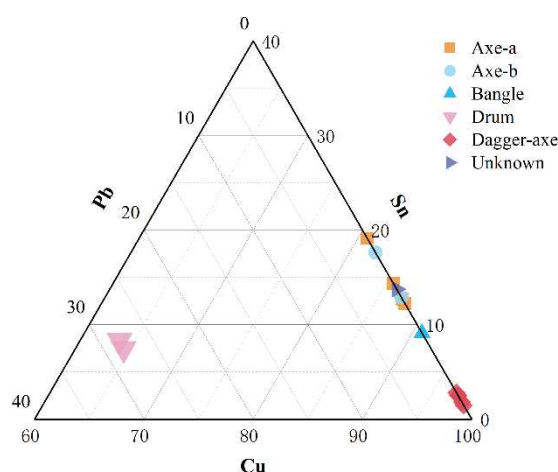


Fig. 3 Ternary diagram showing the major component of the copper-based artifact samples (Cu, Sn, Pb); the data is normalised

The dagger axes contained only a small amount of tin (between 1.5% and 2.5%), which is close to that of copper. Because the actual artefacts would have had a soft texture and the blades of these dagger axes were slender, it is highly possible that they did not serve as practical weapons in the traditional sense. The axes' average tin content was 13.86%, and the level of tin content is consistent for both types of copper axes. The tin content of the dagger axes was stable, but that of the axes varied widely, ranging from 11.62 to 17.98%. A tin content of 10% or more results in copper having good mechanical properties; thus, unlike the dagger axes, these axes should have had a practical function. VL1 (drum) had essentially identical main element content in the

drum foot and drum face, so the drum should have been cast from the same type of alloy as a whole.

Microstructure

Most of these copper wares were produced using the casting technique (Sun et al. 2011), according to the etched sections of the samples, and some samples had incomplete extensive annealing microstructure (Scott 1991), without any traces of intentional hot working by the craftsmen. The graphic below (Fig. 4) shows some typical sample microstructures. Fig. 4a shows the pronounced dendritic structure of the cast bronze. Section compression during sampling may have caused edge deformation. In Fig. 4b, the dendritic segregation was partially removed, indicating that the sample underwent incomplete extensive annealing. Fig. 4c shows the presence of dendritic segregation accompanied by obvious strain lines produced by heavy working; the cause of the strain lines is unclear. Fig 4d, similar to TK3, also shows incomplete extensive annealing. Extensive annealing only exists in the sections of the axes, and it is unclear whether the craftsmen or users intentionally heated the copper axes and then cooled

