# HEMICAL COMPOSITION AND LEAD ISOTOPY OF COPPER AND BRONZE FROM NURAGIC SARDINIA

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Abstract: We present data on the chemical and lead isotope composition of copper and bronze objects from Nuragic Sardinia. The sample suite comprises, inter alia, objects from the hoard finds at Arzachena (21 objects), Bonnanaro (10), Ittireddu (34), and Pattada (20), all in northern Sardinia. With one exception, all ingot fragments (49) consist of unalloyed copper; the exception comes from Ittireddu and contains 11 per cent tin. In contradistinction, all implements (21) are made from standard bronze with a mean tin content of 10.8 per cent. A dozen sword fragments from the Arzachena hoard, all of fairly uniform small size, are pieces of a large number of different swords. The low tin content of only about 1 per cent would have made for poor weapons, confirming the archaeological identification of the fragments as pieces of votive swords. Scrap metal from Arzachena is remarkable for its wide range of trace element contents and lead isotope abundance ratios. It is dissimilar to all other metal samples investigated, possibly representing metal from local smelting experiments using a variety of different copper ores. Lead isotope data and trace element patterns, alone or in conjunction, do not allow us to tell oxhide ingots from plano-convex (bun) ingots. Most ingot fragments have a lead isotope signature similar to those of Cypriot copper ores but there are also a number of ingots whose lead isotope fingerprints are fully compatible with them being local products. Of the bronzes, none has lead with an isotopic composition characteristic of copper ingots from Cyprus. All contain local lead, suggesting the bronze implements were manufactured locally. Isotopically-fitting lead is found in copper and lead ore deposits from the Iglesiente-Sulcis district in south-west Sardinia and from Funtana Raminosa in central Sardinia.

Keywords: lead isotopy, Nuragic artifacts, oxhide ingots, provenance studies, Sardinian Bronze Age



#### Introduction

Sardinia is rich in natural resources. Moreover, it is the second largest island in the Mediterranean, with a number of safe natural harbors to give shelter to ships and a strategic geographic position to make it well suited to partake in Mediterranean exchange networks. Indeed, the earliest evidence for such trade goes back to the Neolithic of the sixth millennium BC when Sardinian obsidian was traded to Corsica, central Italy and southern France (see Tykot 1992, 1996, 1997; Tykot and Ammerman 1997). Later, from the third millennium onward, a connection with the Aegean and the Near East is demonstrated by objects of art and religion such as the 'Cycladic idols', the 'steatite pans' and a monument such as the Monte d'Accoddi (Sassari) 'ziggurat'. Influences and exchange with the Iberian Peninsula to the west are documented by pottery from the Bell Beaker culture. In the midst of all these influences, Sardinia, nevertheless, managed to keep its cultural identity, as is abundantly illustrated by many unique and splendid products from the Neolithic and Chalcolithic.

Metal objects are rather rare during the earliest periods. Generally speaking, in Sardinia the advent of Chalcolithic metallurgy appears to have been delayed in comparison with Anatolia and the Balkans. In the Balkans, the middle Chalcolithic with its flourishing copper industry has been radiocarbon-dated to the end of the fifth millennium BC (Görsdorf and Bojadziev 1997), while, in western Anatolia, late Chalcolithic copper grave offerings at sites such as Ilipinar date to the first half of the fourth millennium BC (Begemann et al. 1994). In Sardinia, on the other hand, the earliest metal derives only from the Ozieri culture, dated to the end of the fourth to the early third millennium BC. Interestingly, some of these early metal objects are made from silver and lead (Lo Schiavo 1989). Since, during this period, the use of these two metals was much more advanced in the eastern Mediterranean, this suggests that metallurgy might have come to Sardinia from the east.

With the rise of the Nuragic culture, metal artifacts become more abundant. This coincides with numerous finds, especially of ceramics, that indicate contact with the Aegean and other regions of the Mediterranean. In fact, imported and locally imitated pottery of Aegean types form the essential basis for Bronze Age chronology on Sardinia (Jones and Vagnetti 1991). Other materials indicating long-distance trade include ivory, faience and glass beads, as well as cylinder seals. It is generally accepted that there is a remarkable interconnection between Cyprus, the Aegean and the central Mediterranean in this period, especially in the thirteenth and twelfth centuries BC (Catling 1980; Vagnetti and Lo Schiavo 1989), impressively demonstrated by the occurrence on Sardinia of sophisticated prestige objects such as Cypriot bronze tripod stands, vessels, and figurines (Lo Schiavo et al. 1985; Lo Schiavo 1998a).

More controversial is the provenance of the so-called copper oxhide ingots. Based on their similarity in shape and their 'inscriptions', Pigorini (1904) believed them to be imports from the Aegean. Buchholz (1959) more specifically attributed them to Cyprus. Chemical analyses were interpreted to cast some doubt on this hypothesis,



Location of archaeological sites investigated.

however, and Balmuth and Tylecote (1976) concluded that the four oxhide ingots from Sardinia which they investigated did not come from Cyprus. Zwicker et al. (1980) compared the composition of slag inclusions in copper ingots from Sardinia with those in copper smelted in the laboratory from ores from Funtana Raminosa and suggested that oxhide ingots were produced on Sardinia itself. This was readily accepted by Buchholz (1980) and local production of oxhide ingots was also considered possible by Lo Schiavo et al. (1985). Finally, to close the circle, lead isotope analyses have been argued again to indicate a Cypriotic origin (Gale and Stos-Gale 1987; Gale 1991; Stos-Gale et al. 1997) although such a conclusion has been contested by Budd et al. (1995) and Knapp (2000).

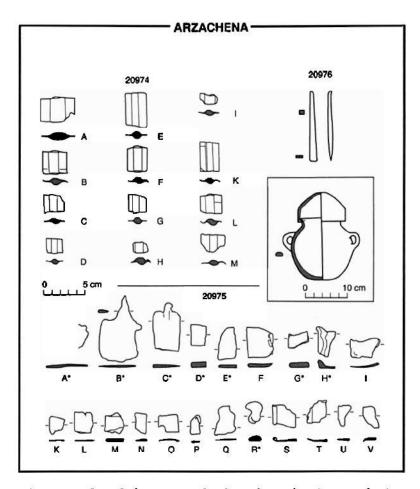
We made use of new finds of Nuragic metal hoards on Sardinia to broaden the database under discussion with the aim of shedding new light on the provenance of the oxhide ingots. To this end, we have analyzed some 40 ingot samples and another 45 artifacts for both trace element concentrations and lead isotopic composi-

tion. Contrary to widespread belief (e.g. Chippindale 1994), the information to be gained from these two material features is not redundant. Rather, the combination of the two has turned out to be a much more powerful tool than each by itself for identifying ore sources, smelting processes and metal workshops (Pernicka et al. 1997). Moreover, since provenancing of metal must, of necessity, rely on a comprehensive database of ores, we have also investigated the lead isotope composition of a number of copper ores from 20 Sardinian occurrences, some ancient copper slags, and also some lead ores. Although the number of samples available from each ore deposit is not large enough to cover the full range of potential variations, the data do serve as a valuable basis for an overview as well as for future work.

#### ARCHAEOLOGICAL SITES AND FIND DESCRIPTION

Arzachena (Sassari), Nuraghe Albucciu hoard

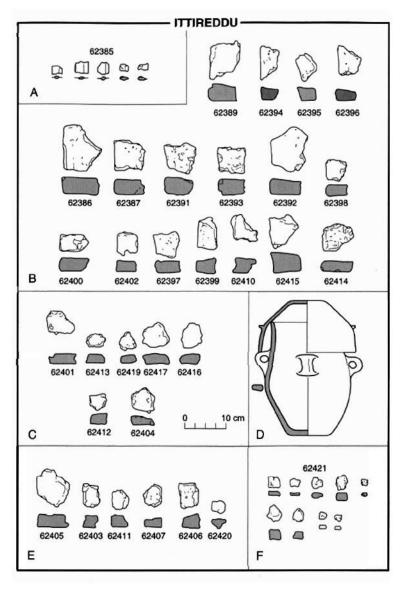
In 1962, during the archaeological excavation of the nuraghe Albucciu (Arzachena, Sassari) an impasto pot, covered with a bowl, containing fragments of bronze objects and scrap metal was found under the pavement of a terrace (Lo Schiavo 1982,



**Figure 2.** Sword fragments, chisel, and nondescript metal pieces (no. 20975) from the Arzachena, Nuraghe Albucciu hoard. The eight analyzed pieces from no. 20975 are marked with an asterisk (A, B, C, D, E, G, H, R).

1990b). Chronologically, the hoard was first thought to be from the advanced Iron Age; now, further studies have dated the complex to about the end of the middle Bronze Age and the beginning of the late Bronze Age (MBA 3/LBA) (Ferrarese Ceruti 1962; Lo Schiavo and Vagnetti 1980; Campus and Leonelli 1999).

Altogether the pot contained 1 small chisel, 6 ingot fragments, 12 fragments of votive swords and 21 small nondescript pieces of scrap metal, as it may have been collected from a bronze workshop to be remelted together with the rest of the hoard (Fig. 2). The six ingot fragments can be attributed to the oxhide type because of their straight sides and constant thickness. (No data reported here. Results of chemical analyses by atomic absorption spectrometry in Maddin and Merkel 1990.) Judged from the cross section of the central rib, which is flattened, rounded and more or less irregular, the 12 votive sword fragments are pieces of at least three or four different weapons. The small chisel with a square cross-section and flattened blade appears curved and damaged at the top.



**Figure 3.** Metal fragments from the second Ittireddu, Nuraghe Funtana hoard. A: sword fragments. The ingot fragments are arranged according to the type of their parent ingots. B: oxhide ingots; C: plano-convex (bun) ingots; E: questionable; F: indeterminate metal pieces.

Ittireddu (Sassari), Nuraghe Funtana, second hoard

A first group of eight fragments of oxhide ingots, most probably a hoard, was brought light to during the excavation of the courtyard at the foot of the tower (Galli 1984). All objects have been analyzed for their chemical composiby Maddin Merkel (1990), two of them for the isotopic composition of their lead by Stos-Gale et al. (1997).

A second hoard (total weight 19.7 kg) was found in the corridor of the main tower. It consisted of 35 fragments of oxhide and plano-convex ingots (plus nine smaller fragments), together with five fragments of votive swords (Fig. 3), collected inside a fourhandled pot (Fig. 3, D), covered with a bowl (Galli 1985/1989; Lo Schiavo 1990a; Campus and Leonelli 1999). Of the ingot fragments, 19 are without doubt fragments of oxhide ingots and 10 of plano-

convex ingots; the remaining six and the nine smaller fragments cannot be assigned with certainty to any particular ingot shape (analyses in Maddin and Merkel 1990). Although tiny and in poor condition, the five metal fragments can unmistakably be attributed to votive swords. The chronology of this typical Nuragic weapon has been discussed by Lo Schiavo (1994).

Metalworking on the site is attested by the discovery of a melting pot containing traces of vitrified copper slag. The pot has a short handle with a 5 cm deep indentation, made by the tip of a spearhead or a dagger. Possibly, the indentation was meant for such an implement to be inserted in order to lift the pot (Galli 1985/1989: Fig. 11).

