

LEAD ISOTOPE AND TRACE ELEMENT ANALYSIS IN THE STUDY OF OVER A HUNDRED SOUTH INDIAN METAL ICONS*

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Technical investigations were made on 130 South Indian statuary images and a few miscellaneous artefacts mostly sampled from the Government Museum, Madras, India, and from the Victoria and Albert Museum and the British Museum, UK. Lead isotope investigations were attempted on 60 of these, and compositional analysis for 18 elements on 115, using inductively coupled plasma optical emission spectroscopy; thus, for 40 objects both lead isotope and trace element analysis was done. From the isotopic and elemental framework, insights are obtained into some art-historical problems of images and artefacts of the Pre-Pallava, Pallava, Chola (i.e., Vijayalaya Chola), Later Chola (i.e., Chalukya-Chola and Later Pandya), Vijayanagara (and Early Nayaka) and Later Nayaka (and Maratha) dynasties, spanning the Early Christian era to the nineteenth century, along with a few other regional styles. Inferences are also made regarding provenance of the lead and the early use of zinc and brass in the early historic period (c. fourth century BC–fourth century AD).

KEYWORDS: SOUTH INDIA, EARLY HISTORIC, MEDIEVAL, INDUCTIVELY COUPLED PLASMA OPTICAL EMISSION SPECTROSCOPY, LEAD ISOTOPE ANALYSIS, TRACE ELEMENT ANALYSIS, ICONS, COPPER ALLOY, LEAD, ZINC, COMPOSITION, PROVENANCE, STYLE

METHODOLOGY AND SCOPE OF TECHNICAL STUDIES

Introduction

The making of metal icons (of Hindu, Buddhist and Jaina affiliations) was a pan-Indian, and, indeed, pan-Asian phenomenon with several regional developments. Of these the lost wax image-making traditions in South India were perhaps the most prolific and longest lasting, continuing into the present day. While some metal icons are known from the early historic period (c. 300 BC–AD 500), it was from the early medieval period, that is, about the eighth century onwards, that fully developed traditions of making metal icons blossomed in South India, based on iconographic formulations of the *silpasastras*. Regional styles of bronzes have been debated based on affiliations with major dynasties such as Satavahana, Pallava, Chola, Vijayanagara or Nayaka. Of these, the Chola bronzes from modern Tamil Nadu state have received the most attention from connoisseurs including Rodin (1921) for the arresting iconography of Siva Nataraja, the Lord of Dance, and for their classic idiom.

While the iconography of South Indian images has been well studied, there is, nevertheless, a lack of consensus amongst art historians on stylistic, chronological and provenance attributions so that there is relevance in exploring objective criteria for making such attributions. With the exception of some early statuary from the Deccan, few bronzes from South India have been

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recovered from excavations, so that the chronology and provenance, for most, cannot be contextually determined. Few are inscribed and most are from hoards buried to protect against iconoclasm (Nagaswamy 1988) or are still in worship in countless shrines and temple vaults (Barrett 1965, preface). Some of the problems concerning the objective stylistic dating of South Indian bronzes have been explored by Schwindler (1975, 1–19) who mentioned that dates for less than half a dozen bronzes could be accepted totally without reservation. The existing methods of dating by comparisons with stone sculpture or by drawing inferences from dedicatory temple inscriptions, if they existed, can induce uncertainty, while the portability of metal icons confuses the identification of regional styles. While most scholarship has concentrated on the Chola period (c. tenth century AD), there is uncertainty regarding bronzes of earlier and later periods.

Technical studies are increasingly reported in art-historical discourse to evaluate typological and stylistic classifications of Asian artefacts, such as those from the Far East and China (Sayre *et al.* 1992), and from the Indo-Tibetan region (Reedy 1986) which used a statistical approach. However, few studies have been made on South Indian bronzes. The analyses of medieval South Indian images include one by Paramasivan (1941, 54) of four images by gravimetry, while Johnson (1972) analysed four of the twelfth century AD by spark source mass spectrometry. Werner (1972) analysed by emission spectrography 330 copper alloys from India, South-east Asia and East Asia, including 77 South Indian images; however, these belong mostly to the eighteenth to twentieth centuries (*ibid.*, 138–49), with only two or three earlier pieces.

The investigation reported here is fairly representative with sampled images forming a continuous sequence from the early to late medieval periods, with a few from the early historic period. The sampled collection of about 130 bronzes included several important images, about half each from the Victoria and Albert Museum, London, and the Government Museum, Madras (including several published in Srinivasan 1963), and the rest from miscellaneous collections including the British Museum, London. The sampled images were predominantly from the region of modern Tamil Nadu which sustained by far the most prolific image-casting tradition and were hence the most accessible, while some bronzes from other regions were also included for comparison. For each stylistic group an attempt was made to sample a stylistically coherent 'control' group of bronzes for which there was general consensus of attributions using conventional art history against which comparisons could be made. However, since the samples depended on objects which could be accessed for study, the number of objects from each tentative stylistic group was uneven. While aspects concerning major alloy composition are reported elsewhere, this paper focuses on the usefulness of lead isotope and trace element analysis in resolving some art-historical problems, based on which as much as half the sampled collection was re-catalogued.

Notes on methodology of compositional and lead isotope analysis

While thermoluminescence dating or petrographic analyses may be attempted on clay cores of hollow cast images, South Indian images are overwhelmingly solid cast, unlike images from northern India, so that choices for technical investigations were limited to metal analysis.

Elemental compositional analysis has been used to group together artefacts based on empirical observations of elemental similarities. These can be due to shared histories of metallurgical processing (which may include geological source) and alloying, thus suggesting that the objects were probably made at the same time and place. Although the composition of

intentionally alloyed elements can vary randomly, the intrinsic composition of trace elements and their variation can be archaeologically significant. However, the concentrations of trace elements can vary from ore to processed metal due to the numerous variables governing trace element partitioning during metal processing from the stages of smelting, refining and alloying and contributions from fluxes and refractories (some aspects of which are explored by Berthoud *et al.* (1980), Merkel (1983) and Tylecote *et al.* (1977)) and due to the variations in the geochemistry of the deposit, and added to these are, of course, analytical uncertainties in measuring trace elements.

Compared to elemental analysis, lead isotope analysis is a more rigorous technique in archaeometry since the relative proportions of isotopes from ore to artefacts are not measurably affected by chemical or pyrometallurgical processes, while there is evidence to show that lead isotope ratios for different ore deposits are characteristic (Begemann *et al.* 1989; Brill and Wampler 1965; Gale and Stos-Gale 1982). Lead isotope analysis has been used for exploring groupings between archaeological artefacts and possibly relating them to ore sources if lead isotope data on ore sources are available, especially in the case of Mediterranean archaeology (*ibid.*), or at least excluding particular ore deposits as the source of metal if the lead isotope ratios of the artefact do not match (Stos-Gale 1993).

Lead isotope investigations may be also used with alloyed or leaded artefacts to separate batches of artefacts with probable similar histories of manufacture and, therefore, similar conditions of smelting and alloying of lead. Isotopic matching of artefacts may be due to the lead coming from a single common source, or due to the objects having been made from the same batch of metal containing lead from mixed sources which may have archaeological significance for shared geography or chronology. Indeed, Mabuchi *et al.* (1985) and Sayre *et al.* (1992) have, thus, used lead isotope investigations to separate chronological groups of Chinese and Japanese bronzes and brasses, without dwelling on problems of geological provenance. There is significance in using lead isotope analysis in conjunction with trace element analysis, as pointed out by Cherry and Knapp (1991, 99), which can be complementary, if not related, based on shared metal processing and/or provenance.

Although mass fractionation for lead has been generally thought to be negligible in relation to the high atomic mass of lead, a concern in exploring the usefulness of lead isotope data for archaeological provenancing has been the possibility that lead isotopes may fractionate significantly enough during geological, extraction or manufacturing processes to affect the lead isotope methodology (Budd *et al.* 1995). However, such fractionation has been shown to be minimal (Gale and Stos-Gale 1996).

For the images, all samples were taken from the main body of the image, that is, the main casting. Compositional analysis was undertaken using inductively coupled plasma optical emission spectroscopy (ICP-OES) for simultaneous multi-element determination at major, minor and trace concentration levels with low detection limits. Such simultaneous analysis also ensured that all artefacts could be analysed under one set of standardized conditions which made comparisons between the results more valid. Procedures for copper-alloy analyses by ICP-OES were those developed and standardized at Royal Holloway and Bedford New College, Egham, by Dr Nick Walsh (Walsh and Thompson 1989) and Nigel Blades (*pers. comm.*). Eighteen elements were analysed using a Philips combined simultaneous sequential ICP-OES spectrometer (Cu, Zn, Pb, Sn, Fe, Ni, As, Sb, Bi, Co, P, S, Cr, Mn, V, Cd, Ag, Au). Samples were made up in solution in a 1/1000 dilution factor, with 100% in the sample equivalent to 1000 ppm in solution with a 10% acid matrix. The final results were drift corrected, weight corrected and

corrected mathematically for optical interferences of copper on lead and zinc. Drift correction was done by calculating the drift in the composition of the standard (BAS standard 183/4) for every five analyses and scaling the composition of samples accordingly.

However, atomic absorption spectroscopy (AAS) was also used to verify the results of ICP analysis, especially for certain elements for which AAS has good sensitivity and detection limits, such as silver, zinc and copper, and for which analyses by ICP-OES were thought to be prone to matrix, drift or interference effects. AAS analysis was done using a Pye Unicam SP-9 flame atomic absorption spectrophotometer at the Wolfson Technical Laboratories, Institute of Archaeology, University of London, using procedures described by Hughes *et al.* (1976, 20). The same solution made up for ICP analysis was diluted to a 1/400 dilution factor for analysis by AAS which shows good sensitivity and linearity in the response curve for low concentrations of solute. As for the accuracy of trace element measurements for ICP analysis, the error in measurement using SRM UZ-40 of 0.22 wt% Fe and 0.22 wt% Ni over different measurements at intervals of samples was no more than 4–10 wt%. For AAS analysis the error in measurement using SRM UE-51 was about 10 wt% at 0.235 wt% Fe so that the results were comparable.