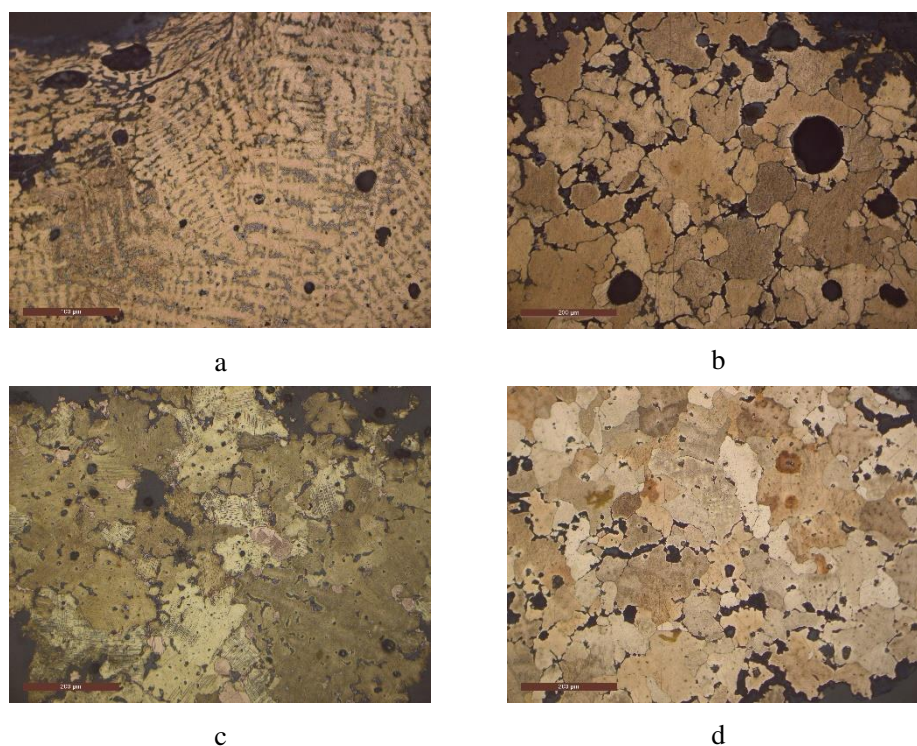


Fig. 4 Section from part of the samples etched in alcoholic ferric chloride

4a TK1(axe) magnification 200x, casting; **4b** TK3(axe) magnification 100x, incomplete extensive annealing; **4c** TK6 (axe) magnification 100x, casting with heavy working; **4d** VL3 (dagger-axe) magnification 100x, similar to TK3, incomplete extensive annealing

them slowly. The alloy types and inclusions are listed in Table 2.

Table 2 SEM-EDS results , alloy types, and processing technology of samples

Lab Num	Cu	Sn	O	S	Pb	Alloy	Microstructure	inclusion
TK1	83.90	12.18	3.50	0.42	0	Cu-Sn	As cast	copper sulfide & copper
TK2	75.89	16.24	7.52	0.36	0	Cu-Sn	As cast	copper sulfide & copper
TK3	84.66	11.80	3.33	0.21	0	Cu-Sn	Extensive annealing	copper sulfide & copper
TK4	78.59	11.62	9.30	0.48	0	Cu-Sn	As cast	copper sulfide & copper
TK5	79.65	13.34	6.65	0.37	0	Cu-Sn	As cast	copper sulfide
TK6	76.02	17.98	5.55	0.45	0	Cu-Sn	Extensive annealing with strain lines	copper sulfide & copper
TK8	79.88	12.72	7.24	0.16	0	Cu-Sn	As cast	copper sulfide & lead
VL1-bottom	61.85	7.14	4.03	0	26.99	Cu-Pb-Sn	As cast	copper sulfide
VL1-top	60.81	7.99	4.39	0	26.82	Cu-Pb-Sn	As cast	copper sulfide
VL2	87.10	8.72	4.18	0	0	Cu-Sn	As cast	Lead
VL3 blade	96.60	2.45	0.95	0	0	Cu	As cast	--
VL3	96.41	1.64	1.95	0	0	Cu	Extensive annealing	--
VL4	97.93	1.48	0.60	0	0	Cu	As cast	copper sulfide
VL5 blade	93.21	2.69	4.10	0	0	Cu	As cast	--
VL5	94.15	1.74	4.11	0	0	Cu	As cast	--
VL9	98.13	0.00	0.95	0.92	0	Cu	As cast	copper sulfide & lead

Trace elements

ICP-AES was used to identify the following elements: Cu, Sn, Pb, Zn, Fe, Co, Ni, As, Sb, Se, Te, Ag, Au, Bi, Sr, and Mn. Table 3 presents the findings.

Trace elements, also known as fingerprint elements, are important indicators of copper's raw material source. This research was conducted using the qualitative analysis method proposed by Mark Pollard (Pollard et al. 2018), which focused on four trace elements: arsenic (As), antimony (Sb), nickel (Ni), and silver (Ag). There are 16 possible 'Copper Groups' based on all possible combinations of presence/absence (cut-off value = 0.1%) for the four elements. Bronze mineral characteristics can be described as specific combinations of multiple 'Copper Groups' in terms of assemblage. This method has been successfully applied in China (Hsu et al. 2021, Li et al. 2021, Li et al. 2022).

According to the results of the above method, the pattern for this particular assemblage was of low complexity, with $CG1(Cu) = 58.8\%$ and $CG2(Cu + As) = 41.2\%$. CG2 includes TK7, VL9 (MLA); TK3, TK5 (axe); VL1 (drum); and VL2 (tubular bangle).