### Pattada (Sassari), Sedda Ottinnera hoard

This hoard was discovered by a workman on 5 March 1997 (Lo Schiavo 1998a, 1998b. 1999). It consists of 23 pieces (Fig. 4). Its main importance lies in its typological mixture, since it is the first time that oxhide ingot fragments have been found associated with implements, weapons, and vessels. ingots together weigh 7.73 kg, the other pieces altogether 5.69 kg. The seven fragments of oxhide ingots are recognizable as such from having a straight side and raised edges.2

The tools and weapons are not only characteristic of the Nuragic late Bronze Age but they in themselves represent the typical association of the Monte Acuto region,

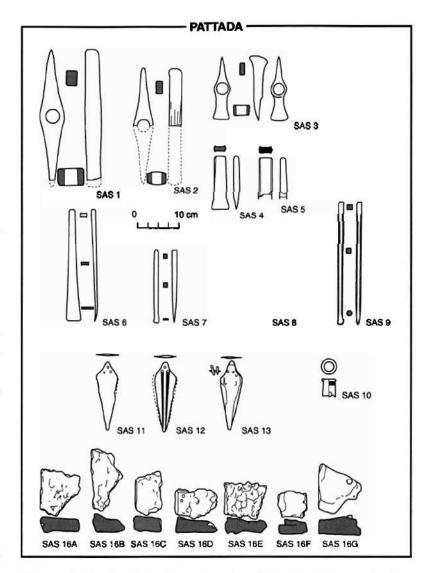


Figure 4. Analyzed implements and oxhide ingot fragments from Pattada, Sedda Ottinera hoard.

as already documented by the well-known Chilivani hoard (Lo Schiavo 1988a, 1990a). The typology of the objects is part Cypriot late Bronze Age, but mostly Nuragic late Bronze Age (Lo Schiavo et al. 1985; Ferrarese Ceruti and Lo Schiavo 1992; Lo Schiavo 1980, 1990b, 1992, 1998a; Lo Schiavo and Usai 1996) and indicates a date about the eleventh century BC.

#### Bonnanaro (Sassari), Funtana Janna hoard

In 1951, a large hoard was found by agricultural workers, but it was robbed soon after the discovery. Only a few objects, namely four ingots, four more or less complete axes and the blades of two more, are preserved in the Archaeological Museum of Sassari (Lo Schiavo 1981: Fig. 297, 1982: Fig. 5, 1986: Fig. 43; Lo Schiavo et al. 1987:179–180).

Worth noting are the four ingots (Fig. 5). One is a large ingot of truncated conical shape (no. 10708), the second has a shape between truncated-conical and conical with a rounded bottom (no. 10709), the third is a large plano-convex ingot, slightly

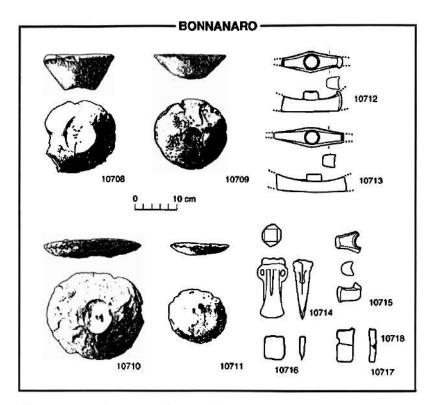


Figure 5. Implements and ingots from Bonnanaro, Funtana Janna. Note the unusual truncated-cone shaped ingots nos 10708 and 10709.

flattened on one side, and the fourth is a typical plano-convex ingot. Only recently, two more ingots of the same truncated conical shape, but of distinctly lesser dimensions, have been found in the hoards of Sant' Imbenia, Alghero (Sassari) (Bafico et al. 1995: Fig. 3, 2).

The three double axes are almost identical in type and dimension but they are in very different states of preservation. All of them are broken and their outside appearance shows clear evidence of having been used extensively. One piece is only a fragment (no. 10715). The type of double axe with vertical and converging blades, round shaft hole and collared socket protruding up on one side is a local evolution from the Cypriot shape of the massive double axes, dated to late Cypriot III (= late Helladic IIIC) (Lo Schiavo et al. 1985: Fig. 8, 3).

The double-loop socketed axe (no. 10714) is of a peculiar Iberian type – '42A West Portugal' with three horizontal decorative ribs along the socket and one vertical rib along the blade. The name derives from the westernmost occurrence (Monteagudo 1977, nos 1727, 1728), dated to the second half of the late Bronze Age because of the presence of three examples in the Monte Sa Idda hoard (Taramelli 1921, nos 29, 30, 33).

One massive axe-blade (no. 10716) and possibly a second one, broken into two pieces (nos 10717 and 10718), can be related to flat axes or to heavy chisels known from other hoards in the south of the island, like Monte Sa Idda, Decimoputzu (Cagliari) (Taramelli 1921: Figs 8–11). Many steatite moulds are known for the production of these tools (Lo Schiavo 1978:11, no. 4, Plate XL).

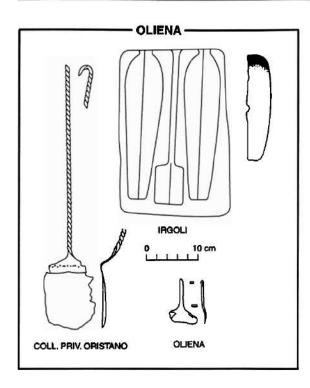


Figure 6. Charcoal shovel fragment from Oliena, Sa Sedda 'e Sos Carros. Identification of the object is essentially based on the similarity with a more complete shovel from a private collection (left) and a steatite mould for a shovel from Irgoli (top, centre).

Oliena (Nuoro), Sa Sedda 'e Sos Carros. Charcoal shovel fragment.

Charcoal shovels with long twisted handles, ending in a hooked terminal shaped as a bird's head, are a well known tool in Cyprus within the twelfth and the beginning of the eleventh century BC. Close parallels are known from Megiddo and Amathus, associated with a group of vessels from the Cypro-Geometric I period (1050-950 BC) and with an obelos of a distinctive Atlantic type, a fragment of which has been found in the Monte Sa Idda hoard in southern Sardinia (Karageorghis and Lo Schiavo 1989). In Sardinia, the 'best-preserved example, identical to the Cypriot ones, is in a private collection in Oristano (Lo Schiavo et al. 1985:27, Fig. 10,2).

Although many of the characteristic features are missing from the Oliena sample (Fig. 6), we are confident that

this implement is such a charcoal shovel. It consists of a piece of twisted handle where the blade has been broken off. It was found among the bronzes of the Nuragic complex of Sedda 'e Sos Carros, which date, in general, from the end of the late Bronze Age to the beginning of the early Iron Age, with sporadic later elements. A steatite mould of a shovel of similar shape is known from Irgoli (Lo Schiavo et al. 1985:27, Fig. 10,3) which indicates local production imitating the Cypriot model.

## CHEMICAL COMPOSITION AND LEAD ISOTOPY<sup>3</sup>

#### Ingots

Generally speaking, all ingots consist of fairly pure copper. Except for one instance with 11 per cent tin, 4 cases with more than 1 per cent iron, and a few with around 1 per cent of arsenic, all ingots are at least 99 per cent copper. Moreover, the trace element contents of all ingots are also remarkably constant (Table 1). Where results for arsenic (As), iron (Fe), nickel (Ni), silver (Ag), zinc (Zn), lead (Pb) and, for Ittireddu, also cobalt (Co), have already been reported by Maddin and Merkel (1990), the agreement with our data is satisfactory in most cases and especially good for nickel and cobalt. For low silver contents, there are 10 samples listed by Maddin and Merkel that contain 0.000 per cent silver, while we find between 28 and 127 ppm. Apparently, an entry of 0.000 per cent by Maddin and Merkel does not imply the concentration to be below 0.001 per cent, which would correspond to 10 ppm.<sup>4</sup>

Again in agreement with Maddin and Merkel, but contra Garagnani and Martini (1996), we find no significant differences between oxhide ingots and plano-convex (bun) ingots in the trace element abundances. This also holds true for elements like iron and arsenic, which, upon remelting and recasting of metal without taking special precautions to avoid it, will preferentially be oxidized so that they are depleted in secondary melts as compared to primary ones. Thus, the data do not allow us to draw conclusions with regard to the question whether one of the two types of ingot might have been the precursor of the other and have served as the starting material from which the other was derived by remelting – and, if so, which ingot type it was. An implication of this result is that chemical data do not help to identify the type of parent ingot from which those fragments of ambiguous shape derive.

In spite of the general chemical homogeneity of the ingots, there are a few notable exceptions. Undoubtedly, the most exciting one is a fragment from Ittireddu (no. 62405) because it contains 11 per cent tin. Indeed, if this were a piece from an ingot it would be one of the extremely rare cases of bronze alloy ingots ever having come to light in Sardinia.5 Maddin and Merkel (1990), who had already identified the fragment as a tin bronze, argued that it cannot be a primary ingot. They suggested it was likely to be a secondary alloy ingot to be processed into artifacts or to be remelted and mixed with more tin. The latter seemed necessary because the tin content, in duplicate analyses, of 6.82 per cent and 2.02 per cent, respectively, was lower than the standard in contemporary Nuragic bronzes of around 11 per cent tin (see later). According to our analysis, since the tin content is 11.0 per cent, no addition of tin is required. Actually, according to the chemical composition as well as the isotopic composition of its lead, this metal might well be a ready-for-use ingot or a spilled left-over from casting artifacts. The lead content is about 10 times higher than in all our other artifacts (see later) but, in the literature, there are numerous instances where similarly high lead contents have been reported (Riederer 1980; Craddock and Tite 1984; Maddin and Merkel 1990).6

There is, at Ittireddu, one more ingot fragment (no. 62417, bun type) that is also deviant in its trace element contents, although not as conspicuously so as the previous one. It is the only ingot in our suite of samples that contains a measurable trace of tin (130 ppm). Moreover, it is low in arsenic, cobalt, gold, and tellurium but high in lead and silver; the silver/gold ratio of 610 is about 100 times higher than normal (Table 1, Fig. 7). The higher-than-normal lead content of 830 ppm (compared to an average lead concentration in Ittireddu ingots of 48 ppm) goes together with an isotopic composition of this lead that is also different from that in all other ingots (Table 2). All 206Pb-normalized abundance ratios are higher; in both panels of Fig. 8 the data point is shifted towards no. 62405 and many of the artifacts. But it should be noted that in the 208Pb/206Pb vs 207Pb/206Pb presentation the data point does not fall on a mixing line between these high ratios and those in any of the other ingots, which is unambiguous proof that the anomalous lead isotope signature cannot have come about by mixing any two, or more, of the other ingots from our suite of samples. Actually, the same follows also from the trace element contents, because the extremely low concentrations of arsenic,

Table 1. Neutron activation analyses of samples from Sardinian ingots. Lead was determined by mass spectrometry with isotope dilution.