Lead isotope analysis was done using thermal ionization mass spectrometry (TIMS) facilities at the Isotrace Laboratory, Research Laboratory for Archaeology and the History of Art, Oxford University, using equipment and methodology standardized by Noël Gale and Zofia Stos-Gale (Stos-Gale 1993), which allows for highly accurate measurements of lead isotopes with the percentage standard error being no more than 0.01% over 60–100 measurements of three independent ratios. Lead was separated from the samples by anodic deposition and was deposited on a rhenium filament as a lead nitrate salt and introduced into a fully automated multicollector VG 38-54-30 double focusing thermal ionization mass spectrometer. Recent lead isotope measurements made by laboratories using comparable methods and NBS standards are thought to fall within the range of experimental uncertainty of $\pm 0.1\%$ of the mean ratios (Stos-Gale 1993; Stos-Gale *et al.* 1995). Taking together recent advances in the use of ultraclean facilities for separation of lead and extraction of very small amounts of lead in the sample, the mounting of the sample using the silica gel and phosphoric acid technique minimizing mass fractionation during ionization, the use of multicollectors and the use of internationally accepted NBS standards (such as NBS SRM 981) to correct for mass fractionation, it may be noted that isotopic ratio measurements made prior to these advances, that is, before 1985, would not be seen to be of sufficient accuracy to be compatible with recent data from reliable laboratories. The results of lead isotope analysis undertaken on South Indian images and copper alloys are presented in Table 1, while results of lead isotope investigations on Indian galenas from the data bank of the Isotrace Laboratory as well as some obtained by the author are in Table 2. Due to limits on access to equipment, and in some cases sample sizes, elemental analysis could not be done on all images for which lead isotope analysis was done.

Use of a combined lead isotope and trace element perspective

A majority of the images analysed were leaded tin-bronzes, while some were brasses. Although some trends in the use of alloys over different periods were noted, it was generally found that the major element trends were not that useful for exploring groupings amongst artefacts and this may be expected since intentional alloying is an external variable. More useful for stylistic characterization were the trace element trends, particularly Co, Ni, Sb, As and Bi. (The results of simultaneous ICP-OES analysis of the investigated South Indian images and copper alloys for 18

Table 1 Lead isotope ratios of South Indian images and miscellaneous copper alloys (c. AD 200–1800)

No.	Accession no.	Description	Findspot	Collection*	Pb ^{207/206}	Pb ^{208/206}	Pb ^{206/204}
1	IM-13-1934	Vishnu	Tamil Nadu	V&A	0.83509	2.07860	18.83000
2	IM-14-1938	Elephant with rider	Uncertain	V&A	0.86552	2.10866	18.08700
3	721/74	Jaina image, inscribed (13th century AD)	Gidangil, Tamil Nadu	MM	0.86188	2.10780	18.23969
4	Lead coin	Pallava (c. AD 600–800)	Tamil Nadu	Misc.	0.86363	2.11150	18.27040
5	Zinc ingot/coin	Inscribed (4th–5th century AD)	Deccan	Misc.	0.84857	2.08775	18.43100
6	IM-9-1924	Octagonal bowl	Krishna delta, Andhra Pradesh	V&A	0.85093	2.09111	18.38400
7	OA-1830-6-12-4	Bodhisattva Tara, gilt	Batticoala, Ceylon	BM	0.85069	2.09431	18.37000
8	IPN-2639	Buddha, gilt	Uncertain	V&A	0.85254	2.09555	18.39355
9	OA-1957-2-11-1	Vishnu	South India	BM	0.85303	2.09617	18.37800
10	OA-1974-12-9-2	Parvati	Deccan	BM	0.85584	2.09682	18.44900
11	53/38	Siva Nataraja	Kuram, Tamil Nadu	MM	0.85648	2.10241	18.35150
12	43-IS-1887	Kiratarjunamurti	Kerala	V&A	0.85261	2.10064	18.34200
13	IM-70-1935	Parvati	Tinnevelly district, Tamil Nadu	V&A	0.85437	2.10145	18.40730
14	315/55	Rama	Tiruvelangadu, Tamil Nadu	MM	0.85377	2.10375	18.38590
15	46	Bodhisattva Maitreya	Nagapattinam, Tamil Nadu	MM	0.85465	2.10791	18.36200
16	260	Seated Parvati	Vadakapoyur, Tamil Nadu	MM	0.85539	2.11413	18.34550
17	IPN-2657	Rukmini	South India	V&A	0.85502	2.11420	18.33911
18	1326-1855	Buddha	Uncertain	V&A	0.85534	2.11511	18.33740
19	IM-118-1924	Karaikal Ammayar	Tanjavur district, Tamil Nadu	V&A	0.85419	2.11631	18.47400
20	IS-204-1959	Kaliya Krishna	Tamil Nadu	V&A	0.85461	2.11651	18.38998
21	IM-6-1924	Narasimha	Avadiyakovil, Tamil Nadu	V&A	0.85503	2.11657	18.37370
22	IM-72-1935	Parvati	Tinnevelly district, Tamil Nadu	V&A	0.85589	2.11672	18.32210
23	IM-71-1927	Rama	Madura, Tamil Nadu	V&A	0.85554	2.11770	18.35326
24	47-4/36	Parvati	Kudiakadu, Tamil Nadu	MM	0.85628	2.11771	18.32400
25	IM-1-1934	Rama	Tamil Nadu	V&A	0.85609	2.11803	18.36223
26	275-09	Hanuman	Ceylon	V&A	0.85574	2.12045	18.40033
27	40/36	Siva Nataraja	Melaperumbalam, Tamil Nadu	MM	0.85651	2.11243	18.35166
28	OA-1967-10-17-1	Siva as Bhairava	Western Deccan	BM	0.85401	2.11414	18.37000
29	8/27	Jaina Tirthankara	Polur, Tamil Nadu	MM	0.85803	2.11740	18.32300
30	OA-1967-7-27-1	Siva as hunter	Deccan	BM	0.84371	2.10228	18.61300
31	223	Somaskanda	Tiruvelangadu, Tamil Nadu	MM	0.84190	2.10748	18.61918
32	IM-137-1927	Bhudevi	Coimbatore district, Tamil Nadu	V&A	0.84298	2.08730	18.65630
33	240	Siva Nataraja	Okkur, Tamil Nadu	MM	0.84340	2.08488	18.60200

Table 1 (continued)

No.	Accession no.	Description	Findspot	Collection*	Pb ^{207/206}	Pb ^{208/206}	Pb ^{206/204}
34	OA-1905-12-18	Jaina Tirthankara, inscribed (9th century AD)	Buddhapad, Andhra Pradesh	BM	0.84352	2.08879	18.65500
35	220	Somaskanda	Needur, Tamil Nadu	MM	0.84370	2.09003	18.66530
36	233	Siva Nataraja	Belur, Tamil Nadu	MM	0.84395	2.08795	18.62783
37	234	Siva Nataraja	Velankanni, Tamil Nadu	MM	0.84411	2.08634	18.58200
38	IM-149-1927	Devi	Coimbatore district, Tamil Nadu	V&A	0.84426	2.08889	18.61991
39	IM-71-1935	Siva Nataraja	Tinnevely district, Tamil Nadu	V&A	0.84444	2.08587	18.57100
40	IM-75-1935	Sambandar	Tinnevely district, Tamil Nadu	V&A	0.84518	2.09140	18.62210
41	37	Seated Buddha	Nagapattinam, Tamil Nadu	MM	0.84558	2.09112	18.57672
42	280	Kali, inscribed (c. AD 1000)	Senianvidudi, Tamil Nadu	MM	0.84562	2.08846	18.57476
43	336	Devotee	Kandarakottai, Tamil Nadu	MM	0.84570	2.08619	18.56540
44	273	Mahishasuramardini	Turaikadu, Tamil Nadu	MM	0.84606	2.08947	18.58000
45	IM-2-1934	Siva Nataraja	Tanjavur district, Tamil Nadu	V&A	0.84680	2.09262	18.57200
46	IM-158-1929	Somaskanda	Tinnevely district, Tamil Nadu	V&A	0.84736	2.09333	18.56875
47	238	Siva Nataraja	Kankoduvannittavam, Tamil Nadu	MM	0.84750	2.09321	18.56870
48	450/61	Tripurantaka	Tirukodikaval, Tamil Nadu	MM	0.84574	2.09100	18.59786
49	495/61	Seated Vishnu	Orukannankadu, Tamil Nadu	MM	0.84610	2.08899	18.53100
50	IS-8-1989	Yakshi (c. AD 200)	Deccan	V&A	0.84393	2.09338	18.65076
51	.495/65	Rama	Tanjavur district, Tamil Nadu	MM	0.84768	2.09246	18.50400
52	19	Buddha	Nagapattinam, Tamil Nadu	MM	0.84860	2.09613	18.53755
53	720/73	Parvati	Tiruvengimalai, Tamil Nadu	MM	0.83927	2.08689	18.70320
54	OA-1965-12-14-1	Laxmi	Kerala	BM	0.84066	2.08639	18.72400
55	OA-1969-12-16-1	Siva Nataraja	Tanjavur district, Tamil Nadu	BM	0.84266	2.08188	18.63200
56	752/75	Siva Nataraja	Kunniyur, Tamil Nadu	MM	0.84113	2.08239	18.64400
57	OA-1957-10-12-1	Jaina Ambika	Deccan	BM	0.84211	2.08285	18.62100
58	930-IS	Jaina image, inscribed (c. AD 1250)	Gujarat, western India	V&A	0.90269	2.15615	17.35980
59	289	Ganesha	Nellore, Andhra Pradesh	MM	0.95594	2.20460	16.30100
60	IS-44-1966	Buddha	Nagapattinam, Tamil Nadu	V&A	0.90951	2.15287	17.31757

* V&A: Victoria and Albert Museum, London; MM: Government Museum, Madras (Chennai); BM: British Museum, London; Misc.: miscellaneous.