Table3 Trace element contents of the samples ($\mu\text{g/g}$)

Lab Num	CG	Fe	Co	Ni	As	Sb	Se	Te	Ag	Bi	Mn	Zn	Cd
TK1	1	0	0	70	306	104	133	82	54	244	6574	105	0
TK2	1	708	0	221	723	152	164	240	61	397	8118	57	0
TK3	2	0	0	105	1226	206	192	198	27	402	7070	102	0
TK4	1	0	0	45	251	126	207	185	3	431	8060	62	0
TK5	2	176	0	49	1044	142	237	227	4	451	9457	66	0
TK6	1	0	0	45	331	146	243	213	13	487	10413	71	1
TK7	2	0	0	125	1908	271	155	45	184	265	8388	101	0
TK8	1	0	0	23	49	47	124	96	0	242	4699	66	0
VL1-bottom	2	2496	52	213	15489	536	157	270	369	657	9392	116	0
VL1-top	2	1467	30	123	8523	322	96	118	205	379	2652	61	0
VL2	2	10	0	65	1658	82	194	108	10	501	4961	70	0
VL3	1	0	0	32	143	67	274	230	0	586	8233	32	0
VL3-ren	1	0	0	17	42	18	147	87	0	265	4894	50	0
VL4	1	0	0	24	98	29	144	328	0	559	4438	49	0
VL5	1	0	0	22	83	32	104	121	0	301	3641	70	0
VL5-ren	1	0	0	41	189	37	141	267	0	558	10579	129	0
VL9	2	0	0	66	1273	287	162	114	15	500	5143	86	0

Lead isotopes

All results are in the range of $^{207}\text{Pb}/^{206}\text{Pb} > 0.80$ and $^{206}\text{Pb}/^{204}\text{Pb} 20.0$ (Table 4), which do not belong to the highly radiogenic lead category but to the common lead. $^{207}\text{Pb}/^{206}\text{Pb}$ varied between 0.848 and 0.860, with 1.41% variation, whereas $^{208}\text{Pb}/^{206}\text{Pb}$ varied between 2.113 and 2.095, with 0.90% variation. The rate of change is slightly higher than the rate of common lead (0.3%–1.0%) for the same ore body, mineral deposit, or ore province (Isotope Geology Research Unit 1979). There are two explanations for the higher rate of data variation: One is that the lead isotopes in the Vilabouly deposit are relatively stable, and the test results prove that these copper wares contain ore materials from a source other than this mine. The other explanation is that the distribution of lead isotopes in the Vilabouly deposit had significantly variability, possibly as a result of the mixing of two different common leads (Doe 1970).

Because VL1 (drum) contains a significant amount of lead, the lead isotope ratios indicate the source of lead. The lead isotope ratios of the remaining samples refer to the copper source because the amount of lead introduced in cassiterite is much less than the copper introduced (Killick et al. 2020).

Table 4 Lead isotope ratios of the samples

Lab Num	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$
TK1	38.539	15.633	18.308	2.1051	0.8539
TK2	38.198	15.519	18.175	2.1021	0.8540
TK3	38.396	15.594	18.313	2.0950	0.8499
TK4	37.956	15.395	18.044	2.1031	0.8530
TK5	38.598	15.659	18.367	2.1011	0.8523
TK6	37.842	15.353	18.064	2.0946	0.8497
TK7	38.560	15.698	18.254	2.1134	0.8603
TK8	38.618	15.714	18.286	2.1119	0.8593
VL1-foot	38.870	15.703	18.495	2.1025	0.8493
VL1- tympanum	38.846	15.705	18.492	2.1023	0.8494

VL2	38.852	15.699	18.498	2.1020	0.8493
VL3	38.103	15.452	18.137	2.1009	0.8519
VL3-blade	37.757	15.324	17.947	2.1038	0.8539
VL4	37.857	15.315	18.057	2.0972	0.8483
VL5	37.872	15.378	18.037	2.1001	0.8527
VL5-blade	38.227	15.521	18.188	2.1015	0.8531
VL9	38.635	15.697	18.293	2.1092	0.8575

Preliminary analysis

This section's test data (hereinafter referred to as TKVL) include major elements, trace elements, and lead isotope ratios. This section uses the aforementioned data to discuss raw material sources for copper wares. We plotted a scatter plot of lead isotope ratios and used 'Copper Groups' and principal component analysis (PCA) of trace elements to comprehensively analyse these data; the results are shown in Fig. 5.