SARDINI	SARDINIAN INGOTS				ð								,	
	Sample	ð	Sn	Pb	As	Sb	ර	ï	Ag	Au	Fe	Zu	Se	Te
		%	bbm	mdd	mdd	mdd	mdd	mdd	bbm	bbm	mdd	mdd	bbm	mdd
Oxhide ingots	rots													
Ittireddu	62386	46	09>	22	2100	47	310	201	28	8.5	<170	36	158	96
	62387	86	o6>	150	5300	128	720	410	172	26.4	740	139	81	26
	62389	100	< 80	9	2870	79	360	320	64	10.6	<200	71	147	88
	62391	86	<70	30	5800	144	268	274	93	10.8	130	33	7	82
	62392	46	<65	12	2900	89	259	460	37	7.0	230	31	25	59
	62393	96	<75	40	4700	114	340	360	127	19.6	490	22	54	84
	62394	92	09>	9	6400	143	380	293	132	19.5	410	48	20	103
	62395	86	<50	20	2460	51	390	260	23	4.2	252	30	93	22
	62396	100	<110	20	4000	88	227	242	69	10.0	247	44	118	117
	62397	91	<65	35	2670	78	226	296	42	5.9	320	36	79	43
	62398	95	<110	20	5200	114	1460	320	40	8.4	3000	237	51	121
	62399	86	<70	50	2000	115	93	265	06	11.0	<180	24	20	70
	62400	94	< 90	40	4200	86	2340	430	79	47.0	11600	< 80	22	79
	62402	95	<100	20	2600	130	200	274	75	12.8	450	09>	107	107
	62410	100	<170	30	5400	120	840	400	112	37.0	089	132	98	115
	62414	46	< 90	8	1300	31	144	250	17	3.2	096	26	143	89
	62415	94	<150	06	6300	152	610	380	130	20.4	420	92	46	142
Pattada	SAS-16A	86	<50	20	3100	64	410	231	43	8.4	<150	89	86	84
	SAS-16B	94	09>	22	1990	43	1040	290	38	7.4	2570	200	132	29
	SAS-16C	82	<50	10	1100	27	201	263	21	3.9	00009	22	35	28
	SAS-16D	100	< 70	10	2000	137	540	290	102	16.4	910	129	94	128
	SAS-16E	100	< 70	7	5100	121	086	410	116	19.9	1060	132	51	86
	SAS-16F	100	< 90	45	2220	520	15	229	145	0.96	< 200	14	148	< 20
	SAS-16G	46	<20	1980	550	51	29	93	220	0.24	27900	259	61	<7

	<b>&gt;</b>	9>	148	142	106	86	163	7	93	39		65	18	89	119	271	69		8 >	<7
	17	12	95	142	26	48	88	6	22	61		45	22	40	100	130	33		23	<1
	239	9>	<50	20	210	110	82	17	188	48		225	67	122	73	< 70	120		410	380
	6700	<100	530	370	2800	1840	009	<150	3500	4100		1530	1230	006	1380	1410	1270		13700	4200
	8.9	0.34	15.3	20.2	18.8	17.8	15.6	0.59	35.0	5.5		11.2	3.8	31.0	35.0	29.7	6.7		2.96	0.02
	191	191	86	145	117	42	66	360	165	53		71	282	88	285	235	51		215	282
	78	141	271	289	370	245	296	288	490	200		430	410	320	330	400	380		82	2270
	29	4	370	320	1400	1130	620	9	1690	470		1030	160	540	570	740	086		221	155
	135	155	146	222	121	61	120	28	188	49		131	410	110	207	440	113		64	114
	400	300	0099	0086	4500	2990	5100	006	7700	2160		5100	2290	4400	2006	16100	2000		165	9200
	1570	320	20	40	65	20	30	830	06	20		8	45500	22	65	130	20		09	300
	<30	09>	<40	<120	<140	<100	<150	130	<150	09>		<100	110000	< 70	<150	<120	<80		<20	<20
	66	86	94	96	96	86	100	100	100	66		86	28	26	96	94	100		101	66
	SAS-710	SAS-711	62401	62404	62412	62413	62416	62417	62419	62421	nun ingots	62403	62405	62406	62407	62411	62420	30800	SAS-708	SAS-709
Bun ingots	Bonnanaro		Ittireddu								Oxhide or <i>t</i>	Ittireddu					62420	Tenneated	Bonnanaro SAS	

Table 2. Content and isotopic composition of lead in Sardinian copper ingots. In this and the subsequent tables the experimental uncertainties of all isotope abundance ratios (95 per cent confidence level) are 0.1 per cent or less.

	Sample	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	)
Oxhide ingots					
Ittireddu	62386	25	2.0695	.8393	.05381
	62387	150	2.0760	.8419	.05402
	62389	60	2.0729	.8411	.05401
	62391	30	2.0728	.8405	.05389
	62392	12	2.0701	.8398	.05387
	62393	40	2.0749	.8414	.05395
	62394	65	2.0719	.8406	.05398
	62395	20	2.0645	.8372	.05365
	62396	20	2.0705	.8398	.05386
	62397	35	2.0724	.8406	.05392
	62398	50	2.0704	.8399	.05387
	62399	20	2.0726	.8406	.05391
	62400	40	2.0732	.8408	.05390
	62402	50	2.0758	.8413	.05381
	62410	30	2.0736	.8412	.05401
	62414	8	2.0653	.8379	.05378
	62415	90	2.0756	.8416	.05376
			2.0750	.0410	.03390
Ozieri, Bisarcio	SARD 7-Z	80	2.0723	.8409	.05402
Pattada	SAS-16 A	50	2.0681	.8392	.05390
	SAS-16 B	55	2.0738	.8423	.05408
	SAS-16 C	10	2.0920	.8549	.05483
	SAS-16 D	10	2.0718	.8407	.05399
	SAS-16 E	7	2.0778	.8445	.05420
	SAS-16 F	45	2.0820	.8434	.05400
	SAS-16 G	1980	2.1265	.8735	.05589
Serra Ilixi	SARD 2-Z	70	2.0710	.8402	.05395
erru illiki	SARD 2-Z	500	2.0677	.8392	.05387
	SARD 3-Z	10	2.0747	.8451	.05424
			2.0747		.03424
Inknown	SARD 67-Z	110	2.0725	.8414	.05403
Bun ingots					
Bonnanaro	SAS-10710	1570	2.0742	.8372	.05363
	SAS-10711	320	2.0906	.8479	.05425
Ittimodda			2.0715		05390
Ittireddu	62401	50		.8402	.05389
	62404	40	2.0780	.8440	.05418
	62412	65	2.0728	.8410	.05400
	62413	20	2.0707	.8401	.05393
	62416	30	2.0711	.8401	.05392
	62417	830	2.0874	.8623	.05532
	62419	90	2.0728	.8410	.05401
	62421	20	2.0702	.8402	.05381

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m 11 A	251201000000000000000000000000000000000
Table /	continued

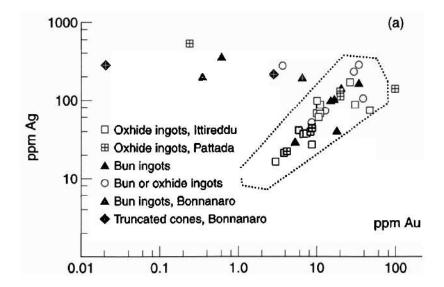
	Sample	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>204</sup> Pb/ <sup>206</sup> Pb
Oxhide or bun ingo	ots				
Ittireddu	62403	90	2.0718	.8402	.05387
	62405	45500	2.1241	.8745	.05581
	62406	25	2.0740	.8412	.05396
	62407	65	2.0777	.8431	.05406
	62411	130	2.0719	.8406	.05395
	62420	20	2.0705	.8397	.05387
Settimo San Pietro*	SARD 104b-Z	330	2.0865	.8459	.05424
Unknown	SARD 113-Z	80	2.1012	.8559	.05474
Truncated cones					
Bonnanaro	SAS-10708	60	2.0832	.8427	.05390
	SAS-10709	300	2.0709	.8318	.05299

<sup>\*</sup> Metal piece. Questionable whether ingot fragment. Samples marked 'Z' were provided by Prof. U. Zwicker, Erlangen.

cobalt, and especially gold cannot have come about by mixing together any other two or more specimens, which all have higher concentrations. This, then, leaves the puzzle of the tin content of 130 ppm in this sample. Of course, it is conceivable that a tiny splinter of tin, or of a tin bronze, somehow found its way into the melt from which this ingot derives, which would barely have affected the content of any other element. But what makes this trivial explanation hard to accept is that such an accidental contamination should have happened only to a sample that is quite distinct from all the others anyhow. More likely, all the anomalous features in trace element concentrations and lead isotopy were indigenous to the source ores of this ingot. That there should be local 'tin-contaminated ore deposits' (in the Iglesiente) had already been suggested by Tylecote et al. (1983) to account for the 'perceptible and significant' tin content (up to 1.8 per cent) of many of their ingot samples from the Nuragic site of Abini in central Sardinia.

The general trend among the ingots is that anomalous trace element concentrations are accompanied by lead with an anomalous isotopic fingerprint. This is also true for the four ingots from the Bonnanaro hoard (Fig. 8) which, moreover, are of a somewhat atypical shape and weight (Fig. 5).

At Ittireddu, the lead isotopic compositions of all ingots but the two already mentioned (nos. 62405 and 62417) fall into narrow bands. This is true for all ingots, independent of type. Most of the samples are comparable to other oxhide ingots from Sardinia and various locations in the eastern Mediterranean analyzed previously, although there is a tendency for the Oxford data to be displaced slightly from our data. If real, however, this systematic shift would be well within the quoted experimental uncertainties for both ratios of  $\pm 0.1$  per cent (2 $\sigma$ ). Note that two oxhide ingot specimens (nos. 62395 and 62414) extend the range of ratios hitherto



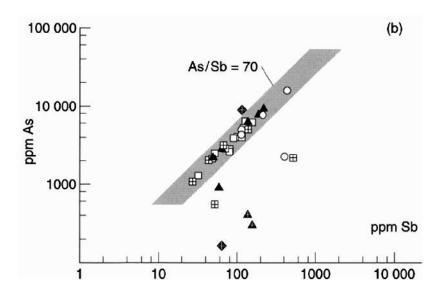


Figure 7(a). Gold and silver concentrations in copper ingot fragments from Sardinia. The outlined field shows the range of concentrations determined previously in oxhide ingots from Cyprus and various other locations in the eastern Mediterranean (Stos-Gale et al. 1997). Experimental uncertainties are smaller than the symbol size. (b). Arsenic and antimony concentrations in copper ingot fragments from Sardinia are highly correlated. For an absolute range in concentrations of a factor of 20 the As/Sb ratios range between 34 and 49. (The shaded bar delineates the field with As/Sb ratios between 30 and 70.) Exceptions are the unusual ingots from Bonnanaro and a few scattered fragments which are also outliers in the Ag-Au diagram (a) and in the isotopic composition of their lead.

observed. Moreover, in the  $^{208}$ Pb/ $^{206}$ Pb vs  $^{207}$ Pb/ $^{206}$ Pb diagram, these two samples fall outside the 'Old Cypriot (ore) field' as formerly defined by Stos-Gale and Gale (1994) and also well away from all new ore data reported for a number of ore occurrences from Cyprus that were not covered in previous publications (Gale et al. 1997). They are close enough, however, to make it not improbable that ores with such an isotopic signature may eventually be discovered on Cyprus or may have existed there in antiquity but now be exhausted.

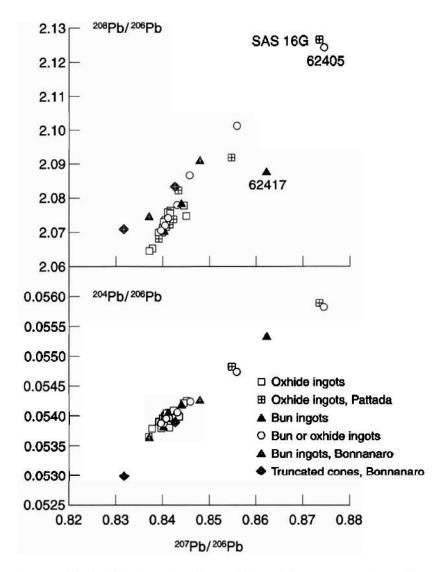


Figure 8. Lead isotope abundance ratios in Nuragic copper ingot fragments. No. 62405 is a bronze alloy fragment from Ittireddu, containing 11.0 per cent tin. In this and all subsequent figures the experimental uncertainties are comparable with, or smaller than, the size of the symbols.