Table 2 Lead isotope ratios of some Indian ores and slags

No.	Location	Region	Pb ^{208/206}	Pb ^{207/206}	Pb ^{206/204}	Sample type
1	Agnigundala	Andhra	2.17031	0.94146	16.699	Ore
2	Agnigundala	Andhra	2.16910	0.94075	16.727	Ore
3	Agnigundala	Andhra	2.16874	0.94079	16.721	Ore
4	Agnigundala	Andhra	2.17334	0.94176	16.745	Ore
5	Agnigundala	Andhra	2.16646	0.94023	16.717	Ore
6	Agnigundala	Andhra	2.16899	0.94080	16.728	Ore
7	Agnigundala	Andhra	2.16722	0.94053	16.708	Ore
8	Agnigundala	Andhra	2.17385	0.94212	16.726	Ore
9	Agnigundala	Andhra	2.17206	0.94186	16.716	Ore
10	Zawar	Rajasthan	2.19244	0.94290	16.730	Pb slag
11	Mochia, Zawar	Rajasthan	2.20495	0.95004	16.493	Ore
12	Mochia, Zawar	Rajasthan	2.19909	0.94345	16.640	Ore
13	Zawar	Rajasthan	2.20259	0.94806	16.575	Pb slag
14	Balaria, Zawar	Rajasthan	2.20051	0.94685	16.614	Ore
15	Balaria, Zawar	Rajasthan	2.21119	0.95244	16.461	Ore
16	Zawar	Rajasthan	2.19648	0.94324	16.661	Ore
17	Zawar Mala	Rajasthan	2.20195	0.94760	16.604	Ore
18	Balaria, Zawar	Rajasthan	2.20800	0.95200	16.470	Ore
19	Balaria, Zawar	Rajasthan	2.19100	0.94600	16.530	Ore
20	Balaria, Zawar	Rajasthan	2.19600	0.94600	16.550	Ore
21	Balaria, Zawar	Rajasthan	2.20500	0.95100	16.400	Ore
22	Balaria, Zawar	Rajasthan	2.20230	0.95048	16.397	Ore
23	Dariba	Rajasthan	2.21200	0.96200	16.100	Ore
24	Dariba	Rajasthan	2.20500	0.95900	16.160	Ore
25	(Rajpur) Dariba*	Rajasthan	2.2345	0.96543	16.061	Ore
26	Dariba	Rajasthan	2.20876	0.94961	16.379	Pb slag
27	Dariba	Rajasthan	2.21438	0.95271	16.307	Pb slag
28	Ambaji	Gujarat	2.14843	0.90079	17.313	Slag
29	Ambaji	Gujarat	2.14883	0.90085	17.318	Slag
30	Ambaji	Gujarat	2.09020	0.86951	18.003	Ore
31	Ambaji*	Gujarat	2.1596	0.90405	17.315	Ore
32	Ambadongar*	Deccan	2.07	0.84	18.57	Ore

Galena samples 1–9 were collected by the author through Hindustan Zinc, Bandlamottu, and analysed at IsoTrace Laboratory, Research Laboratory for Archaeology and the History of Art, Oxford Univ. Results on samples 10–24 and samples 26–30 were given courtesy of Dr Z. Stos-Gale, of the above institution, with ore and lead slag samples obtained from Dr Paul Craddock, Dept. of Scientific Research, British Museum. Results on sample 25 (galena) and sample 31 (chalcopyrite with Pbs and Zns) are from Hegde and Ericson (1985) and on sample 32 (galena) from Venkatasubramanian *et al.* (1982).

* Samples not analysed at IsoTrace Laboratory.

elements undertaken at Royal Holloway and Bedford New College, Egham, are available on request to the author and are also listed in the author's unpublished Ph.D. thesis (Srinivasan 1996.) The scatter plots of Co versus Ni (Fig. 1 (upper)), Bi versus As (Fig. 1 (lower)) and Sb versus Co indicated that for many bronzes from the 'control group' of well-dated bronzes, the values of Co, Ni, As, Bi and Sb showed certain typical patterns. These trends or patterns are summarized in the 'Appendix' (see 'Trace element categories').

The significance of trace element groupings lies in the fact that ore deposits can exhibit trace element characteristics based on geochemistry. Indeed, inter-element ratios of trace elements are

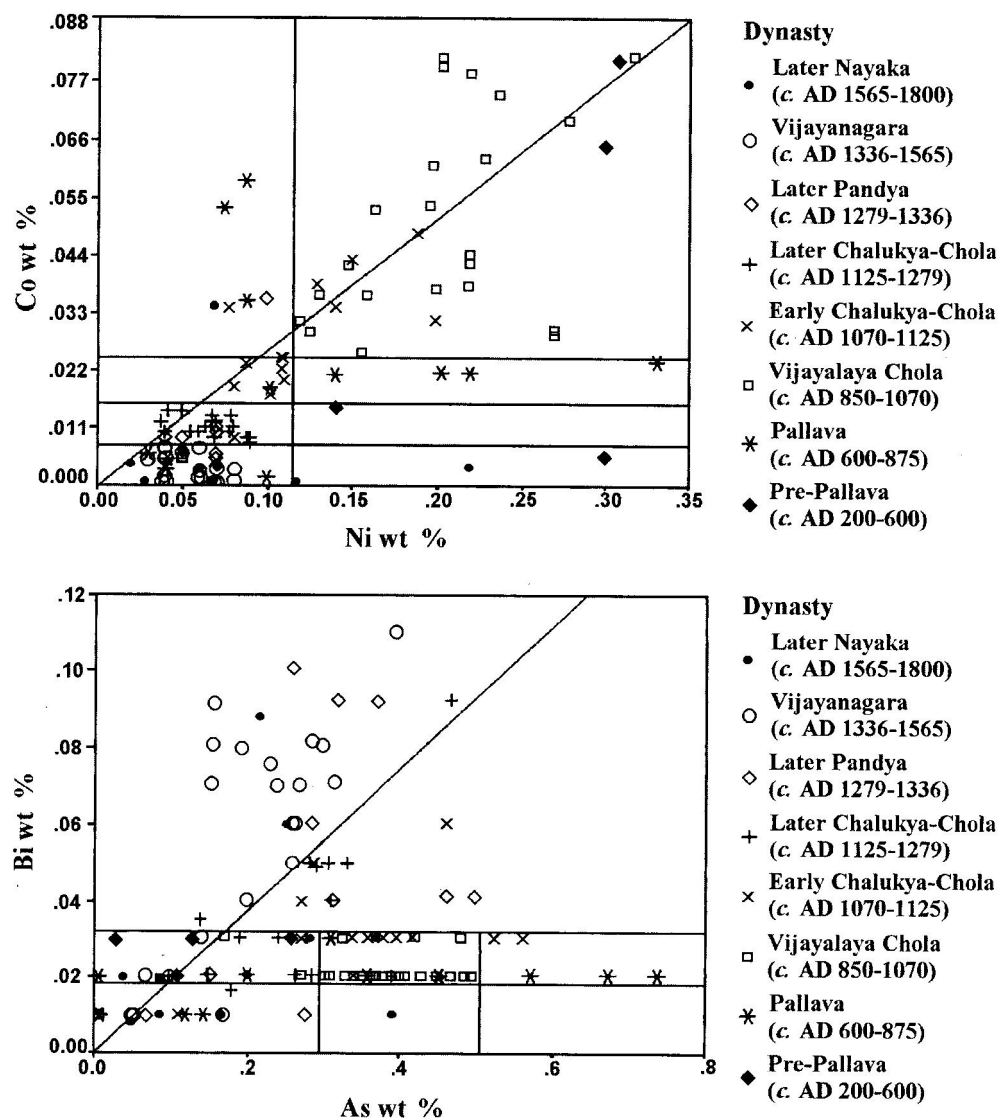


Figure 1 Scatter plots of (upper) cobalt versus nickel content (wt%) and (lower) bismuth versus arsenic content (wt%) in analysed South Indian bronzes and copper alloys, classified by dynastic group. The perpendicular lines indicate boundaries of absolute values of elements for bronzes of different periods as indicated in the 'Appendix' ('Trace element categories'). The diagonals are arbitrarily drawn to indicate (upper) that the Co and Ni values of Vijayalaya Chola bronzes show some linear variation, (lower) that Bi/As ratios for most Vijayanagara and Early Nayaka bronzes are greater than 0.19.

often used in geological literature to explore characteristics of ore deposits and may, thus, be useful to discriminate empirically between groups of contextually related artefacts. Loftus-Hills and Solomon (1967, 228-42) suggest that Co/Ni ratios are useful as discriminators of different mineral and ore deposits and that Co/Ni ratios can be greater or less than one depending on

whether the ore deposits are volcanic or sedimentary. As another example, Mookherjee and Philip (1979, 46) found that nickel and cobalt can vary linearly in the case of sulphidic copper ores from Ingaldhal. Studies on partitioning of elements during copper smelting indicate that Ni, Co and Ag are amongst the most chalcophilic (Merkel 1983; Tylecote *et al.* 1977), while an attempt has been made to quantify the relationship of these elements from ores to processed metal (Berthoud *et al.* 1980). Despite losses, As, Sb and Bi would also be sufficiently retained in the processed metal from ore although As and Sb can be altered in subsequent processes (Tylecote *et al.* 1977, 329, 324).

Although Ag and Au are amongst the most chalcophilic elements and, hence, amongst the most useful for characterization studies, as used in Gale (1991), these were not so useful in this study as there was a fair amount of random scatter in the Au/Ag and Co/Ag plots. One explanation could be the fact that, as told by a traditional icon manufacturer or *Sthapati* from Swamimalai, Tanjavur district, interviewed by the author in 1990 (Srinivasan 1996, 101), South Indian bronzes were known as *Pancha-loha* or five-metalled icons because minor additions of gold and silver could be made during casting, of about 100–500 mg, added for ritual purposes. The Swamimalai craftsman interviewed in mid-1990 also mentioned that the addition of these small amounts of gold or silver were particularly made into the runner behind the face of the casting since it was thought to enhance the lustre of the face of the image (*ibid.*). But this amount of gold or silver may not be detected as a deliberate addition as it would constitute only about 0.001 wt% of a typical solid cast copper-alloy image of an average weight of 50 kg. (Field investigations made by the author in mid-1990 of traditional icon manufacture at Swamimalai, and comparisons with ritual prescriptions in ancient treatises such as the *Manasara* and the *Manasollasa*, along with technical examination of medieval images, are reported in Srinivasan (1996).)