According to the previous section, copper wares belonging to CG1 and CG2 in the collection account for 58.8% and 41.2%, respectively. Copper wares classified as CG2 contain arsenic in its alloys that exceeds a manually defined cut-off. However, there are also differences between copper wares classified as CG2. The arsenic content of the VL1 (drum) alloy was significantly higher than that of the other copper wares in the same group. The PCA results provide comparable evidence. Fig. 5a shows that VL1 (drum) was clearly distinct from the rest of the collected data.

The scatter plot of $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ (Fig 5-b) divides the data into four categories. Category A contains the majority of copper halberds and copper axes; Category B contains TK3 (axe) and TK6 (axe); Category C contains drums: VL1 (drum) and VL2 (tubular bangle) (although lead isotope ratios are similar, VL1 should indicate the source of lead); Category D contains TK7 (MLA), VL9 (MLA), and TK8

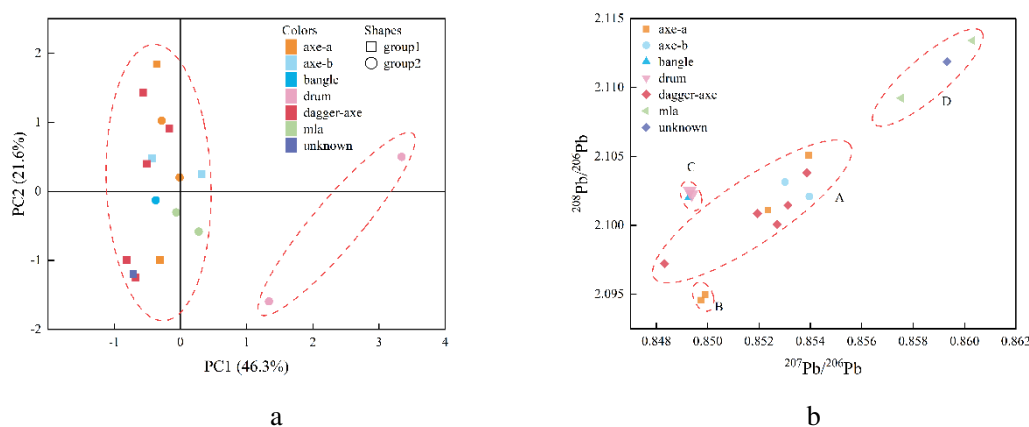


Fig. 5 Graphical representation of the outcomes of principal component analysis and lead isotope ratio scatter plot

5a PCA result; **5b** lead isotope ratio scatter plot

(unknown). The characteristics of the alloys in different copperware categories may be inconsistent.

In conclusion, bronze drums are clearly different in terms of materials from other samples; aside from bronze drums, MLA ore characteristics may differ from axes and dagger axes; additionally, some copper wares contain arsenic. Because of the small sample size, this section yields only limited information. In the following section, relevant data from additional sites are presented.

Discussion

This section collects previously published metallurgical data from Laos (Pryce et al. 2014, Cadet et al. 2019, Cadet et al. 2022), Thailand (Yasuda et al. 2013, Pryce et al. 2014, Pryce et al. 2018), Cambodia (Yasuda et al. 2013, Pryce et al. 2014, Pryce et al. 2017), Vietnam (Pryce et al. 2014, Pryce et al. 2021), Myanmar (Pryce et al. 2014, Pryce et al. 2018), and other countries, and compares the data to data measured in the previous section (hereafter referred to as TKVL). This section investigates the exchange of raw materials between the central region of Laos and surrounding sites from the perspective of Vilabouly and the medium-range interaction zone where it is located.