Of the other ingots, the more interesting ones are those which, according to their lead isotope composition, do not belong to the main group. Aside from nos. 62405 and 62417 from Ittireddu (see earlier), this group includes all four ingots from Bonnanaro and also at least three specimens from Pattada (Fig. 8). While the lead in the main Ittireddu ingot group is different from that of all Sardinian copper ores analyzed so far, most of the 'maverick ingots' are comparable to Sardinian copper ores (Fig. 9). Of course, the most straightforward explanation is that these ingots are local products deriving from Sardinian ores. In the case of the Pattada specimens, classified as oxhide ingots, early production by relatively inexperienced craftsmen is perhaps indicated by the very poor casting quality. Drilling into the ingots upon taking samples for our investigations indicated a spongy and porous texture rather than solid metal. Indeed, the bulk density of 16B is some 25 per cent lower than that of solid copper (6.5 g/cm<sup>3</sup> as compared to 8.9 g/cm<sup>3</sup>) and

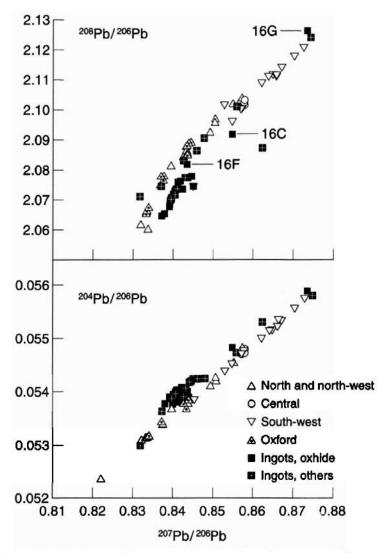


Figure 9. 'Main group' copper ingots have (as yet) no necopically matching copper ores (open symbols) in Sardinia, but for a number of 'maverick' ingots the lead isotope abundance ratios are perfectly con putible with this copper to derive from Sardinian sources.

that of 16C is only half that of massive copper. This is not the case for the Bonnanaro specimens, whose casting quality is quite good.

The importance of the result that the lead isotopy alle ws for a local origin of the ingots cannot be overemphasized. Leaving aside intriguing questions concerning early, pre-Nuragic, metallurgy on Sardinia, current archaeological evidence indicates that Sardinians became reacquainted with copper via the arrival from Cyprus of metal and metallurgical expertise during the twelfth century BC. Among the 'imports', oxhide ingots figure prominently, although it is not even clear whether they arrived in the course of normal trade relations between Sardinia and the eastern Mediterranean, whether they served as currency to pay for the harboring and resupply of the ships, or whether they are accidental 'imports' as cargo of a stranded vessel or vessels en route to more westerly destinations. But finding these imports together with locally produced ingots indicates that it did not take long to (re)introduce into Sardinia the art of extractive pyrometallurgy. It also brings up the question

why it should have been necessary to import a commodity that was locally produced as well. Quality is clearly not the critical factor (Muhly and Stech 1990), since the ingots from Bonnanaro are, in every respect, as technically good as any of the oxhide ingots. If it were not that demand exceeded supply, the most logical explanation might be that the oxhide ingots indeed served as currency in exchange for goods or services.

Using minor constituents to trace bulk metal to a particular ore deposit requires that the constituents in question indeed do derive from the ores. This holds true for trace elements as well as lead isotopy, for ingots as well as metal artifacts. The method fails if the trace element in question has been introduced to more than a negligible extent from any extraneous source. Since direct demonstration of this fundamental condition is impossible, circumstantial evidence must be invoked. One possibility as to how such an obliteration of ore fingerprints may have arisen has been extensively discussed in the literature (Gale and Stos-Gale 1982; Muhly 1985; Budd et al. 1995 and comments by Gale and Stos-Gale 1995; Hall 1995; Muhly 1995; Pernicka 1995; Sayre et al. 1995): the reuse and mixing of metal, e.g. by mixing virgin copper with scrap. As far as the reuse of scrap bronze is concerned, this possibility can be excluded in the present instance. The absence of measurable concentrations of tin, less than about one hundredth of one per cent, is unambiguous proof that these ingots are not contaminated with bronze; an upper limit to any contamination by standard bronze, with 10 per cent tin, is less than one part in a thousand.8 Because non-bronze artifacts do not contain a similarly well-suited tracer element - such as tin in the case of bronze - we can only conjecture, but not prove, that any potential admixture to the ingots of non-bronze scrap should also be low.

#### Artifacts

A total of 45 artifacts has been analyzed. Among them are 20 recognizable tools, such as axes, chisels, and daggers, one short piece of a decorated cylindrical handle (Pattada, no. 10, Fig. 4), and one charcoal shovel with twisted handle (Oliena, no. 58981, Fig. 6). According to their shape, two of the sampled fragments from Bonnanaro (nos. 10717 and 10718) are pieces of the same massive axe blade or chisel (Fig. 5); since they agree in the isotopic composition of their lead and their trace element contents, they will be considered to represent a single object. Fifteen other objects are sword fragments (12 from the Arzachena, Nuraghe Albucciu hoard and three from the second Ittireddu, Nuraghe Funtana hoard) while eight samples, all from the Arzachena hoard, were taken from pieces of sheet metal or fragments that are otherwise rather nondescript (Fig. 2).

It transpires that such a cursory phenomenological classification of the artifacts is surprisingly well-reflected in the chemical and lead isotope characteristics:

1. Chemically, all implements are made of standard bronze with tin content ranging between 6.8 per cent and 14.3 per cent (average 10.8 per cent tin (Sn); Table 3). Silver concentrations range between 207 ppm and 960 ppm, gold between 0.6 ppm and 18 ppm, with a negative correlation between the two

Table 3. Neutron activation analyses of samples from Sardinian artifacts. Lead was determined by mass spectrometry with isotope dilution.

SARDINIAN ARTIFACTS	N ARTIFA	CTS	S	ğ	<b>5</b>	ಕ	ć	N.	<b>₹</b>	:	ģ	7	5	Ę
	oampie	3%	mdd	mdd bbm	bbm Bbm	bbm Bbm	b midd	bbm	mdd bbm	mdd	ppm	ppm	bbm	bbm
Implements Arzachena chisel	20976	68	143000	069	3800	135	530	289	300	14.8	<550	31	61	82
Bonnanaro double axe double axe	10712 10713	98	110000	2400	2530 750	390 1610	97	206	890	3.5	220 520	70	40	<20 <25
socketed axe		8 6 8 8 8 8	101000 123000	3650	1090 840	530 134	16 65	520 283 370	300	1.34	<300 980	<15 57	13 42	\( \frac{20}{10} \)     \( \frac{1}{10} \)    \( \frac{1}{10} \)     \( \frac{1}{10} \)     \( \frac{1}{10} \)     \( \frac{1}{10} \)     \( \frac{1}{10} \)     \( \frac{1}{10
axe blade axe blade axe blade	10717* 10717* 10718*	8 2 2 2	103000	250 160	2120	370 310	16 16	380 370	340 293	10.2 9.1	<250 138	11	30 32	\$15 \$15 \$10
Oliena spatula	58981	91	139000	430	5300	161	233	320	207	17.8	<700	16	61	102
Pattada double axe	1	90	103000	2560	2770	97	221	295	610	3.3	2390	198	27	<15
axe adze	1 th =	88	99000	2410	8300	188	137	247	580	3.5	550	256	24	× × × × × × × × × × × × × × × × × × ×
flanged axe	4 70	68	121000	1580	3200	124	265	285	430	6.2	2680	133	36	× 40 × 40
chisel chisel	46	8 8	85000 $110000$	1960 4000	3400 2590	118 133	470 172	390 297	570 580	5.9	5800 2280	166 116	31 32	< 24 < 20
chisel chisel	8 6	8 8	112000 68000	1000 3780	5500 3700	91	320 149	202	800 710	3.6	4900 8200	380 239	39	<30
nandle or a chisel dagger	10	83	109000 94000	2040 1680	3400 2400	116	254 170	291 249	850 320	3.1	10700	298	40	<20 <20



	179 35 <30 196 25 <15		222 31 < 390 79									210 86 37 44			26	21	1660 24 <80	13	<1	73	23	2	
	5600 1 3800 1		<1000 2 740 3						2000	5 84807		1540 2			13700	11900	12300	160	<2000	610	<1000	<500	
•	960 8.8 370 3.6		13600 2.4 30 5.0									246 5.7					4400 0.98						
	360 281		<7 13 340									340	210		<75	165	520	79	<	114	< 70	<35	
	154 380 84 232		97 2 83 61									118 100					1940 74						
	3300 1700		9200 2170				• •					2530	ir 18-00		21	53000	39000	18100	4400	480	6300	23000	
_	74000 4780 112000 3370		<200 2730 16200 50	7400 20 9300 510		3300 90		8500 150	21700 390			75000 7400			09 09>	<70 4090		.4	٠,		31	<75 10	
	90 74 85 112			98 17 100 9	,	_		106 8		97 7		101 83 77			102	P== 441						66	
	13 13	gments	20974A 20974B	20974C 20974D	20974E	20974F 20974G	20974H	209741	20974N 20974I	20974M		62385 67385A	62385D	ript metal	20975A	20975B	20975C	20975D	20975E	20975G	20975H	20975R	
	dagger dagger	Sword fragments Arzachena									Ittireddu			Non-descript metal									

\* Same object.

elements in the sense that high silver goes together with low gold, and vice versa (Fig. 10).<sup>10</sup> Omission of the objects from Bonnanaro reduces the range in gold content by a factor of five and clustering of the data points becomes very much tighter, particularly for cobalt/nickel ratios. Lead concentrations fall between 160 ppm and c 5000 ppm, while the <sup>206</sup>Pb-normalized isotope abundance ratios are all high. In both panels of Figure 11, they plot in the upper right, with the objects from Pattada forming a particularly tight cluster. Because the samples from Pattada are a very homogeneous chemical group as well, they almost certainly derive from a single ore source. To the same group might belong the chisel from Arzachena. It is lower in iron and zinc but this can be accounted for by secondary changes during casting and remelting.

The six tools from Bonnanaro must clearly be assigned to a different source, not so much because of the differences in their lead isotopic composition but because of differences in their trace element signature. Cobalt contents are very much lower and arsenic/antimony ratios are only about five, as compared to 30 at Pattada (Fig. 10).

2. In the sword fragments, tin content clusters around 1 per cent. The exception is no. 20974A from Arzachena, with less than 200 ppm tin. But this piece is also differentiated from all others by its high silver content of 1.36 per cent and its low concentrations of nickel and, in particular, cobalt. Interestingly, this fragment is also distinct from all others in that the central rib is rectangular in cross-section (Fig. 2). Except for this outlier, silver contents are very much lower than in the implements; moreover, silver and gold concentrations are positively correlated, the ratio silver: gold being about six (Fig. 10). Cobalt concentrations also tend to be lower than in the implements; the same is true for lead. Isotopically, the lead is very heterogeneous (Fig. 11); isotope abundance ratios range from very high to very low, with some clustering, in five samples out of fifteen, at intermediate values around <sup>208</sup>Pb/<sup>206</sup>Pb ~ 2.095, <sup>207</sup>Pb/<sup>206</sup>Pb  $\sim 0.855$ ,  $^{204}$ Pb/ $^{206}$ Pb  $\sim 0.0546$ . For two pairs of fragments (nos 20974F and G; 20974I and L), the isotope abundance ratios of the members of each pair agree within the experimental uncertainties; they may be fragments of the same swords. The respective geometrical cross-sections of the fragments (Fig. 2) are compatible with such an assignment. What argues against it, however, are the threefold differences in tin content and the differences in cobalt concentration of about a factor of two.

Taking all the evidence together, we conclude that the 12 sword fragments from the Arzachena hoard derive from at least 10, perhaps even 12, different swords. For a hoard of scrap metal, this is about as enigmatic as the fact that all fragments are of approximately the same small size, the largest one being just over 4 cm long. All this argues against the sword fragments constituting a utilitarian hoard for traders or for founders. Traders, or merchants, hoards, by definition, consist of *complete* objects, usually more than one of a kind. Founders' hoards are typified by broken implements or pieces of metalwork and by ingot fragments destined for remelting. The nondescript bits and pieces of metal from Arzachena might perhaps fall into this category but not

an ensemble consisting of so many sword fragments of fairly constant size. Possibly, the whole collection of metal pieces from Arzachena is a treasure hoard of 'de facto refuse' collected and hidden for its value, both utilitarian and ritual, but never to be reused. The find context - a covered urn buried under the pavement of the terrace of a nuraghe - suggests it to be a deposit intended to procure divine favor to ensure the well-being of the nuraghe and its inhabitants. Such an explanation also would account for the low concentrations of tin in the sword fragments. At a time when the superior quality of bronze was fully realized in the production of axes, chisels, and daggers, it would be unreasonable to use soft, pliable copper for making swords if they were intended to be used as weapons. More likely, these swords were specifically made for ceremonial or votive offerings where the symbolic value was more important than the potential utility of the objects. The implication is that the deities whose favors were sought had no particular preferences for the metallurgical quality of the offerings or, more mundanely, that tin was an expensive commodity.