The use of scatter plots and trends in the absolute values of trace elements of Ni, Co, Sb, As and Bi was found to be adequate for discriminating between groups of bronzes at this stage of study. Although preliminary k-means cluster analysis was attempted, it did not give clear results although some weak trends could be observed for variables of Ni, Co, Sb, As and Bi content with an optimum of ten clusters (using SPSS-Windows 6.0 software) (Srinivasan 1996, 237–9). The usefulness of multivariate statistics was limited because the sample was not balanced, there being several images from one stylistic group and very few from others. Since many of the attributions were in the first place uncertain, there were not large enough control groups to use statistical methods meaningfully both for trace element and lead isotope analysis. Furthermore, as suggested before, the data on two element ratios can have actual geological or metallurgical significance, and these essentially arbitrary relationships would not perhaps be so easily picked up by statistical methods. For instance, as indicated later, a trend was noted for one group of artefacts in the Bi/As ratios being greater than a certain value. Hauptmann (1989, 121) also uses an arbitrary but empirically observed relationship of Co/Ni versus Sb/As in the typological classification and discrimination of artefacts. Finally, for the ultimate objective of intimate re-assessment of style of individual bronzes the scatter plots proved more useful, while Cherry and Knapp (1991) point out that statistical discrimination procedures required prior knowledge of groups to be successful. Indeed, Begemann *et al.* (1989), Cherry and Knapp (1991, 99), Hauptmann (1989, 121), Hauptmann *et al.* (1992, 17, 25) and Gale (1991, 210, 218) have used trace element ratio scatter plots for discriminating between artefacts; in many cases these were found to complement findings from lead isotope analysis.

The first consideration for lead isotope groupings based on provenance is to ascertain the

expected range of isotopic variation for ore deposits, and to know the constancy of the lead isotope composition in a given mine. Studies on European and West Asian ores indicate that, for ore deposits with a relatively simple geological history, the range of lead isotope ratios falls within $\pm 0.25\%$ of the mean for that group (Stos-Gale 1993). In fact, the range of experimental uncertainty for the measurement of lead isotope ratios for galena samples from six mines in Anatolia was less than $\pm 0.1\%$ (Hauptmann 1989, 273). In order to interpret the lead isotope data on artefacts, the spread in lead isotope ratios for individual mines and within a general mining region was investigated for the Indian ore deposits. For this, the existing data on ore sources at the Isotrache Laboratory, including western Indian deposits such as Zawar and Dariba (with samples provided by P. T. Craddock), analyses of western Indian ores by Ericson and Shirahata (1985) and Hegde and Ericson (1985), were inspected together with new analyses made at Isotrache on ore samples obtained by the author from the Bandalamottu mine in southern India. It was seen that in the Indian context the spread of lead isotope ratios is much larger than that usually found elsewhere in the world, which could be due to the fact that Indian ores are amongst the oldest in the world, so that new as well as old formations could give a range of ratios. The spread of ratios within a mining area (i.e., covering several mines) was found to be as high as $\pm 1-\pm 2\%$ of the mean for some western Indian ore deposits such as Ambaji (plotted in Fig. 2; see also Table 2), although for some individual mines the variations could be within $\pm 0.25-\pm 0.6\%$ of the mean and even within $\pm 0.25\%$ of the mean as seen for the Bandalamottu mine.

For exploring groupings within lead isotope ratios, Stos-Gale *et al.* (1995) use diagrammatic

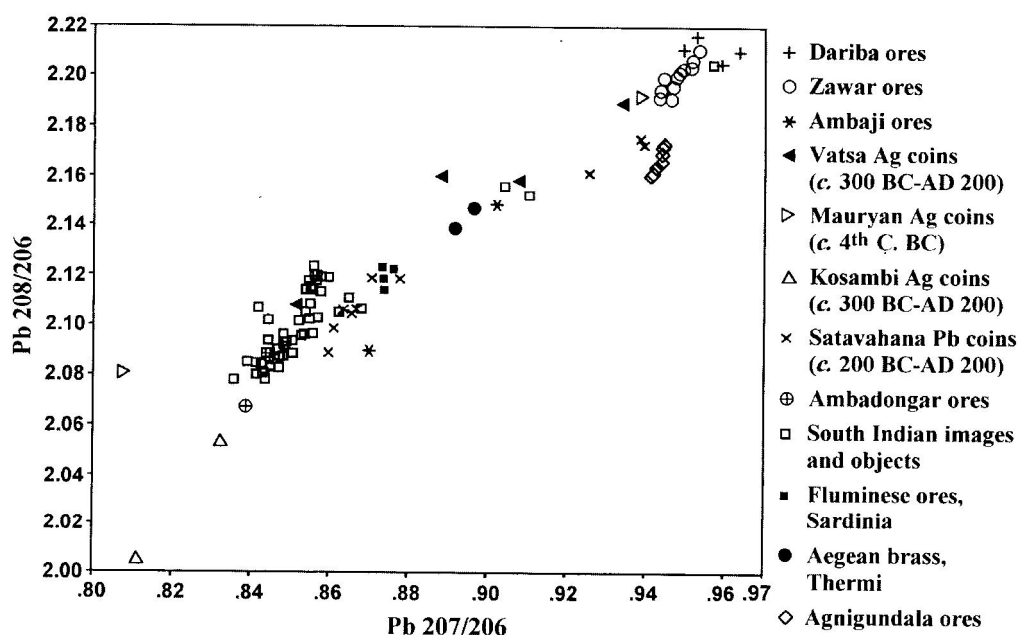


Figure 2 Scatter plot of lead isotope ratios of $Pb^{208/206}$ versus $Pb^{207/206}$ for investigated South Indian images and copper alloys, plotted against some ores and other artefacts. These include Satavahana coins (Turner 1985) and Mauryan coins (Isotrache data bank, courtesy of Z. A. Stos-Gale and P. T. Craddock), Hellenistic brass (Stos-Gale 1992) and Sardinian ores (Stos-Gale *et al.* 1995). Ratios for all ore samples were analysed at the Isotrache Laboratory, except for Ambadongar (Venkatasubramanian *et al.* 1982).

representations of two sets of ratios, that is, $Pb^{208/206}$ versus $Pb^{207/206}$ and $Pb^{206/204}$ versus $Pb^{207/206}$. This study used this schema as well as the plot of $Pb^{206/204}$ versus $Pb^{208/206}$ and $Pb^{207/206}$ versus $Pb^{208/206}$ for added visual perspectives and objects were grouped only if there was sufficient matching in all four plots. Figure 3 shows the three ratio plot of $Pb^{208/206}$ versus

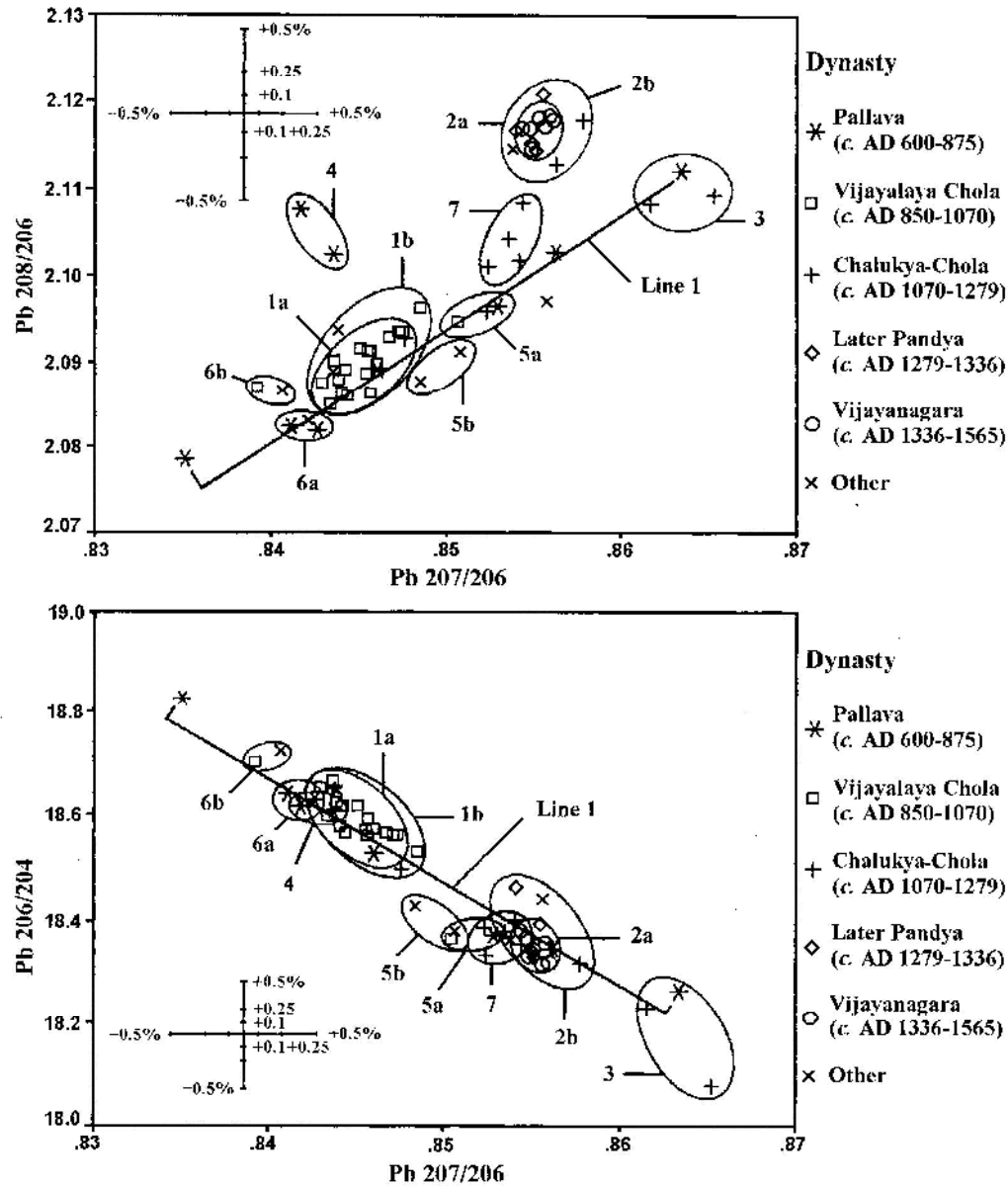


Figure 3 Exploded scatter plots of lead isotope ratios of South Indian group of artefacts, classified by dynastic group, for (upper) $Pb^{208/206}$ versus $Pb^{207/206}$ and (lower) $Pb^{206/204}$ versus $Pb^{207/206}$. The ellipses are arbitrarily drawn to indicate the groups (1-7) set out in the 'Appendix' ('Lead isotope groups') based on the spread in isotopic ratios. Line 1 is arbitrarily drawn to indicate a linear trend noted for one stylistic group.

$Pb^{207/206}$ and $Pb^{206/204}$ versus $Pb^{207/206}$, and the groups indicated on the figure correspond to those defined in the 'Appendix' (see 'Lead isotope groups'), based on the isotopic spread.