Considering the inconsistency between the methods used to determine trace elements in the above research and the accuracy of the instruments used in this study, the comparative study of the raw material provenance analysis of bronzeware in this section is mainly based on lead isotope analysis. Owing to their unique characteristics, VL1 (drum) and VL2 (tubular bangle) are discussed separately in Section 5.2, and are not included in the comparative analysis in Section 5.1.

Internal interaction

The following Vilabouly sites have well-preserved wooden mine shafts: Kha Nong A2, Thengkham South C pit & Puen Baolo, Thengkham South D pit, Thengkham East, Thengkham North, and Pha Bing A & B pits. Based on these discoveries, scholars presumed that these were Vilabouly's main ancient mining areas. Two sites have clear evidence of ore processing: The cultural layer of Puen Baolo revealed fragments of ore processing, along with slag and crucibles around the majority of the ash pits (Chang 2013, Chang 2013, Tucci et al. 2014), and evidence related to copper smelting was discovered at Thong Na Nguak in the form of slag, crucibles, ore, and final products (Chang 2009, Tucci et al. 2014).

In addition to archaeological materials, the scatter plot of lead isotope ratios (Fig. 6) demonstrates that Puen Baolo was an ore-processing site. Fig. 6b shows that nearly half of the samples from Puen Baolo were distributed in area A ($0.853 < {}^{207}\text{Pb}/{}^{206}\text{Pb} < 0.856$, $2.102 < {}^{208}\text{Pb}/{}^{206}\text{Pb} < 2.110$). Within this small area, 70% of the data pertained to the intermediate products and slag, which have relatively stable lead isotope characteristics. The close relationship between the buried copperware and bowtie ingots in area A may indicate that at Puen Baolo, craftsmen processed ore into intermediate products and used them to produce their final products. In the lead isotope ratio scatter plot, for example, the data points of PBL/14, PBL/33, PBL/36, and PBL/37 (bowtie ingots) almost coincided with those of PBL/24 (axe) (Fig. 6). Similarly, final products

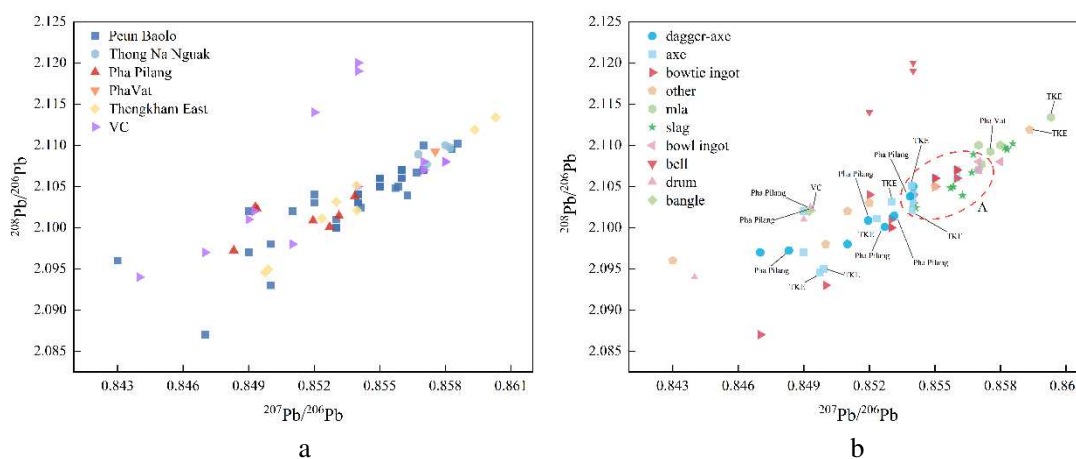


Fig. 6 Lead isotope scatter plot for copper wares excavated at Vilabouly
6a Classified by sites; **6b** Classified by types

such as PBL/25 (axe), PBL/26 (axe), PBL/30 (axe), and PBL/31 (spear) have very similar characteristics to those of the ingot materials. Although the strata at Puen Baolo have been disturbed and a clear stratigraphic sequence cannot be established, most copperware and bowtie ingots are associated with deeper strata, whereas iron axes associated with MLA are frequently found in the upper layers (Cadet et al. 2019). Copper smelting activities at Puen Baolo may have continued from the Bronze Age to the Iron Age, based on the approximate stratigraphic relationship of samples in area A of Fig. 6b.