3. The sheets, fragments and cakes of metal, other than the sword fragments, from the Arzachena hoard are a mixed bag in every respect. The eight pieces sampled contain no detectable tin (less than 250 ppm in all cases, two of them less than 30 ppm); two of them have 5.3 per cent arsenic which is the highest by far among our suite of artifacts but, at the same time, another sample (no. 20975A) is extremely poor in arsenic, its 21 ppm being the lowest by far. Silver content tends to be high, while gold is lower than in any of the other artifacts (Fig. 10). Nickel and cobalt are generally low but show the largest variation among the three groups of artifacts. Lead isotope abundance ratios are quite variable and distinct. In two cases (nos. 20975G and R) the lead is extremely radiogenic; in both panels of Figure 11 the data points plot far off scale to the lower left. The remaining six are, in the lower part of Figure 11, similar to the other artifacts but in the <sup>208</sup>Pb/<sup>206</sup>Pb vs <sup>207</sup>Pb/<sup>206</sup>Pb panel they all plot well below the trend line as defined by the other samples. Note that, with but one exception, the sword fragments from the same hoard are entirely different from this scrap metal in their material properties. The exception is the outlier among the sword fragments already mentioned (no. 20974A) that agrees in the isotopic fingerprint of its lead and in its trace element contents with nos 20975E and 20975H. These three samples also have in common the exceptionally high silver content of more than 1 per cent and the unusually low cobalt content of 2 ppm or less.

The meaning of such a collection of bits and pieces, stored together with fragments of votive swords, is not clear at all. Indeed, it is not even clear what this metal is in the first place. Obviously it cannot be remelted material from fragments of broken implements or discarded objects as they were in use at the time. Such remelts would be similar to the objects they came from, in trace element contents and lead isotopy, which is clearly not the case. In this respect it is important to note that each of the pieces is extreme in at least one aspect, either the lead isotopic

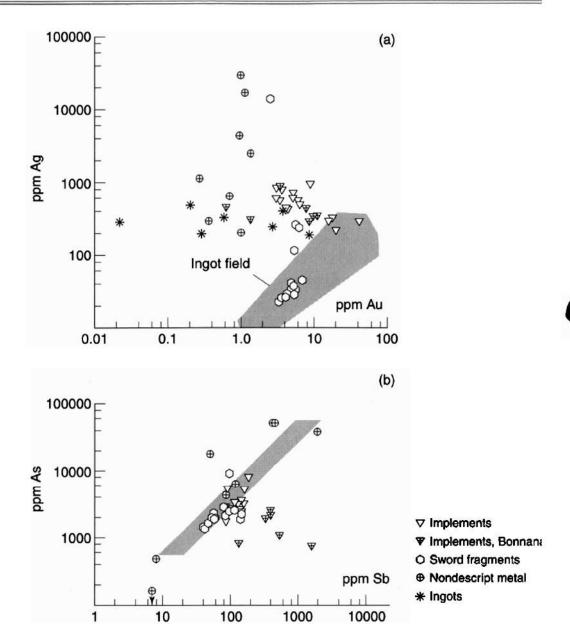
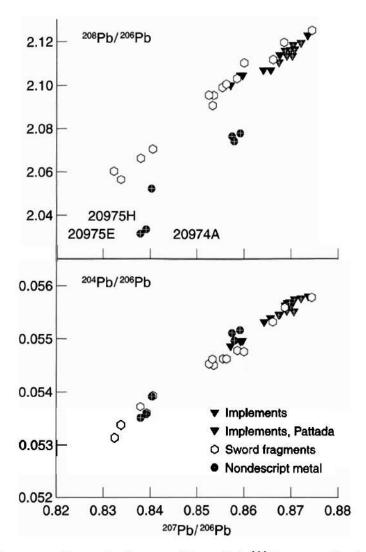


Figure 10(a). In a silver-gold diagram most artifacts plot far away, at elevated silver concentrations, from all ingot fragments while the sword fragments from Arzachena fall right into the ingot field. Isotopically, on the other hand, the same sword fragments are quite distinct from the ingots (Fig. 12). Apparently, the information content of lead isotope signatures and trace elements is complementary, not redundant (Pernicka et al. 1997). (b). Arsenic/antimony ratios in artifacts are less uniform than in ingots. (As a visual aid the grey bar from Fig. 7, indicating As/Sb ratios between 30 and 70, is shown for comparison.) Note that the implements from Bonnanaro are set apart by low As/Sb ratios as is also the case for the ingots from the same hoard.



**Figure 11.** Bronze implements all have high <sup>206</sup>Pb – normalized isotope abundance ratios which are particularly constant at Pattada. Sword fragments and the nondescript pieces of metal, on the other hand, show an extreme scatter, with two pieces (nos. 20975G and R) even plotting far off scale to the lower left. Note that sword fragments and the nondescript metal, both from the Arzachena, Nuraghe Albucciu hoard, are entirely different from one another in their lead isotope fingerprints.

composition or the concentration of one or more trace elements. This makes it impossible to explain them as mixtures of any of the kinds of metal analyzed. Perhaps we are seeing the products of tinkering with various kinds of ores. If so, apparently none of the sources, or of the products, were found to be entirely satisfactory because none of this metal shows up in any of the artifacts, the exception being the flat sword fragment no. 20974A already discussed.

#### Artifacts vs Ingots

A striking result of the lead isotope analyses is the complete absence among the bronze implements of the isotopic fingerprint of the lead from the oxhide ingots (Fig. 12). Not a single one of our 21 implements, nor any of the 10 bronzes from

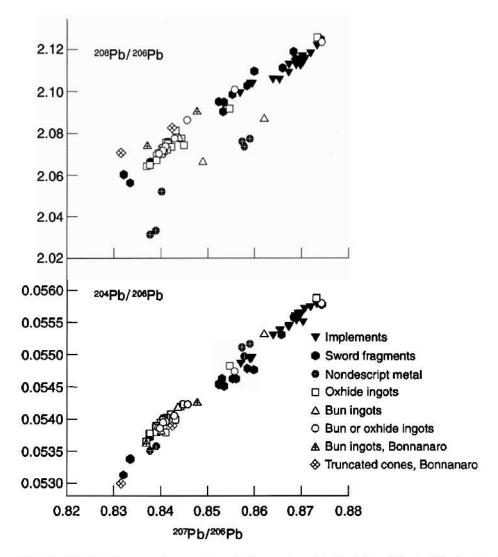


Figure 12. Lead in 'main group' copper ingots does not match that in artifacts. Obviously, in artifacts extrinsic lead from other sources dominates, which accords with the observation that the lead content of artifacts tends to be very much higher than that of ingots.

the Santa Maria in Paulis hoard analyzed at Oxford (Gale and Stos-Gale 1987), contains lead that is even remotely similar to that in the main group of oxhide ingots. Since the lead content of the implements is much higher than that of the ingots, we can think of three possibilities to account for this observation.

- Oxhide ingots were not utilized for the manufacture of useful things like implements or weapons.
- Oxhide ingots were utilized but their lead was diluted beyond recognition by large amounts of extraneous lead brought in together with the tin, or cassiterite, added to produce bronze.
- Oxhide ingots were never employed alone to make bronze but were always mixed with copper from other sources, and this copper contained high concentrations of lead with an isotopic composition different from that in the oxhide ingots.

None of these suggestions is entirely satisfactory, however. For oxhide ingots never to have been alloyed with tin to make bronze artifacts implies a solely non-utilitarian, ceremonial, almost sacred meaning of the ingots for which there is no evidence. Although oxhide ingots have been found in contexts that suggest a special importance in Nuragic life (Lo Schiavo 1986), there is also ample evidence to the contrary. Both hoards from Ittireddu, for example, as well as the heaps of scrap metal together with fragments of both oxhide and plano-convex ingots at Villagrande Strisaili (Vagnetti and Lo Schiavo 1989) clearly indicate that oxhide ingots were also valued simply for their metal.

More plausible is that the isotopic signature of the oxhide lead does not show up in the artifacts because extraneous lead, with a different isotopic fingerprint, found its way into the bronzes where it now dominates. At Pattada, for example, in the hoard where most of our implements came from, the average lead content is 2500 ppm, which is about 50 times higher than the typical lead concentration in the ingots of, say, 50 ppm. Clearly the isotopic signature of the latter will hardly be recognizable any more in the presence of 50 times more lead if that has a different isotopic composition. As to the question of how extraneous lead might have been introduced, there is the obvious possibility that it came in together with tin because these are the two elements, in addition to silver, that are much more abundant in bronzes as compared to the ingots. 12 In the extreme case that essentially all lead had been brought in together with tin, or with cassiterite if this should have been used directly (Tylecote 1977; Buchholz 1980), the tin/lead ratio would have to have been the same as it is in the bronze, namely about 40. But for tin to have contained some 2.5 per cent lead poses a problem in so far as all tin ingots analyzed until now are rather poor in lead. Among 15 ingots, 6 from Kefar Shamir, 5 from Hishuley Carmel, and 4 from Uluburun (Maddin 1989; Begemann et al. 1999), the highest lead content was 0.22 per cent, and in most samples it was even some 50 times smaller than that. In the absence of any tin ingots from Sardinia one can only speculate what their lead content might have been. Perhaps the tin utilized in Sardinia came from entirely different sources from that retrieved from shipwrecks in the eastern Mediterranean. Perhaps western tin from Cornwall, Brittany or Iberia was used in Sardinia – or possibly even provided by local sources; whatever tin was used, it may have contained a small percentage of lead as a natural contaminant.13

There are problems, however, with such an explanation. First, the lead/tin ratio in the implements is not constant, not even in objects from the same site. Taking again Pattada, the lead/tin ratio varies about sevenfold, between 0.9 per cent and 6.5 per cent, and this is much more than can be accounted for by analytical uncertainties. Possibly, such variations might have come about if cassiterite, rather than metallic tin, had been added to the copper to make bronze. In particular, if the unavailability of fine-grained placer cassiterite led to the use of chunks of tin ore, it is conceivable that different ore charges were quite variable in their lead admixture. However, the rather constant tin content of the bronzes argues against such an explanation. Moreover, this explanation would not account for the fact that the lead isotope abundance ratios in implements from different sites are distinctly different from one another.

Quite clearly, the lead utilized at different sites, or at different times, was not the same, and this is hard to understand if the lead had been associated with tin.

The interesting analytical side-effect of adding lead together with tin, so as to bring the lead content of oxhide ingots up to the level observed in bronze, would be that presumably one need not much worry about other trace elements. Tin and cassiterite are, as a rule, rather free from large amounts of elements used to characterize and trace back copper. Hence, their addition to copper will leave the concentrations of these trace elements and their abundance pattern largely unaffected. On the other hand, changes in trace element characteristics may have resulted if coppers of different provenance had been mixed together and the mixture subsequently been alloyed with pure tin. We shall return to this topic later in connection with the provenance question. But the main difficulty with the mixing-ofcoppers hypothesis is, of course, the extreme scarcity of ingots made from the putative copper that is high enough in lead content and has the required high isotope abundance ratios. With the possible exception of SAS 16G from Pattada, there are only four, possibly five, more ingots reported that would qualify. 14 Why, then, the extreme dominance among the ingots of those with low lead content and 'normal' isotopic composition? Why should there be only four or five ingots of one kind, out of a total of 80 or so that have been analyzed, and why not about an equal number of both kinds? Since oxhide ingots are more liable to break than, say, the truncated-cone variety from Bonnanaro, statistics based upon ingot fragments can be quite misleading. But we doubt that the apparent dominance of oxhide ingots is solely due to an observational bias.