Based on certain principles concerning lead isotope geochemistry, groupings within artefacts were explored as follows. If the lead isotope ratios of a significant group of artefacts fall within $\pm 0.25\%$, or 0.5% of the mean, they are highly likely to come from a single source or mine so that the groups could, hence, be archaeologically significant, such as group 1 and group 2. These are divided into core groups 1a and 2a for which the lead isotope ratios fall within $\pm 0.25\%$ of the mean, and the peripheral groups 1b and 2b where the lead isotope ratios fall on the periphery of the core groups within a range of $\pm 0.5\%$ of the mean. For the latter it is inferred that the lead (from alloyed lead or intrinsic trace lead in unalloyed copper) probably came from the same mining area or mine with a higher internal range of lead isotope ratios or that there was some mixing of lead whereby lead isotope ratios from one source prevailed because it was in higher concentrations. The latter can perhaps also be inferred for the minor isolated 'satellite' groups of no more than two to three artefacts which lie close to group 1 such as groups 6a, 6b, 5a, 5b. The existence of such minor groups of artefacts may also be explained if the objects were cast or recycled from the same batch of metal with lead from mixed sources, or from mixing in a constant proportion, which again has archaeological implications. (Of these minor groups, group 5a consists of copper images and 5b of a brass and zinc ingot, unlike the leaded bronzes of group 1. It is possible that, even if the copper, lead and zinc come from the same source, there can be a slight difference in the associated lead isotope ratios as seen for chalcopyrite and galena ores from Dariba (Hegde and Ericson 1985).) For objects made with lead mixed randomly from two sources they will have lead isotope ratios which are a linear combination of the lead isotope ratios of the two sources, roughly plotting along a straight line connecting the two sources as seen in line 1 (Fig. 3). These ideas were used in exploring groupings between artefacts. Based on the above principles, the scatter plots of lead isotope ratios of $Pb^{208/206}$, $Pb^{207/206}$ and $Pb^{206/204}$ were found to show certain patterns of isotopic matching or relationships, within which internal stylistic coherence could be discerned; this is discussed further below.

It is also noted that the patterns from lead isotope analysis and trace element analysis were somewhat consistent with each other for many of the 40 objects for which both analyses were performed (although this could not be attempted for all artefacts). This is visually indicated in the combined three-dimensional scatter plots of $Pb^{208/207}$ versus $Pb^{207/206}$ versus Ni/Co and of $Pb^{208/207}$ versus $Pb^{207/206}$ versus As/Bi (Fig. 4). This greatly strengthens the case for compatibility between the stylistic and analytical evidence. It may also be inferred that, if both lead isotope ratios and trace element trends are matched, it is likely that both lead and copper sources were the same for that group of artefacts since the former relates to lead sources and the latter mainly to copper sources.

IMPLICATIONS FOR STYLISTIC STUDY

For the lead isotope ratio plots of $Pb^{208/206}$ versus $Pb^{207/206}$ and $Pb^{206/204}$ versus $Pb^{207/206}$ and trace element plots of Co versus Ni (Fig. 1 (upper)), Bi versus As (Fig. 1 (lower)) and Sb versus Co, scatter plots were made using dynastic chronology and style as well as findspot/provenance as indices or markers for the bronzes. It was found that it was not necessarily findspot/provenance which gave the most coherent patterns, as might have been expected, but dynastic chronology and style, suggesting that different sources were widely used during different periods. (Although some bronzes of other regional styles were included for comparative study,

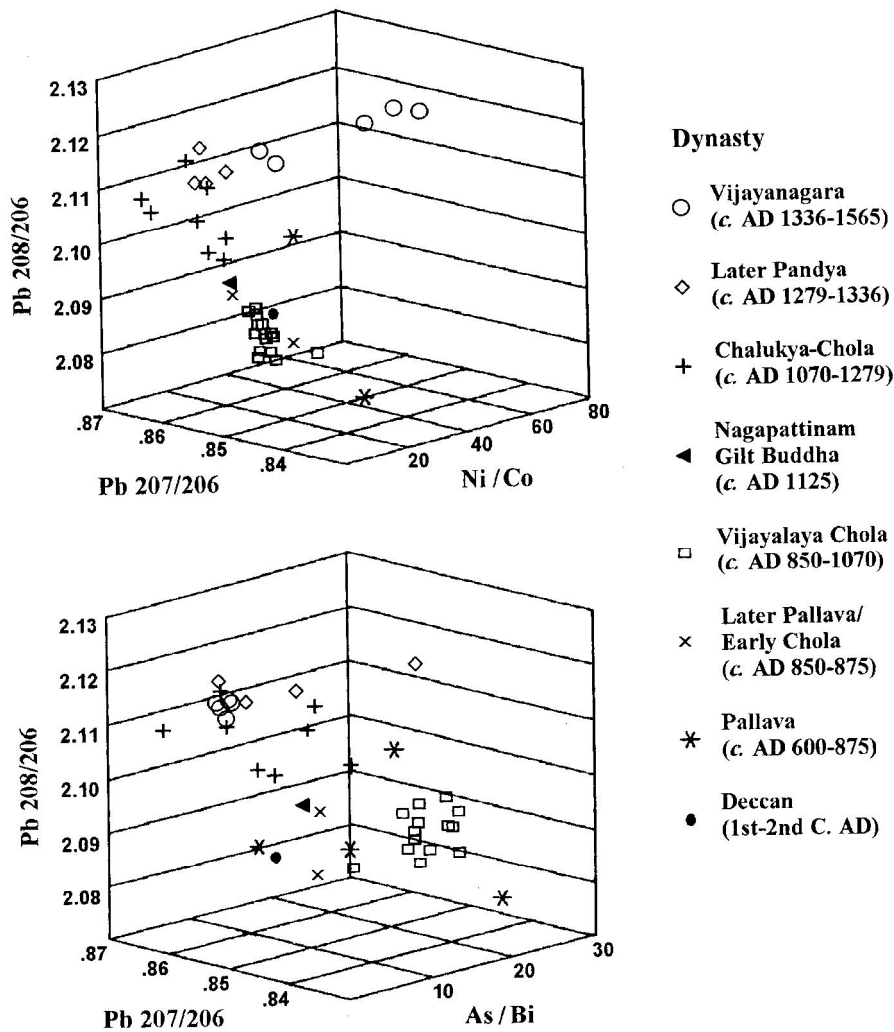


Figure 4 Combined three-dimensional scatter plots of (upper) $Pb^{208/206}$ versus $Pb^{207/206}$ versus Ni/Co and (lower) $Pb^{208/206}$ versus $Pb^{207/206}$ versus As/Bi, which gives a purely visual idea of shared lead isotope and trace element trends for different stylistic groups of South Indian images.

inferences for these are discussed elsewhere.) Where possible, inscribed images or dated coinage, seals or charters were also analysed.

The trends observed based on dynastic chronology are summarized in the 'Appendix' (see 'Summary of typical analytical trends of lead isotope ratios and trace elements...'). The first part of the Appendix, 'Lead isotope groups', describes groupings discerned and marked on lead isotope ratio plots based on the spread in lead isotope ratios and such criteria. The second part of the Appendix, 'Trace element categories', describes trace element patterns or groupings discerned from the trace element scatter plots of Co versus Ni, Bi versus As and Sb versus Co. The third part of the Appendix, 'Summary of typical analytical trends of lead isotope ratios and trace elements...', summarizes the major analytical trends observed for different stylistic

groups defined by dynastic chronology, and also indicates those bronzes that formed the control group and the total number attributed to that stylistic group by comparisons based on both analytical signatures and style. Objects are assigned to particular groups only if both the lead isotope ratio groups and trace element groups were compatible. The broad stylistic groups discussed here were devised based on a fresh re-assessment of stylistic attributions made from the existing literature and were classified for convenience under the most prominent dynasty for the particular period.

The prevalence of isolated later bronzes with trace element or lead isotope trends matching earlier groups can be interpreted as being due to recycling of metal. If there are many such, this can be explained as the continued use of a source known in earlier periods in later periods. However, the prevalence of mixed elemental trends can suggest that a source known in earlier periods was mixed with a new source which then became widely prevalent in the later period.

Vijayalaya Chola bronzes (c. AD 850–1070)

This group refers to bronzes attributed to Chola rulers of the Tanjavur region of present-day Tamil Nadu, in South India, who were direct descendants of Vijayalaya Chola, the founder of the Chola lineage. Most of the classic Chola bronzes are attributed to rulers of this lineage such as the queen Sembiyan Mahadevi and the king Rajaraja Chola. This group shows marked coherence in the lead isotope ratios, sufficient to suggest that the lead came from one source, falling mostly into group 1a (Fig. 3). The same is noted for the elemental trends seen in the Co versus Ni and Bi versus As plots (Fig. 1). The Co/Ni values may be linearly correlated, roughly falling along a line through the origin which may relate to the characteristics of the copper source. The combined three-dimensional scatter plots of $Pb^{208/207}$ versus $Pb^{207/206}$ versus Ni/Co and of $Pb^{208/207}$ versus $Pb^{207/206}$ versus As/Bi (Fig. 4) show that the analytical signatures of this group can be discriminated well from the other stylistic groups, suggesting that the lead and copper sources for the artefacts were uniformly similar, although both copper and lead need not have come from the same source. This group is further subdivided into an Early Vijayalaya Chola group (c. AD 850–940) from the reign of Vijayalaya Chola, Aditya Chola to Parantaka Chola, with a few bronzes with analytical trends shared with earlier Pallava bronzes, and a High Vijayalaya Chola group (c. AD 940–1070) from the reign of the queen Sembiyan Mahadevi and Rajaraja Chola to the end of the Vijayalaya Cholas. The widowed matriarchal queen Sembiyan Mahadevi surely ranks as one of the most remarkable patrons, not only of Chola art but of Indian art as a whole, with a village in Tamil Nadu still named in honour of her (Barrett 1965; Dehejia 1990; Srinivasan 1996), while gender issues in the art and patronage of South Indian bronzes are discussed in Srinivasan (1996). The High Vijayalaya Chola group contained an inscribed Chola-style image of the goddess Kali from Senianvidudi (no. 42, Table 1; Fig. 5 (upper left)), palaeographically dated to the tenth or eleventh century, for which both lead isotope ratios and trace elements matched the analytical trends for this group. This image bears marked stylistic affinities—in the accoutrements and the hair (depicted erect like flames)—to an image of Dancing Kali in the Tiruvelangadu temple, attributed by Nagaswamy (1988) to Sembiyan Mahadevi's patronage (c. AD 950). A well-known archetypal Chola image which also fits the analytical signatures of this group is the Siva Nataraja (c. AD 985) (no. 37, Table 1); this was the subject of an admiring exposition by Rodin (1921) who compared the grace of its gestures to the Medici Venus's. The highest amount of tin in cast images was from a Bhudevi image (no. 32, Table 1) which fitted the trace element and lead isotope trends of the group with 15 wt% tin.