Based on a method that uses the relative distance of data points on the lead isotope ratio scatter plot to reflect the degree of similarity of ore characteristics, we can establish a connection between the intermediate and final products and investigate the metallurgical activities involved in Puen Baolo. As shown in Table 1, TKVL includes both final and intermediate products from multiple sites, which can be utilized to

investigate diverse Vilabouly metallurgical activities using similar methods.

First, we looked for a connection between the intermediate products of the assemblage and Vilabouly's final products. In TKVL, there are two intermediate products, VL9 and TK7 (MLA), neither of which is within the range of Area A (Fig. 6b). None of the final products data is highly similar or even coincides with these two points. VL9 fell within the range of Puen Baolo and Thong Na Nguak in the lead isotope ratio scatterplot (Fig. 6a). Currently, the academic community lacks sufficient lead isotope data of intermediate products to determine whether there are subtle differences in the copper materials of Thong Na Nguak and Puen Baolo (Cadet et al. 2022); thus, proving the specific source of VL9's raw material is not sufficient. Regardless of the specific source, the discovery of VL9 undoubtedly provides evidence of intermediate product exchange in Vilabouly, as VL9 was discovered in Pha Vat, and there is currently no public archaeological data to prove that it was an ancient mining area or major ore processing site. TK7 was a singularity far from the majority of the data and not within the range of intermediate and final products at any site, proving that locating the exact source of the raw material is currently impossible.

Second, we established a link between the final products in the assemblage and the intermediate products in Vilabouly. In TKVL, the lead isotope ratios of TK2 (axe) are similar to those of PBL/22 (axe), PBL/38 (axe), and PBL/15 (bowtie ingot); TK3 and TK6 (axe) are similar to those of PBL/10 (bowtie ingot); and VL5 (dagger axe) is similar to those of PBL/20 and 21 (bowtie ingot) (Fig 6-b). These could be signs of a product exchange between Thengkham East and Puen Baolo, with the possibility of intermediate and final product exchanges.

Vilabouly appeared to have two distinct types of dagger axes. PBL/8 underwent heat forging and had a tin content greater than 10%. In addition to PBL/8, the collected dagger axes (VC/2 or VL4) had very low average tin content in the comparative material (only 3.10%), which is fairly similar to the tin content level of the dagger axes in TKVL. The remaining dagger axes were made by casting and had a relatively simpler shape and decoration style than PBL/8 (Cadet et al. 2019). The distinct process and alloy ratio demonstrate that PBL/8's usage and application scenarios differ from those of the other daggered axes.

In summary, Vilabouly may have had an internal metallurgical technology system. Several major mining areas had similar technical characteristics, all employing multiple narrow and deep wooden shafts for mining, and ore selection and final product

production were relatively unified (Tucci et al. 2014). The lead isotope analysis indicates that a product exchange within Vilabouly may have existed. For example, consumer site Pha Vat did not engage in mining but chose to import intermediate products (ingots or MLA) from Puen Baolo or Thong Na Nguak. Furthermore, the final products excavated from Thengkham East share similar raw material characteristics with the intermediate products excavated from Puen Baolo, and casting technology was widely used for decorative items and intermediate products throughout Vilabouly. Craftsmen in Puen Baolo and Thengkham East intentionally used alloys with improved mechanical properties for weapons and tools with practical functions. Furthermore, Baolo began using heat forging technology to improve the functionality of alloys.

External interaction

Section 5.1 discussed the origins of the raw materials used in the production of copper wares in Vilabouly. This section investigates the interactions between central Laos and its surrounding areas. Lead isotope analysis points to the source of lead (Fig. 7a) in some of the copper goods in the comparison material because they contain a certain amount of lead ($w(\text{Pb})2\%$), while the remaining data reveal the source of copper (Fig. 7b).