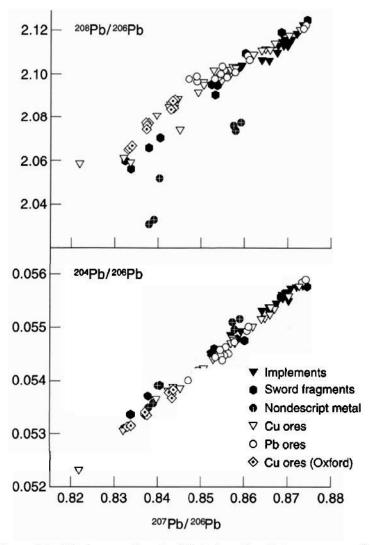
#### POTENTIAL ORE SOURCES FOR INGOTS AND ARTIFACTS

Provenance studies of metal artifacts based on the isotopic composition of their lead can only be as good as the database of ores available for comparison. It is understandable, then, that considerable scepticism met the announcement that none of the Sardinian oxhide ingots was made from Sardinian ores (Gale and Stos-Gale 1987). Since, at that time, the sampling program for Sardinian ore deposits was far from complete, and since a large number of published analyses (e.g. Boni and Köppel 1985) appeared to have been unknown to the authors, such a conclusion appeared to be premature at best. We wish to emphasize that even now, with a much more comprehensive database at hand (Stos-Gale et al. 1997; this article, Table 5), this striking result still holds true (Fig. 9). The only possible exceptions are three samples from Pattada (16C, F, G). But, for three of us (F.B., E.P., S.S.-S.), their classification as fragments from oxhide ingots is not unequivocal in the first place (Fig. 4).

There are other ingots, on the other hand, where the lead isotope fingerprints are fully compatible with a derivation of their copper from Sardinian ores (Fig. 9). For some of them, the high lead content indicates deliberate addition of lead, vitiating any attempt to trace back the copper. But there is no reason to believe that this should also be the case for the four ingots from Bonnanaro. At least three of these have perfect matches for their lead isotopy among copper ores from northern

Sardinia: SAS 10708 agrees, within experimental uncertainties, with a chalcopyrite from Capo Marargiu (CMCP, Stos-Gale et al. 1997), SAS 10710 with malachites from schist ores at Castello di Bonvei (Stos-Gale et al. 1997), and SAS 10711 (almost) with a malachite from Terra Padedda (Table 5). Unfortunately, trace element data are not of much help in adjudicating on these assignments. For the ores measured at Oxford, there are no such data available at all and just a single sample has been analyzed from Terra Padedda (Table 6). If this should be representative, however, then there will be serious problems with establishing a genetic connection between ore and artifact because the concentrations of gold, nickel and, in particular, cobalt are all very much higher in the ore than in the respective ingot. 15

Subsequent to the suggestion that the Sardinian oxhide ingots were Cypriot imports (Gale and Stos-Gale 1987), there has been a heated debate over the certainty and unambiguity of such an assignment (Cherry and Knapp 1991;



**Figure 13.** Most copper-based artifacts have Sardinian copper and/or lead ores to match the isotopic composition of their lead. Surprisingly, this is not the case for the pieces of nondescript metal scrap from Arzachena although they certainly are expected to be local products.

Table 4. Content and isotopic composition of lead in copper and bronze artifacts from Nuragic Sardinia.

		Jurumu	•		
SARDINIAN	ARTIFACTS	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>204</sup> Pb/ <sup>206</sup> Pb
Implements Arzachena					
20976	chisel	690	2.1138	.8677	.05547
20970	Ciusei	690	2.1130	.0077	.03347
Bonnanaro					
10712	double axe	2400	2.1068	.8642	.05533
10713	double axe	950	2.1001	.8572	.05488
10714	socketed axe	710	2.1046	.8593	.05495
10715	double axe	3650	2.1229	.8736	.05581
10716	axe blade	280	2.1044	.8597	.05497
10717*	axe blade	250	2.1045	.8597	.05497
10718*	axe blade	160	2.1045	.8597	.05498
Oliena					
58981	spatula	430	2.1068	.8657	.05540
Pattada	double axe	2560	2.1158	.8696	.05566
1 2	double axe	1350	2.1136	.8691	.05558
3	axe adze	2410	2.1136	.8700	.05568
4	flanged axe	2140	2.1161	.8695	.05567
5	flanged axe	1580	2.1151	.8700	.05563
6	chisel	1960	2.1182	.8705	.05552
7	chisel	4000	2.1162	.8708	.05574
8	chisel	1000	2.1102	.8675	.05546
8 9	chisel	3780	2.1149	.8694	.05559
10	handle of a chisel	2040	2.1191	.8722	.05577
11	dagger	1680	2.1160	.8699	.05562
12	dagger	4780	2.1162	.8692	.05555
13	dagger	3370	2.1154	.8689	.05563
Sword fragm	T-1				
Arzachena			921 (14200000000)		101 111 1 <u>0</u> 1 111 111
20974 A		2730	2.0334	.8392	.05359
20974 B		50	2.0603	.8323	.05313
20974 C		20	2.0565	.8337	.05338
20974 D		510	2.1196	.8686	.05559
20974 E		450	2.1117	.8661	.05533
20974 F		230	2.0952	.8536	.05452
20974 G		90	2.0955	.8525	.05454
20974 H		40	2.0706	.8405	.05393*
20974 I		150	2.0988	.8555	.05463
20974 K		60	2.0907	.8533	.05463
20974 L		390	2.1005	.8563	.05463
20974 M		40	2.0663	.8379	.05372*

continued on next page

C 11	852 859		•
Inhi	04	continue	7

	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>204</sup> Pb/ <sup>206</sup> Pb
62385	450	2.1030	.8586	.05479
62385 A	7400	2.1249	.8745	.05579
62385 D	370	2.1100	.8602	.05477
Nondescript metal Arzachena				
20975 A	60	2.0523	.8403	.05392
20975 B	4090	2.0742	.8580	.05498
20975 C	5130	2.0779	.8593	.05517
20975 D	2350	2.0765	.8575	.05512
20975 E	900	2.0314	.8379	.05350
20975 G	10	1.9074	.7827	.04989
20975 H	3100	2.0335	.8391	.05357
20975 R	10	1.6285	.6813	.04269

<sup>\*</sup> Same object.

Muhly 1991; Budd et al. 1995; Knapp 2000). Our opinion on this question is that, as far as the scientific evidence is concerned, Cyprus is still the most convincing case of all possibilities discussed so far. Moreover, we find it not in any way peculiar that the oxhide ingots should all be traceable to the same origin, presumably to Cyprus. This particular shape appears to have been a common ingot shape on Cyprus but there is no reason why Sardinian copper smelters should have preferred this shape also. There are a number of rational arguments in favor of it. True, the shape facilitates safe storage of ingots on board ship as well as transport by humans and animals. And it is relatively easy to break off pieces, as must undoubtedly have happened before final use. Still, as the copper cargos of the shipwrecks from Uluburun and Cape Gelidonya testify, these were not overruling considerations because these ships carried both oxhide as well as bun ingots (Bass 1967, 1986). This being said, we are nevertheless puzzled by the truncated-cone shaped ingots from Bonnanaro. The heaviest of them (no. 10708) weighs more than 11 kg which, given its shape, makes it particularly unwieldy. It has to be carried with both hands and, for taking off pieces for further use, it has just about the most unfavorable shape. Arguing that it was an intermediary product not yet meant for final use makes one wonder why it should have been desirable, or necessary, to remelt such a charge of perfectly good copper.

Finding potential ore sources for the 21 implements in our suite of samples is not straightforward, because the possibility cannot be excluded that some, or even most, of their lead has been added to the copper. Hence, it is not clear among which kind of ores one has to search. If the lead should have been brought into the bronzes together with the tin, there is not much we can do at present. As discussed earlier, no tin ingots with the required high lead contents have been reported. Among

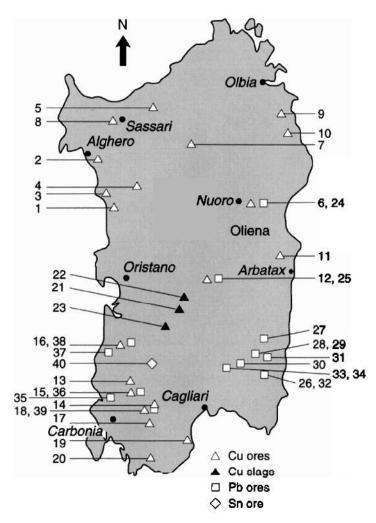


Figure 14. Location of ore occurrences investigated. Numbers are keyed to those in Table 5, column 1.

copper and lead ores, on the other hand, the bronze implements from Pattada and the chisel from Arzachena have isotopically matching occurrences in the Iglesiente-Sulcis region in south-west Sardinia (Fig. 13). Nurallao-Nieddiu, Arenas and Nuxis, alone or in combination, cover the whole range observed for those particular implements. The same was also found to be the case for some of the bronzes from S. Maria in Paulis (Gale and Stos-Gale 1987). Actually, the lead isotopic composition at Nurallao-Nieddiu is not very well constrained; it covers even the two artifacts with the <sup>206</sup>Pb-normalized highest abundance ratios, viz. one of the double axes from Bonnanaro (no. 10715) and one of the small sword fragments from Ittireddu (no. 62385A). Interestingly, these isotopic compositions agree also with

those in the two Nuragic copper slags from Nurallao-Nieddiu some 60 km to the north-east (Table 4).

An interesting group of artifacts is that with <sup>207</sup>Pb/<sup>206</sup>Pb ratios of around 0.858 and <sup>208</sup>Pb/<sup>206</sup>Pb ~ 2.105. It comprises four implements from Bonnanaro, a sword fragment from Arzachena (no. 20974L), and the two remaining sword fragments from Ittireddu. What makes this group interesting is the agreement of the isotopic fingerprint with those of copper and lead ores from Funtana Raminosa, 'generally thought to have been a potential source of Sardinian copper in ancient times' (Lo Schiavo 1988b). A word of caution must be added, however, because the assignment is not unique. There are also ores in the Iglesiente-Sulcis region, as at Rosas-Sa Marchesa, with a matching isotopic signature. Trace element data are once more missing which might otherwise have allowed plausible differentiation between different ore deposits. Independent of the location of the exact source region of the ores, however, there is every reason to assume these artifacts to be made from local metal. This is important since there is among this group a peculiar double-loop socketed axe of the Iberian type '42A West Portugal' (Fig. 5). Its lead isotopic composition as well as the trace element concentrations are indistinguishable

from those of three other 'normal' axes from Bonnanaro, an indication that all four pieces derive from the same ores. If these indeed were Sardinian ores, this would be another proof that foreign styles in artifacts testify to cultural exchange but not necessarily to an exchange of material goods. The same can also be said of the shovel fragment from Oliena, for which a matching mould has been found in Sardinia (see earlier).

Finally, there is the nondescript metal from Arzachena, where the extreme scatter in trace element contents is accompanied by an extreme spread in the lead isotope abundance ratios. Nonetheless, a common feature of the lead isotopy is that, in the <sup>204</sup>Pb/<sup>206</sup>Pb vs <sup>207</sup>Pb/<sup>206</sup>Pb panels, the data points plot together with those of the other samples while, in the upper panels, they fall consistently below the trend line as defined by the other objects. Clearly, there is a deficit of thorium-derived <sup>208</sup>Pb, which is to say that this lead comes from a magma source with the lower-than-average thorium/uranium ratio typical of deep-seated magmas. Aside from these general remarks that might help to search for such a source, there are at present no copper ores known in Sardinia that would meet these specifications.