Figure 5 (upper left) Image of Kali, from Senianvidudi, Tanjavur district, Tamil Nadu, with inscription on pedestal palaeographically dated to the tenth–eleventh century AD, of a style consistent with that patronized by queen Sembiyan Mahadevi (c. AD 950) of the Vijayalaya Chola dynasty. Photograph: courtesy of Government Museum, Madras (Chennai) (acc. no. 280) (height: 51 cm). (upper right) Image of Kaliya Krishna, of a typical 'lesser' Vijayanagara style (c. AD 1500). Photograph: courtesy of Victoria and Albert Museum (acc. no. IS-204-1959) (height: 64.8 cm). (lower left) Image of Rama, representative of the Later Chalukya-Chola style (c. AD 1200). Photograph: courtesy of Government Museum, Madras (Chennai) (acc. no. 315/55) (height: 95 cm). (lower right) Image of a Yakshi with goose, of the first–second century AD, with analytical signatures and style consistent with an attribution to the Sangam era of the Tamil region. Photograph: courtesy of Victoria and Albert Museum (acc. no. IS-8-1989) (height: 13.3 cm).

Vijayanagara and Early Nayaka bronzes (c. AD 1336–1565)

Innumerable bronzes were produced for the Vijayanagara rulers, with their capital at Hampi in Karnataka and their feudatories the Nayakas in the Tamil region, who ruled a major portion of South India in the fourteenth to sixteenth centuries. Due to the continuance of conventionalized iconography from the Chola period, there have been problems with making attributions for this group. Some art historians would attribute bronzes of a debased style to this period. On the other hand, as far as stone sculpture is concerned, Michell (unpubl. lecture by G. Michell at the 'Kumbakom—Sacred and Royal City of Tamil Nadu' seminar, British Museum, June 1996) has argued for a neo-Chola revival emulating the earlier classic Chola style. Technical investigations indicate that the most marked separation in analytical patterns both for lead isotopes and for trace elements is noted between Vijayanagara and Early Nayaka bronzes (c. AD 1336–1565) and the Vijayalaya Chola bronzes (c. AD 850–1070). The Co and Ni values are much lower (Fig. 1 (upper)) while the Bi/As ratios are distinctive from other groups being generally greater than 0.19 (Fig. 1 (lower)), while the lead isotope ratios also form a discrete cluster (group 2) suggesting the lead is from a distinct source. Thus, using technical investigations, a problem of aesthetic judgment can be overcome whereby High Vijayanagara bronzes can be distinguished from Chola, and provincial Chola bronzes from later Vijayanagara. This study would support the existence of a 'High' Vijayanagara style of bronzes, along with a lesser Vijayanagara style of bronzes, rather than a style specifically imitative of the Cholas. A typical High Vijayanagara image fitting the analytical signatures of this group is an image of Rama (no. 23, Table 1) with 21 wt% zinc; this image is published in Michell (1995, 199). An image which is stylistically typical of the greater majority of 'lesser' Vijayanagara images and fits the analytical trends for this group is the Kaliya Krishna (no. 20, Table 1; Fig. 5 (upper right)), with a typical helmet-shaped crown.

Pallava bronzes (c. AD 600–875)

A major art-historical problem has been the existence of Pallava bronzes, associated with the Pallavas under whom a fine stone sculptural idiom flourished at centres such as Mahabalipuram in Tamil Nadu. Attributions of Pallava bronzes have been tentative and open to debate due to the lack of epigraphically-dated bronzes (Balasubrahmanyam 1971, 279; Barrett 1965, 1–5; Khandalavala 1995; Nagaswamy 1995). However, this study does indeed support the existence of a distinct Pallava school of bronzes from their discrete metallurgical profile. Whereas bronzes from other groups tend to form discrete clusters in the lead isotope ratios, ratios in Pallava bronzes seem to show a general linear trend (tending to fall along line 1 in Fig. 3), probably from the random mixing of two lead sources, one of which is group 3 which in fact also contains a Pallava lead coin. Although it has been problematic to distinguish Pallava from Chola bronzes, the scatter plot of Co versus Ni indicates that the coordinates of (Co, Ni) do not overlap with those for Chola bronzes, making it possible to discriminate between them using the parameters of lead isotope ratios and elemental trends. Also investigated were Pallava artefacts comprising two seals, a lead coin and a dated copper plate charter which were found to be consistent with the analytical signatures of the group. An image attributed to the Pallava period (Dr J. Guy, pers. comm.) which also tends to fit both the lead isotope and trace element trends of this group is a seated Vishnu (no. 1, Table 1). The subtle masterpiece of Somaskanda (no. 31, Table 1), attributed by Nagaswamy (1988) to Rajasimha Pallava (c. AD 700), also fits trends for this group. The Pallava group has been further sub-divided into Middle Pallava (c. AD 600–850) and

Later Pallava (c. AD 850–875) groups, of which the Middle Pallava group shows the most consistent analytical trends for this group, while the Later Pallava group shows some trends from the Vijayalaya Chola group. Indeed Vijayalaya Chola (c. AD 850–875), the first ruler of this lineage, was a contemporary of the Later Pallavas who were vanquished by his successor, Aditya Chola, around AD 875.

Chalukya-Chola bronzes (c. AD 1070–1279), including Early Chalukya-Chola (c. AD 1070–1125) and Later Chalukya-Chola bronzes (c. AD 1125–1279)

The groups of bronzes intermediate between Chola and Vijayanagara periods have received little attention from art historians. Technical study seems to help to discriminate between these problematic bronzes with confusing stylistic attributions. Although these bronzes are usually referred to as Later Chola bronzes, in this study these are classified as Chalukya-Chola bronzes based on a stylistic change postulated by Dehejia (1990, 93–5) from the ‘classic’ or High Vijayalaya Chola period to a slightly more provincial idiom in Later Chola bronzes due to the ascent of an eastern Chalukya ruler, Kulottunga I, on the Chola throne around AD 1070. The control group of bronzes was further sub-divided into an Early Chalukya-Chola group (c. AD 1070–1125) consisting of bronzes which show provincial features akin to royal portrait bronzes of this period with a remnant classic Chola style, and a Later Chalukya-Chola style (c. AD 1125–1279) till the end of the dynasty which shows a greater departure from the classic Chola style. This study suggests that the stylistic change is also detectable from the technical fingerprint since the Later Chalukya-Chola bronzes may be differentiated on the basis of trace elements, especially their Co and Ni trends. The elemental trends of the Early Chalukya-Chola period are a mix of the earlier Vijayalaya Chola and the Later Chalukya-Chola. The lead isotope ratios of Chalukya-Chola bronzes in general tend to fall in lead isotope group 7. An inscribed Jaina image of Adinatha from South Arcot, Tamil Nadu (no. 3, Table 1), dated to the thirteenth century, fits the analytical signatures of the Later Chalukya-Chola group. An image which can be classified as typically Later Chalukya-Chola, and which fits the elemental trends for this group with lead isotope ratios falling in group 7, is a Rama image (no. 14, Table 1; Fig. 5 (lower left)) from Tiruvelangadu, which was also dated by Dr S. Desikan (pers. comm.) to the twelfth century.

Later Nayaka and Maratha period (c. AD 1565–1800)

Usually bronzes from about the fourteenth century onwards are loosely classified as Vijayanagara or Nayaka and the attributions are often somewhat arbitrarily made, for example, to the eighteenth century. Indeed it is not easy to distinguish between these since later bronzes tend generally to follow the earlier Vijayanagara style. In this study an attempt was made to regroup these bronzes according to the major ruling dynasties of the period and, hence, the group of Later Nayaka and Maratha was devised. The stylistic indicators kept in mind were the temples of the Madurai Nayakas and the Maratha-style Tanjore paintings. In particular, the Bi versus As scatter plot indicates that the (Bi, As) coordinates for these bronzes are generally different from those for the Vijayanagara and Early Nayaka bronzes, which helps to distinguish them. The Sb and Co values seem the lowest of the groups studied. The low level of trace elements may also relate to a higher degree of refining of metal in later bronzes. A Siva Nataraja image (Victoria and Albert Museum, London, accession no. IM-1062-1873), with an inscription palaeographically datable to the nineteenth century, fitted the trace element trends of this group (lead isotope analysis was

not carried out); its pot-shaped *prabha* or circle of flames with the *kirtimukha* motif is typically seen in Late Nayaka temple sculpture.

Later Pandya/Later Pandya–early Vijayanagara transitional (c. AD 1279–1336)

This group was invoked to account for bronzes produced during the hiatus between the collapse of the Chola kingdom and the rise of the Vijayanagara kingdom in AD 1336, a period to which bronzes are rarely attributed. The dominant dynasties during this period were the Later Pandyas who ruled from Madurai from AD 1190 and rose to predominance by AD 1310 (Tapsell 1983, 443–4), although this was a period of instability due to the advances made by the Muslim Sultanate from Delhi who ruled Madurai between AD 1310 and 1340 (*ibid.*). The dominant style of this period would be a somewhat debased Chola style with a regional Madurai influence. The lead isotope trends fall on the periphery of those for the Vijayanagara period, group 2, while the trace element trends are also a mix of the earlier and later periods. This suggests that a new source may have begun to be used during this period (which was used more widely during the Vijayanagara period) but with some mixing of metal used in the earlier Chalukya-Chola period. Another distinguishing feature was that the average lead content was the highest, at 12 wt%, with a maximum of about 24 wt% lead for an image of Ganesa (Victoria and Albert Museum, London, accession no. IM-112-1924; only composition was analysed). An image with lead isotope ratios falling in group 2a and with elemental trends consistent with this group is that of the emaciated woman saint Karaikal Ammayar (no. 19, Table 1), depicted with provincial vigour; this has about 21% lead. Hence the above fits with the theory of a new lead source, that is, group 2.