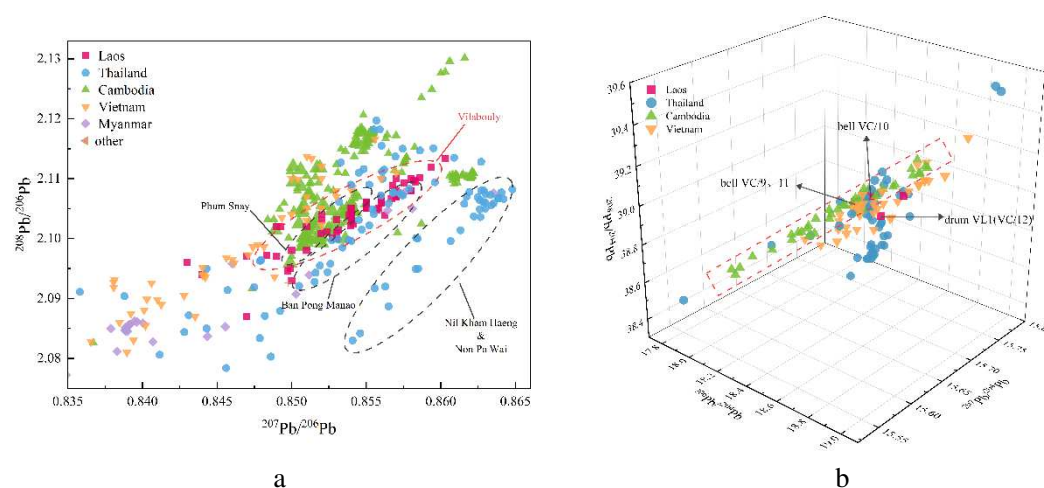


Fig. 7 Scatter plot of lead isotope ratios (Laos and neighboring countries)

7a Source of copper (partial enlarged version): **7b** Source of lead

Non Pa Wai, as an occupation in the early Bronze Age in Thailand, excavated copper wares that represented the raw material characteristics of the early Khao Wong Prachan Valley (Fig. 7a). The majority of the raw materials from Ban Non Wat's bronze Age 1–3 periods are consistent with Non Pa Wai's materials, whereas copper ware with Vilabouly characteristics appeared during the Iron Age. Simultaneously, the distribution of copper materials in Vilabouly (abandoned outliers), some data from Phum Snay in northwestern Cambodia, and Ban Pong Manao in central Thailand were roughly consistent, indicating that the raw materials in the three sites may have had similar features. Phum Snay and Ban Pong Manao are roughly contemporaneous and date back to the Iron Age (200 BC to 500 AD). Phum Snay's raw material supply was complicated; Vilabouly could have been a source of copper, and the Song Toh lead mine in Thailand could have been a source of lead (Hirao et al. 2013). Based on this, can Cambodia be included in the medium-range interaction zone that Pryce et al. (2022) defines as encompassing central Thailand, northeast Thailand, and central Laos? The evidence presented above demonstrates that Vilabouly replaced the Khao Wong Prachan Valley

as the primary source of copper for the Indo-China Peninsula during the early Iron Age (420 BC to 100 AD) (Higham et al. 2021).

Two artefacts in TKVL have unique qualities: VL1 (drum) and VL2 (tubular bangle). Drums and tubular bangles are widely used in China's Yunnan–Guizhou Plateau and in many Southeast Asian countries. Drums are crucial tools for sacrificial rituals and symbols of wealth and authority (Wan et al. 2018), with the potential to exchange and disperse wealth and power. Previous research on Vilabouly Bronze and Iron Age archaeology hardly mentioned tubular bangles. Therefore, these two types of copper wares require special attention in comparative studies.