#### SUMMARY AND CONCLUSIONS

Until about 30 years ago, Nuragic Sardinia was considered to have been an island of 'withdrawn isolation' (Tylecote et al. 1983). Since then, the situation has changed dramatically. About 100 Nuragic hoards from all over Sardinia have come to light that testify to extensive long-distance connections with the Aegean in the east and the Iberian peninsula to the west. The hoards are also clear evidence for a widespread use in Sardinia of copper and bronze towards the end of the second millennium BC. Nonetheless, while the number of hoards and of objects known till now are impressive, the critical points brought up in the discussion of Muhly and Stech (1990) remain pertinent. We still know very little about the internal social and economic organization of Sardinia at this time, nor is the relation obvious between nuraghes. Because all of the hoards are chance finds that do not derive from well-dated strata in systematic excavations, their immediate archaeological context is not always clear. Their chronological placement is often related to the degree of complexity of the nuraghe where the hoards were found and on associated imported pottery from other Mediterranean regions. Moreover, with regard to copper and bronze finds, there still is a regrettable lack of evidence in Sardinia for extractive pyrometallurgy. There is no evidence for old mining nor are there heaps of smelting slags that would tell us about the scale of copper production, or reveal technical details of the extraction processes employed. It still is not possible to amplify the conclusion of Muhly and Stech (1990) that, in Sardinia, 'metallurgy does not seem to have created social change, but rather responded to it'.

With the present data at hand, it is clear that, during Nuragic times, artifacts made of bronze were not just imported into Sardinia but also manufactured locally. The lead isotope signature of the oxhide ingots suggests them to be of non-Sardinian origin; presumably they are imports from Cyprus. This signature, however, is extremely rare, if not completely absent, among the implements and tools analyzed.



Their lead rather agrees in its isotopic composition with that found in Sardinian copper and lead deposits. We take this to be evidence that this lead was added locally in the course of manufacturing the bronze artifacts, although the technical reason why lead should have been added is not clear at present. Moreover, the lead added to the implements recovered at Bonnanaro is isotopically distinct from that added at Pattada. It would appear that the bronze ingots utilized at the two sites were local products and not supplied by some central ingot foundry, provided the artifacts are contemporary. Otherwise, there may have been a change with time in the provenance of the lead.

That bronze ingots should have existed as an intermediary product between copper ingots and bronze artifacts is, at the present time, conjecture because, puzzlingly, such ingots, or fragments thereof, are almost completely absent from the wealth of several hundred Nuragic metal fragments analyzed. Nevertheless, the implements from hoards such as Bonnanaro and Pattada are so constant in their tin content that we find it inconceivable that the bronze for each object, small or large, should have been prepared individually by mixing just the right amounts of copper and tin. More likely, they derive from larger batches of bronze of fairly constant composition. Where, then, are these bronze ingots?

The limitations in our knowledge about Nuragic Sardinian metallurgy is evident from the present data also. There are scraps of metal with some 5 per cent arsenic, but there are essentially no artifacts with a similarly high arsenic content. Among the 100 and more analyses of artifacts (Table 3; Junghans et al. 1960; Riederer 1980; Tylecote et al. 1983; Craddock and Tite 1984; Maddin and Merkel 1990), there are very few objects reported to contain more than 1 per cent arsenic. Obviously, this is not a secondary feature that came about by preferential oxidation or the removal from the metal of arsenic during extensive reheating and melting of the metal, because copper *ingots* with a high arsenic content are equally rare. Interestingly, the suite of *pre*-Nuragic weapons analyzed by Cincotti et al. (1998) includes two swords and two daggers with around 5 per cent arsenic.

Candidate regions for having supplied the copper of the native Sardinian ingots and the lead in the implements and tools are the Iglesiente-Sulcis district in south-west Sardinia, although verification is needed through more extensive data collection, in particular by measuring the trace element characteristics of these deposits. For the time being, it remains a hypothesis. The same is true for the sourcing to Funtana Raminosa of some of the implements from Bonnanaro and a few sword fragments. Tempting as such an assignment might be, because archaeologists have long suggested these mines to have been the source of copper in ancient times, this again needs support from trace element data.

#### APPENDIX

In the course of the present investigation, a number of ores from various locations in Sardinia were analyzed for the isotopic composition of their lead (Fig. 14), as well as three archaeological copper slags from central Sardinia. The results are presented here (Table A1). Furthermore, the chemical composition of three series of specimens











Table 5. Lead content and lead isotopic composition of Sardinian ores. Numbers after the find locations are keyed to Fig. 14. All lead contents are very approximate because no attempts were made to homogenize large chunks of ore.

Location	Sample	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>204</sup> Pb/ <sup>206</sup> Pb
Cu ores					
North and Northwest					
Bosa (1)	SARD 124	1150	2.0850	.8425	.05382
Calabona (2)	SARD 121a	40	2.0864	.8429	.05379
Calabona (2)	SARD 121b	4	2.1040	.8573	.05482
Calabona (2)	SARD 121c	2360	2.0874	.8435	.05377
Calabona (2)	CA 2	2	2.0850	.8436	.05389
Capo Marargiu (3)	SARD 123	40	2.0891	.8446	.05386
Mara (4)	SARD 122	10	2.0616	.8320	.05308
Nulvi (5)	SARD 118	410	2.0970	.8507	.05419
Nuoro (6)	SARD 46	30	2.0813	.8396	.05367
3					
Ozieri (7)	SARD 120	3900	2.0956	.8506	.05426
Ozieri, Su Elzu (7)	SUE 2	18000	2.1017	.8551	.05454
Sassari (8)	SARD 112	6	2.0597	.8338	.05317
Terra Padedda (9)	TP 1	90	2.0923	.8492	.05410
Torpe (10)	SARD 119	3	2.0591	.8219	.05234
Central					
Baunei (11)	SARD 125	780	2.1025	.8571	.05471
Funtana Raminosa (12)	SARD 41c	2000	2.1016	.8580	.05480
Funtana Raminosa (12)		1000	2.1025	.8578	.05475
Funtana Raminosa (12)		4800	2.1035	.8579	.05473
Southwest					
Arenas (13)	SARD 128	21500	2.1122	.8648	.05519
Campana Sissa (14)	SARD 130	40	2.1007	.8576	.05479
Domusnovas (15)	SARD 97b	900	2.1114	.8659	.05525
Domusnovas (15)	SARD 108	210	2.1212	.8727	.05578
The could be the property of the contract of t	SARD 108	n.b.	2.0968	.8547	.05455
Domusnovas (15)		11000		.8703	
Domusnovas (15)	SARD 129a		2.1183		.05559
Domusnovas (15)	SARD 129b	6700	2.1147	.8671	.05536
Domusnovas (15)	SARD 129c	100	2.1117	.8639	.05517
Montevecchio (16)	SARD 127	22000	2.1095	.8621	.05503
Nuxis (17)	SARD 132	280	2.1120	.8661	.05539
Rosas (18)	SARD 131	6800	2.1004	.8571	.05473
Sarroch (19)	SARD 133	80	2.0747	.8451	.05387
Teulada (20)	SARD 134	170	2.1021	.8529	.05441
Cu alam					
Cu slags					
Central	0.1 DD 445	-	0.4400	0.600	05540
Gesturi (21)	SARD 105	1	2.1108	.8633	.05510
Nurallao (22)	SARD 12a	<1	2.1231	.8733	.05575
Nurallao (22)	SARD 45b	13000	2.1235	.8746	.05593
Villanovaforru (23)	SARD 103	4	2.1126	.8638	.05535

Table 5. continued

Location	Sample	ppm Pb	<sup>208</sup> Pb/ <sup>206</sup> Pb	<sup>207</sup> Pb/ <sup>206</sup> Pb	<sup>204</sup> Pb/ <sup>206</sup> Pb
Pb ores					
North					
Nuoro (24)	SARD 100	n.b.	2.1004	.8545	.05459
Central					
Funtana Raminosa (25)	SARD 42	n.b.	2.1023	.8553	.05448
Southeast					
Baccu Arrodas (26)	SARD 240-B	n.b.	2.0985	.8534	.05445
Baccu Locci (27)	SARD 580-B	n.b.	2.1003	.8564	.05453
Baccu Monte Lora (28)	SARD 170-B	n.b.	2.0969	.8491	.05420
Bruncu Lionaxi (29)	SARD 100-B	n.b.	2.0989	.8559	.05465
Bruncu Molentinu (30)	SARD 140-B	n.b.	2.0979	.8471	.05401
Bruncu Ventura (31)	SARD 70-B	n.b.	2.0993	.8489	.05418
Monte Narba (32)	SARD 280-B	n.b.	2.0990	.8540	.05450
Serra S'Ilixi (33)	SARD 390-B	n.b.	2.0980	.8532	.05445
Tacconis (34)	SARD 370-B	n.b.	2.1036	.8549	.05440
Southwest					
Campo Pisano, Iglesias (35)	SARD 1080-B	n.b.	2.1224	.8740	.05587
Domusnovas (36)	SARD 109 b	n.b.	2.1212	.8735	.05585
Ingurtosu (37)	SARD 1020-B	n.b.	2.1088	.8607	.05494
Monte Poni, Íglesias (35)	SARD 1060-B	n.b.	2.1228	.8742	.05588
Montevecchio (38)	SARD 1010-B	n.b.	2.1070	.8611	.05502
Rosas (39)	SARD 1090-B	n.b.	2.1014	.8579	.05474
St.Giovanni, Iglesias (35)	SARD 48 a	n.b.	2.1233	.8742	.05590
	SARD 48 a	n.b.	2.1233	.8742	.05590
Villacidro (40)	SARD 101	5	2.1117	.8645	.05521

Samples marked 'B' were provided by Profs K. Germann and H.-J. Schneider (Freie Universität Berlin), CA 2, SUE 2, and TP1 by A. Krausse, all others by Profs P. Virdis (Cagliari) and U. Zwicker (Erlangen).

from ore deposits in northern Sardinia was determined (Table A2). It should be noted that the *copper* contents listed in column 2 are as measured but that the concentrations of all other elements are expressed as abundance ratios relative to copper. For Calabona and Bena de Padru, there is a clear tendency for these abundance ratios to depend on the actual copper content of the analyzed specimen, indicating that trace elements do not exclusively reside with copper minerals. Since, in antiquity, only copper-rich ores will have been utilized, only the abundance ratios of these copper-rich specimens should be used when comparing the trace element contents of ores with those in copper metal.