Pre-Pallava/Andhra Pallava and Andhra bronzes (c. AD 200–600)

The precursors of the Pallavas were thought to be the Andhra Pallavas (who moved to the Tamil region from Andhra) and the Sangam dynasties, and, hence, this group refers to artefacts of an early Tamil and Andhra influence. Although both elemental and lead isotope analysis was not done for all artefacts, artefacts were attributed to this group only if they fitted either the trace element or lead isotope trends. Some of the trace element trends tended to be similar to Pallava images. However, a trend uniquely noted in all objects of this Pre-Pallava group was that the antimony contents fell below 0.1 wt%, which is shared by some but not all objects of the Pallava group. The lead isotope trends fell into groups slightly peripheral to the Chola group 1a, while retaining the South Indian bias. This group consisted of two reasonably well-dated Andhra high-tin (23 wt%) bronze coins (*c.* fourth century) of the Vishnukundin or early Andhra Pallava period (courtesy of R. Krishnamurthy). These matched some of the trace element trends of the Pallava group. This find is an example of the existence of ancient, indigenous and continuous traditions of the use of high-tin bronze in South India, first reported by the author in 1991 (Srinivasan 1997; Srinivasan and Glover 1995). Another is a zinc ingot from the Deccan, *c.* fourth century AD (no. 5, Table 1; courtesy of Dr N. J. Seelcy), which closely matched the lead isotope trends of an Andhra Buddhist ritual brass vessel (no. 6, Table 1) with 13 wt% zinc (both falling in group 5b), which in turn matched the elemental trends for this group. These findings are, thus, consistent with the tentative dates and analytical signatures for the artefacts and images of this group.

Figure 5 (lower right) shows a Yakshi (Victoria and Albert Museum, London, accession no. IS-8-1989), dated by the Victoria and Albert Museum to the first–second century AD, but for which attributions were uncertain as to whether it was Kushan or Deccan. Since its lead isotope

ratios fall in group 1b of the South Indian images rather than matching any of those seen in North Indian artefacts of the early historic period (see Fig. 4), and since the trace element trends fit those of the Pre-Pallava group, it may be concluded that it is more likely a Sangam-era image from the extreme south (c. AD 200). Indeed, the flexed pose and attitude of the slender image compares well with a maiden depicted on a Sangam-era gold signet ring from Karur and fits such an attribution, as agreed by Dr J. Guy (pers. comm.).

IMPLICATIONS FOR PROVENANCE AND SOURCES OF LEAD AND ZINC

Some aspects concerning source of metal are discussed here. Of the 60 images and artefacts analysed for lead isotope ratios, only two South Indian images could be confidently isotopically matched to known ore sources. One is a brass Buddha (no. 60, Table 1) which fitted lead isotope ratios of Ambaji ores in western India (Fig. 4), along with a remarkable, 1 m high, western Indian brass image (no. 58, Table 1) with an inscription of around AD 1250. This Buddha image had elemental and stylistic trends compatible with the Later Chalukya-Chola period. Both images had around 25% zinc. The other is a leaded bronze Ganesha from Nellore (no. 59, Table 1) which matches ore sources in western India from Dariba. This is the only bronze stylistically attributed to the Later Nayaka and Maratha period for which lead isotope analysis was done, and indeed this result is compatible with the fact that the Marathas were a western Indian dynasty who may well have introduced new sources of metal from north-western India.

While there is not enough lead isotope data on ore sources from the Indian subcontinent for identification of sources for all artefacts by isotopic matching, the lead isotope ratios for the rest of the South Indian bronzes do not match comparable data for ore sources from the Old World (Z. A. Stos-Gale, pers. comm.). The ores scrutinized include sources in the Mediterranean and the British Isles and Iranian, Turkish and Bulgarian ores from the extensive data bank of Isotrache (Stos-Gale 1992; Stos-Gale *et al.* 1995; Z. A. Stos-Gale, pers. comm.) and others analysed using similar standards and conditions, including Chinese samples (Brill *et al.* 1991; Sayre *et al.* 1992), and western Asiatic samples (Hauptmann *et al.* 1992).

The spread in the lead isotope ratios for groups 1 and 2 (Fig. 3) falls within $\pm 0.5\%$ of the mean, which, as mentioned before, is the spread observed with mines from the Indian region. In fact, nearly all the South Indian artefacts investigated, that is, groups 1–7, barring the two samples mentioned above and perhaps barring group 3 (see 'Appendix', 'Lead isotope groups'), can be seen to have a spread of not much more than $\pm 1\%$ of the mean, so that the possibility remains that the metal all came from one mining region in the Indian subcontinent with a large isotopic spread. The spread for ore sources from the Mediterranean, Europe and south-east Asia, on the other hand, is noted to be much narrower. Indeed, ratios for ore samples from Ambadongar in peninsular India (Venkatasubramanian *et al.* 1982) fall within a proximity of $\pm 1\%$ of the mean. (This analysis is only approximate, being measured to only two significant figures, whereas lead isotope measurements at Isotrache are measured to five.)

Figure 4 also shows that samples of artefacts from the Mauryan and early historic period (c. fourth century BC) from northern India, analysed at Isotrache (courtesy of Z. A. Stos-Gale; provided for analysis by Dr P. T. Craddock), do not really overlap with the South Indian bronzes (with one exception) and generally match north-western Indian ores such as Zawar and Ambaji, so that the lead isotope trends for the southern Indian artefacts can be regarded as distinctive. Apart from the medieval brasses, some 'Hellenistic' brass from the Aegean reported to be foreign to that region by Stos-Gale (1992) also uniquely seems to match Ambaji ores, which may

not be surprising given the early evidence for metallic zinc and brass from western India and Zawar (Craddock 1995). However, interestingly, there also seems to have been an early source of zinc for southern India as the lead isotope ratios of the Deccan zinc ingot, dated to the fourth to fifth century AD, closely matched the lead isotope ratios of the votive brass vessel also from the Andhra region and of a roughly similar date, falling in group 5b, the peripheral group to group 1.

In the lead isotope ratio plots, whereas a trend generally based on dynastic chronology was observed for other groups such as groups 1a and 2a, group 3, of three artefacts, differs in consisting of dated artefacts from different periods. These include a Pallava lead coin, while three Satavahana lead coins (c. first to second century AD), for which two ratios were analysed by Turner (1985), also fitted this group. However, the third ratio (i.e., $Pb^{206/204}$) was not analysed for the Satavahana coins which makes the comparison incomplete (Z. A. Stos-Gale, pers. comm.) since isotopic mis-match of this would invalidate the comparison. Nevertheless, the matching of two ratios most likely suggests that Satavahana lead may have been re-cycled in later periods, while there are not enough artefacts in this group to suggest that the source continued to be used on a large scale for later periods. This may also explain the linear trend from mixing for Pallava images if Satavahana lead coins were re-cycled by alloying them to leaded bronze. The $Pb^{208/206}$ and $Pb^{207/206}$ ratios for a single ore sample from Tavoy, Burma (Dayton and Dayton 1986), do match this minor group 3; however, the third ratio is not known which is especially relevant for matching ores to artefacts, while the analysis (made 1979–1982) is to two significant figures and, hence, is not comparable according to Stos-Gale (pers. comm.). Figure 4 indicates that of two other sets of Satavahana lead coins, analysed by Turner (1985) albeit for two ratios, one set matches ore from Bandalamottu in Andhra in South India analysed for this study, while the other set matches Sardinian ore from Fluminense published in Stos-Gale (1992). Indeed, Seeley and Turner (1984) suggest this possibility although the exact sources are not pin-pointed. Moreover, the lead isotope ratios of these Satavahana coins do not overlap with the early historic period Pre-Pallava and Andhra objects analysed for this study, which perhaps helps to distinguish them from roughly contemporaneous Satavahana artefacts.

A final point in exploring the distinctiveness of the analytical signatures for the South Indian bronzes is that of the trace element trends. A comparison was made with trace element trends of Ni, Co, Bi, As and Sb for a dozen Sri Lankan images analysed by atomic absorption spectroscopy (AAS) (by the British Museum Research Laboratory) and published in Von Schroeder (1990, 551). Of these, only the well-known life-size Buddhist Tara (no. 7, Table 1) matched South Indian elemental trends, that is, of the Pallava group. Lead isotope analysis for this image produced ratios which fell into group 5a, matching another gilt Buddhist image (no. 8, Table 1), from the Buddhist centre of Nagapattinam in southern India, for which the elemental trends fitted the Later Chalukya-Chola group. It may be inferred that the Tara may not be of Sri Lankan but of South Indian provenance from Nagapattinam, which is not inconsistent with the style of the image (Srinivasan 1996, 88). In general, the sources of Sri Lankan images seem to be different from South Indian ones based on the preliminary trace element comparisons above.

SUMMARY

Using the combined lead isotope and trace element framework discussed in this paper, it is possible to resolve the analysed South Indian images into stylistic groups of Pre-Pallava and Andhra bronzes (c. AD 200–600), Pallava (c. AD 600–875) consisting of Middle Pallava (c. AD 600–850) and Later Pallava (c. AD 850–875), Vijayalaya Chola (c. AD 850–1070) consisting

of an Early phase (c. AD 850–940) and a High phase (c. AD 940–1070), Early Chalukya-Chola (c. AD 1070–1125) and Later Chalukya-Chola bronzes (c. AD 1125–1279), Later Pandya (c. AD 1279–1336), Vijayanagara and Early Nayaka bronzes (c. AD 1336–1565), and Later Nayaka and Maratha period (c. AD 1565–1800). Some of the groupings are also ratified by comparisons with datable objects such as coins or inscribed artefacts.

The lead isotope and trace element trends of most South Indian bronzes seem to be distinctive from artefacts from other parts of the Indian subcontinent. Lead isotope analysis also indicates that the western Indian deposits such as Ambaji, which was a source of brass for northern India from the late pre-Christian era to the medieval period, were also minor sources of lead and brass for medieval southern India. A remarkable discovery is that, by the early historic period (c. fourth century AD), there appears to have been an early source of metallic zinc for southern India, which ranks amongst the earliest evidence for metallic zinc anywhere in the world. One source for southern Indian Andhra Satavahana lead from the Early Christian era seems to have been the local Bandalamottu mine in Andhra. Exchange of metal with the Mediterranean is indicated in the early historic period by the finding of a Sardinian source for some Satavahana lead and of a western Indian source (Ambaji) for some Hellenistic brass. With one or two exceptions, the South Indian bronzes do show a general bias in the lead isotope ratios; however, the sources are as yet unknown since the lead isotope ratios do not appear to match satisfactorily the known and comparable ratios for ore sources worldwide, while there are no data for many ore sources in southern Asia and India to complete the picture.