Vilabouly's drums contain two types of alloys: lead-tin bronze (VL1 is the same as LA/VC/12) and tin bronze (LA/VC/13). The VL1 lead isotope ratio data were similar to those of the three Dong Son-style drums VN/DS/5, 14, and 17 (Fig. 7b), and VC/13 was similar to VN/DT/5. Current studies indicate that there was little exchange of metallurgical raw materials between Vilabouly and Vietnam, and whether the similarity in the lead characteristics of the two drums implies the possibility of circulation of the final products between the two sites should be investigated further. Fig. 7b shows that the data in Cambodia have a strong linear correlation in area B, and the three bells of Vilabouly are all located in this area. These bells have different sources of lead from VL1, but may have the same source as Phum Snay.

Tubular bangles have been discovered in mainland Southeast Asia and Yunnan, primarily in the regions of Dadingshan in Yunnan and Dong Son in northern Vietnam (Yang 2020). Similar discoveries were made in early iron-age burials in Thailand's Ban Non Wat and Ban Na Di (Higham et al. 2012, Higham et al. 2021). The bangles were similar in shape, with minor differences in detail. Tubular bangles are typically decorated with strings, milk nails, or braided patterns. VL2 is very similar to the bronze bangle excavated in the early Iron Age burial at Ban Non Wat, and both are decorated with hollow spherical ornaments at the end; this type of tubular bangle with spherical ornaments seems to have appeared only in Laos and Thailand (Fig. 8). The lead isotope ratio scatter plot (Fig. 7a) shows that the lead isotope ratio of VL2 is within the range of Vilabouly. It could have been cast from local copper mines in Vilabouly, but direct evidence is needed for confirmation. It is generally believed that tubular bangles in Thailand are cast by the lost-wax method, for which Ban Na Di has revealed clear evidence (Higham et al. 2021). However, no traces of lost-wax casting bangles have yet been discovered in Vilabouly. The alloy ratios of the tubular bangles varied by region.



Fig. 8 Tubular bangles

Left (excavated from Laos), Right (excavated from Thailand)

The bangles unearthed by Ban Na Di were mostly copper-lead-tin ternary alloys, whereas the VL2 was a copper-tin binary alloy. Thus, this particular type of bangle may be a common decorative style. VL2 has no context, and it is impossible to confirm whether there were burial customs for tubular bangles similar to those found in infants in Ban Non-Wat and Dong Son. Researchers have speculated that this type of bangle is representative of a tradition of Vietnam's Dong Son culture and that tubular copper bracelets in Thailand's upper reaches of the Mekong River and Yunnan's Dadian Mountains were influenced by northern Vietnam (Yang 2020). Pryce et al. (2022) proposed that in the late second millennium BC, the technological diffusion path may have spread from north to south along the Red River Delta coast. According to this theory, the decorative style of the tubular bangles may have spread from the upper reaches of the Mekong River to central Laos.

Conclusion

This study used a variety of techniques to expound on metallurgical industry issues in Vilabouly, central Laos, using excavated artefacts in 2018 and the collection of a local museum as the research objects. Although the technical system within the mining area is relatively unified, the lead isotope data between the intermediate products produced by each mining area may vary slightly because of the raw materials. The MLA with suspected PBL/TNN material characteristics was discovered in the Pha Vat pit, which is located far from mining areas. This can be considered compelling evidence of Vilabouly's internal exchange of intermediate products. Future data may help us understand the social organisation and interaction patterns of the metallurgical industry in Vilabouly.

A comparative study with data from other Bronze Age and Iron Age sites in the Indo-China Peninsula confirms the current dominant scholarly understanding of Vilabouly in the Southeast Asian metallurgical exchange network. Newly introduced sites from the Early Iron Age in northwestern Cambodia and central Thailand with similar lead isotope ratios suggest that these sites may also have been copper consumption sites in Vilabouly. The tubular copper bracelets found in Vilabouly are morphologically similar to those found in central Thailand, and the different alloy types may indicate an exchange of copper bracelet styles between the two sites. The main weakness of this study was the paucity of diversity in the sample types. More information on metallurgical relics (slags, crucibles, and ores) would help us develop a more accurate picture of the metallurgical industry in central Laos.

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Author contribution

Li HaiChao conceived and directed this study. Yang ZiShu performed the SEM-EDS, OM, MC-ICP-MS analyses, and wrote preliminary versions of the paper. All authors read and approved the final manuscript.

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