all |

Sample	<del>گ</del> %	As ppm	Sb	Ppm Ppm	iz d	Ag ppm	Au ppm	Se
CA1	2	0099	3200	210	380	69	3.0	370
CA2	17	79	44	38	99	9	0.23	29
CA3	Н	700	360	41	270	151	3.1	48
CA4	12	47	13	26	96	7	0.1	22
CA5	17	24	7	52	114	2	0.07	22
CA8	3	294	218	330	450	77	0.79	23
BdP1	H	31000	089	099	6200	3400	0.45	244
BdP2	2	00086	1100	2560	26000	2300	0.79	151
BdP3	27	7400	280	160	1030	820	0.27	43
BdP4	35	6100	218	134	860	096	0.14	90
BdP5	က	13200	640	1100	9300	970	0.43	247
BdP6	31	8200	240	103	029	770	0.13	41
SuE2	8	3900	290	28	130	169	0.67	12
SuE7	က	009	400	150	280	300	2.53	200
OS1-2	7	287	22	850	1050	8600	3.60	0099
TCB3	1	1000	226	770	1920	520	2.40	1170
TIP1	2	310	270	950	1250	380	2.60	130
	CA1 CA2 CA3 CA4 CA5 CA8 BdP1 BdP2 BdP3 BdP3 BdP4 BdP5 BdP5 BdP5 BdP7 CA5 CA5 CA5 CA5 CA4 CA5 CA5 CA5 CA4 CA5 CA5 CA4 CA5 CA5 CA4 CA5 CA5 CA5 CA5 CA5 CA5 CA6 CA7 CA6 CA6 CA7 CA6 CA7 CA6 CA6 CA7 CA6 CA6 CA7 CA6 CA7 CA6 CA6 CA7 CA6	7 7 7 8 8 7	17 17 17 17 17 17 17 17 17 17 18 33 113 34 18 38 3 3 13 13 13 14 15 16 17 17 17 17 18 18 18 18 18 18 18 18 18 18 18 18 18	2 6600 3 17 79 79 12 447 17 24 3 294 3 13000 1 27 7400 35 6100 31 8500 27 7400 36 600 3 13200 3 13200 3 13200 3 13200 3 13200 3 13200 3 247 7400 1 287 2 <1 287	2 6600 3200 17 79 44 18 47 13 12 47 13 17 24 7 18 24 7 19 88000 1100 2 27 7400 280 6100 218 3 13200 640 1 88 3900 290 400 2 <1 287 22 2 <1 1000 226 2 310 270	17       6600       3200       210         17       79       444       38         11       700       360       41         12       47       13       26         17       24       7       52         17       24       7       52         24       7       52       330         29       29       218       160         27       7400       280       160         35       6100       218       134         31       8500       240       1100         31       8500       240       1103         400       400       150         2       400       226       770         1       1000       226       770         2       310       270       950	2       6600       3200       210       380         17       79       44       38       66         12       700       360       41       270         12       47       13       26       96         17       24       7       52       114         2       24       218       330       450         27       7400       680       660       6200       2         27       7400       280       160       1030       2         35       6100       218       134       860       2         3       13200       640       1100       6300       2         3       600       240       1103       670       8         4       287       22       850       1050       8         5       <	2         6600         3200         210         380         62           17         79         44         38         66         6           11         700         360         41         270         151           12         47         13         26         96         7           17         24         7         52         114         5           2         24         7         52         114         5           2         24         7         52         114         5           2         35         680         680         660         230         3400           27         7400         280         160         2300         2300         2300           35         6100         218         134         860         960         960           33         13200         640         1100         6300         970         970           8         3900         290         28         130         169           9         600         400         150         280         300           1         1         1000         226         770

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#### Notes

- 1. The fragments were re-examined and re-classified. For some of them, the assignment to the parent ingot type is different from that listed in Maddin and Merkel (1990).
- 2. On the occasion of a second site visit, in the company of the man who originally discovered the hoard and indicated it to the Soprintendenza, an eighth fragment of an ingot was found  $(5.5 \times 7 \times 3.3 \text{ cm}$ , weight 384 g, bulk density  $6.4 \text{ g/cm}^3$ ), embedded among the stones at the top of a small rocky cavity. In the soil, there were also two more tiny fragments of a sheet vessel and a fragment of impasto pottery. With this last addition, the Pattada ingots together weigh 8.11 kg.
- 3. The chemical composition was determined by instrumental neutron activation analysis except for lead, which was measured by isotope dilution. Lead isotope measurements were performed by thermal ionization mass spectrometry;  $2\sigma$  uncertainties in isotope abundance ratios are less than 0.1 per cent in all cases.
- 4. The following misprints should be noted in this article (Maddin and Merkel 1990): Inventory number 62103 should read 62403 (pp. 62/63), 62491 should read 62421 (pp. 76/77). In Table IV 'Oxhide or plano-convex ingots' (pp. 82/83) for N.19, no. 62405 the entries under Zn-Fe should each be moved one column to the right and Zn = 0.000. In Table XVI (pp. 152/153) the entry for Ni in SS-F (no. 10709) should read 0.08 per cent as given on pp. 146/147. The tables on pp. 186/187 contain some erroneous entries for Pb. In particular, the average Pb content in oxhide ingots from Ittireddu should read 0.012 per cent, not 0.27 per cent, which makes some of the discussion on pp. 178/179 obsolete because there is no significant difference any more between oxhide ingots and plano-convex ingots.
- 5. There is some confusion concerning the number of *bronze* ingots from Nuragic Sardinia. Vodret (1935) quotes analyses by Agrestini of three 'pani di bronzo studiati dal Pigorini' from Serra Ilixi. Two of them are listed with 12.2 and 14.2 per cent tin, respectively. However, since only three of the five ingots originally found at Serra Ilixi were saved for posterity (two were bought by a coppersmith who presumably destroyed them), it must be the same three ingots which in all later analyses (Vodret 1959; Gale and Stos-Gale 1987; Maddin and Merkel 1990) yielded only insignificant tin. In addition, Vodret (1959) lists two other ingots with 8.94 per cent and 15.6 per cent tin respectively. Since the first one, from Nuragus (Forraxi Nioi), contains also 5.8 per cent zinc, it is unlikely to be Nuragic in age. The last one, from Olmedo, is very atypical in size (151.7 g) and suspected to be re-melted bronze scrap rather than a primary ingot (Vodret 1959).

- 6. As to measured lead contents in the percentage range, it should be noted that this is above the solubility limit of lead in copper or bronze. Accordingly, a metallographic investigation of a polished section from this metal piece by Maddin and Merkel (1990 Fig. 39) showed segregated lead blobs which, moreover, are very inhomogeneously distributed. This makes all lead concentrations determined on very small samples of dubious significance. It may well account for the scatter in the lead concentrations 12.3 per cent and 6.56 per cent in duplicate analyses by Maddin and Merkel, and 4.55 per cent by us. This does not explain, however, the discrepant tin contents of duplicate samples reported by Maddin and Merkel.
  - 7. For a compilation of reliable data from Oxford, see Stos-Gale et al. (1997).
- 8. The two exceptions the plano-convex ingot from Ittireddu (no. 62417) and the one of uncertain type (no. 62405) are also differentiated by their high lead content, not only because of the different isotopic fingerprint of this lead.
- 9. A similar cylindrical object, with a chisel inserted, was found in the Nuragic hoard at Sardara (Ugas and Usai 1987).
- 10. Negative correlations between the concentrations of *trace* elements are notoriously suspect. More often than not, they are just chance associations with no statistical significance.
- 11. For recent discussions of metal hoards in general and useful definitions in particular, see Knapp et al. (1988) and Sommerfeld (1993).
- 12. It is clear that, if lead were introduced into the bronze via tin, this would make futile any attempts to employ the lead isotopic composition for tracing *copper* back to its ultimate source.
- 13. That a small percentage of lead should have been added to tin intentionally makes no sense from a metallurgical point of view. Conversely, there might have been every reason to sell cheap lead at the price of tin if that should have been possible without being found out!
- 14. Again, there is some confusion about the exact number. Gale and Stos-Gale (1987) report one ingot no. 5 from Baccu Simeone to have high abundance ratios and another one from the same site, and with the same sample number, to contain 'normal' lead. Subsequently, Gale (1989) lists only the 'normal' no. 5 but has another one (no. 27) whose isotopy is virtually identical with that of the old 'high' no. 5. There just might have been a mix-up.
- 15. The ore available from Terra Padedda was a low-grade ore with 2 per cent copper. It stands to reason, however, that the high-grade ores presumably utilized in antiquity and such low-grade ores as analyzed here are associated with different kinds and amounts of accessory mineral phases which host the trace elements in question. As a result, the nominal trace element concentrations of the copper listed here may be unreasonably high and misleading.

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#### ABSTRACTS

La composition chimique et l'isotope de plomb d'objets en cuivre et bronze de la Sardaigne Nuragique

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Nous présentons des données sur la composition chimique et l'isotope de plomb d'objets en cuivre et bronze de la Sardaigne Nuragique. La série d'échantillons comprend, entre autres, des objets des dépôts d'Arzachena (21 exempl.), Bonnanaro (10), Ittireddu (34), et Pattada (20), tous dans le Nord de la Sardaigne. À une exception près tous les fragments de lingots (49) consistent en cuivre non allié; seulement Ittiredu contient 11% d'étain. Par contre tous les artefacts (21) sont fabriqués d'un bronze standard, avec en moyenne 10.8% d'étain. Une douzaine de petits fragments d'épées du dépôt d'Arzachena, tous d'une taille plus ou moins uniforme, provient d'épées différentes. Leur contenu en étain de 1% permettrait de produire seulement des armes peu efficaces, ce qui confirme

leur interprétation archéologique comme épées votives. Les épreuves d'Arzachena sont remarquables pour leur grande diversité d'impuretés et leurs proportions abondantes d'isotope de plomb. Ceci les rend différentes de tous les autres échantillons analysés, et fait penser qu'il s'agit de métaux issus d'expérimentations d'extraction locales à base de minerais de cuivre d'origines différentes. L'isotopie de plomb et les impuretés, seules ou combinées, ne permettent pas de différencier les lingots oxyde des lingots plano-convexe (bun). La majorité des fragments de lingots ont une signature isotopique similaire à celle des mines de cuivre chypriote. Pour un nombre de lingots l'empreinte digitale isotopique est par contre entièrement compatible à celle des produits locaux. Parmi les objets en bronzes, aucun n'a une composition isotopique comparable aux lingots chypriotes. Tous contiennent du plomb local, ce qui suggère qu'ils sont de production locale. Du plomb provenant des mines de cuivre et plomb de la région d'Inglesiente-Sulcis en Sardaigne du sud-ouest et de Funtana Raminosa en Sardaigne centrale conviendrait parfaitement du point de vue isotopique.

# Kupfer und Bronze aus der Nuraghenzeit Sardiniens: chemische Zusammensetzung und Blei-isotopie

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Metallobjekte aus den nord-sardischen nuraghischen Hortfunden von Arzachena (21), Bonnanaro (10), Ittireddu (34) und Pattada (20) wurden auf ihre chemische Zusammensetzung und die isotopische Zusammensetzung ihres Bleis untersucht. Mit einer Ausnahme (11 Prozent Zinn) bestehen alle Barren und Barrenfragmente (49) aus unlegiertem Kupfer, alle Werkzeuge (21) aus Bronze mit einem mittleren Zinngehalt von 10,8 Prozent. Fünfzehn Schwertfragmente andererseits enthalten nur etwa 1 Prozent Zinn, zu wenig, als daß dadurch die Eigenschaften als Waffe gegenüber reinem Kupfer signifikant verbessert worden wären. Es handelt sich um Fragmente (einer großen Zahl) verschiedener Votiv-Schwerter. Acht der Arzachena-Proben von undefinierten Metallstückchen und Blechen sind in ihren Spurenelementgehalten und ihrer Blei-Isotopie extrem heterogen. Keine der Proben enthält Zinn, Arsengehalte (bis 5,3 Prozent) sind etwa zehnmal höher als in allen anderen Proben, und Silber (bis zu 3,2 Prozent) ist ungewöhnlich hoch. Die Proben sind chemisch und isotopisch völlig verschieden von dem mit ihnen in derselben Urne gefundenen Meißel und den Schwertfragmenten; möglicherweise handelt es sich dabei um Produkte lokaler Kupfergewinnung, bei der eine Reihe verschiedener Erze versuchsweise eingesetzt wurde. - Die Blei-Isotopie der meisten Barrenfragmente weist auf Zypern als Herkunftsort; einige wenige sind aber verträglich mit einer lokalen Produktion. Mit Ausnahme von zwei Schwertfragmenten findet sich die 'zypriotische' Signatur des Bleis in keinem der Endprodukte; deren Blei hat vielmehr die isotopische Zusammensetzung wie das der lokalen Kupfer- und Bleierze, insbesondere aus dem Südwesten der Insel und bei Funtana Raminosa im Zentrum. Es spricht alles dafür, daß diese Endprodukte lokalen Ursprungs sind.