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APPENDIX

Lead isotope groups

The analysed South Indian images and copper alloys are categorized according to trends in lead isotope ratios, such as the proximity of lead isotope ratios indicated by the spread of ratios about the mean. These groups are also indicated on the lead isotope scatter plots (Fig. 3).

Group 1 This group of 21 bronzes is divided into 1a and 1b; 1a is the major core group with the closest lead isotope ratios and 1b consists of bronzes on the periphery of group 1a.

1a Seventeen bronzes

	Mean Pb ^{207/206} : 0.84505	Spread: ±0.26%
	Mean Pb ^{208/206} : 2.08923	Spread: ±0.2%
	Mean Pb ^{206/204} : 18.6003	Spread: ±0.35%
1b	Four bronzes	
	Mean Pb ^{207/206} : 0.84658	Spread: ±0.24%
	Mean Pb ^{208/206} : 2.09274	Spread: ±0.16%
	Mean Pb ^{206/204} : 18.5558	Spread: ±0.51%
<i>Group 2</i>	This group of 14 bronzes is divided into 2a and 2b; 2a is the major core group with the closest lead isotope ratios and 2b consists of bronzes on the periphery of group 2a.	
2a	Eleven bronzes	
	Mean Pb ^{207/206} : 0.85537	Spread: ±0.1%
	Mean Pb ^{208/206} : 2.11668	Spread: ±0.18%
	Mean Pb ^{206/204} : 18.3656	Spread: ±0.19%
2b	Three bronzes	
	Mean Pb ^{207/206} : 0.85618	Spread: ±0.21%
	Mean Pb ^{208/206} : 2.11466	Spread: ±0.13%
	Mean Pb ^{206/204} : 18.34822	Spread: ±0.12%
<i>Group 3</i>	This is a discrete group of three bronzes with proximal lead isotope ratios	
	Mean Pb ^{207/206} : 0.86368	Spread: ±0.21%
	Mean Pb ^{208/206} : 2.1093	Spread: ±0.18%
	Mean Pb ^{206/204} : 18.19903	Spread: ±0.39%
<i>Group 4</i>	This is a discrete group of two bronzes with proximal lead isotope ratios	
	Mean Pb ^{207/206} : 0.85616	Spread: ±0.11%
	Mean Pb ^{208/206} : 2.09961	Spread: ±0.12%
	Mean Pb ^{206/204} : 18.40025	Spread: ±0.02%
<i>Group 5</i>	This consists of two minor groups 5a and 5b which can be seen as belonging to a group 5 with spread from the mean of no more than ±0.22% for Pb ^{207/206} , ±0.15% for Pb ^{208/206} , ±0.22% for Pb ^{206/204} .	
5a	Three bronzes	
	Mean Pb ^{207/206} : 0.85209	Spread: ±0.11%
	Mean Pb ^{208/206} : 2.09534	Spread: ±0.04%
	Mean Pb ^{206/204} : 18.38052	Spread: ±0.07%
5b	Two bronzes	
	Mean Pb ^{207/206} : 0.84975	Spread: ±0.14%
	Mean Pb ^{208/206} : 2.08943	Spread: ±0.08%
	Mean Pb ^{206/204} : 18.4075	Spread: ±0.13%
<i>Group 6</i>	This consists of two minor groups 6a and 6b which can be seen as being part of a group 6 with spread of no more than ±0.18% for Pb ^{207/206} , ±0.13% for Pb ^{208/206} , ±0.32% for Pb ^{206/204} . Group 6 is a 'satellite' to group 1a suggesting it may contain mixed lead, with lead from the source of group 1a prevailing	
6a	Three bronzes	
	Mean Pb ^{207/206} : 0.84197	Spread: ±0.08%
	Mean Pb ^{208/206} : 2.08237	Spread: ±0.02%
	Mean Pb ^{206/204} : 18.6323	Spread: ±0.06%
6b	Two bronzes	
	Mean Pb ^{207/206} : 0.83997	Spread: ±0.08%
	Mean Pb ^{208/206} : 2.08664	Spread: ±0.01%
	Mean Pb ^{206/204} : 18.7136	Spread: ±0.05%
<i>Group 7</i>	Four bronzes	
	Mean Pb ^{207/206} : 0.85385	Spread: ±0.09%
	Mean Pb ^{208/206} : 2.10344	Spread: ±0.21%
	Mean Pb ^{206/204} : 18.3743	Spread: ±0.18%

Line 1 Some bronzes have lead isotope ratios which are found to fall on or close to a line of the following equation

$$y = mx + c; m(\text{slope}) = 1.3042; c(\text{constant}) = 0.98336$$

Scatter Bronzes which do not fit any group are designated as scatter 1, 2 and 3.

Trace element categories

Trends in the trace element composition for the analysed objects for Ni, Co, Bi, As and Sb are indicated as categories, also evident in the Co/Ni (Fig. 1 (upper)), Bi/As (Fig. 1 (lower)) and Sb/Co scatter plots. In the final, third, part of this appendix (below) typical analytical trends of lead isotope ratios and trace elements are summarized for each stylistic group (groups I–VII).

NB Some of the categories overlap as they are not always mutually exclusive.

∉ Does not belong to the set of co-ordinates defined in brackets.

∈ Belongs to the set of co-ordinates defined in brackets.

- A (Ni, Co) ∉ (0.11–0.4 wt%, 0.022–0.1 wt%)
- B (As, Bi) ∈ (0–0.8 wt%, 0–0.035 wt%)
- C (Co, Sb) ∈ (0–0.09 wt%, 0–0.23 wt%)
- D (Ni, Co) ∈ (0.11–0.4 wt%, 0.022–0.1 wt%)
- E (As, Bi) ∈ (0.3–0.5 wt%, 0.02–0.035 wt%)
- F (Co, Sb) ∈ (0.022–0.1 wt%, 0.1–0.45 wt%)
- G (Ni, Co) ∈ (0–0.1 wt%, 0.008–0.016 wt%)
- H Bi/As < 0.19
- I (Co, Sb) ∈ (0.008–0.016 wt%, 0.15–0.4 wt%)
- J (Ni, Co) ∈ (0–0.1 wt%, 0–0.008 wt%)
- K Bi/As ≥ 0.19
- L (Co, Sb) ∈ (0–0.008 wt%, 0.1–0.4 wt%)
- M (Ni, Co) ∈ (0–0.12 wt%, 0–0.008 wt%)
- N (As, Bi) ∈ (0–0.4 wt%, 0–0.035 wt%)
- O (Co, Sb) ∈ (0–0.008 wt%, 0–0.15 wt%)
- P (Co, Sb) ∈ (0.015–0.05 wt%, 0.15–0.45 wt%)
- R (Co, Sb) ∈ (0–0.9 wt%, 0–0.1 wt%)

Summary of typical analytical trends of lead isotope ratios and trace elements (Ni, Co, As, Bi, Sb) for various stylistic groups (I–VII), using lead isotope groups (1–7) and trace element categories (A–R) (see above), and with reference to Figures 1 and 3

(1) refers to those objects for which only lead isotope ratio analysis was done.

(2) refers to those objects for which only trace element analysis is available.

(3) refers to objects for which both lead isotope analysis and trace element analysis was done.

Group I Pre-Pallava and Andhra Pallava (c. AD 200–600) (total: 6 objects, control group: 2 objects)

(1) of three objects, two fell in group 5b and one in group 1b

(2) of five objects, three showed trends A, B, R and two showed trends D, B, R

(3) of two objects, both shared trends A, B, R and one each fell into groups 5b and 1b

Group II Pallava (c. AD 600–875)

Group IIa: Middle Pallava (c. AD 600–850) (total: 10, control: 7)

(1) of six objects, all tended to fall along Line 1, while some of these also fell into groups 4, 3 and 5a

(2) of six objects, all showed trends A, B, C

(3) one object analysed for lead isotopes and trace elements showed trends of B, C and Line 1

Group IIb: Later Pallava period (c. AD 850–875) (total: 7, control: 1)

(1) of four objects, three fell on Line 1 and also in groups 6a and 1b

(2) of five objects, the trace elements were a mix of categories A, B, C and D, E, F

(3) one object fitted Line 1/group 1b while the trace element trends were D, E, C/F

Group III Vijayalaya Chola bronzes (c. AD 850–1070)

Group IIIa: Early Vijayalaya Chola (c. AD 850–940) (total: 12, control: 4)

(1) of seven objects, all fell into group 1a, and one into group 6b

(2) of 12 objects, all showed trends D, E, F except two which showed trends A, B, C

(3) of seven objects, five showed both signatures of D, E, F and group 1a

Group IIIb: High Vijayalaya Chola (c. AD 940–1070) (total: 16, control: 12)

(1) of ten objects, all fell into group 1a

- (2) of 15 objects, all showed trends D, E, F
- (3) of nine objects, all showed both signatures of D, E, F and group 1a
- Group IV* Early and Later Chalukya-Chola bronzes (c. AD 1070–1279)
 - Group IVa* Early Chalukya-Chola (c. AD 1070–1125) (total: 13, control: 4)
 - (1) of four objects, three fell into group 7 and one into group 1b
 - (2) of 12 objects, all showed trend P and all but one showed trend D
 - (3) of three objects, all fell into group 7, and showed trends D and P
 - Group IVb* Later Chalukya-Chola (c. AD 1125–1279) (total: 17, control: 6)
 - (1) of six objects, one fell into group 7, two in group 3, two in group 2b and one in 5a
 - (2) of 17 objects, 14 showed trends G, H, I, while the rest showed at least one of trends G, H, I
 - (3) of six objects, the common factor was that they showed the trends G, H, I
- Group V* Later Pandya/Later Pandya–early Vijayanagara transitional (c. AD 1279–1336) (total: 11, control: 2)
 - (1) of six objects, four fell into group 2a and one into group 2b and one matched Ambaji ores in western India
 - (2) of 11 objects, the trends were a mix of G, H, I and J, K, L
 - (3) of four objects, the common feature was that they fell into group 2a
- Group VI* Vijayanagara and Early Nayaka period (c. AD 1336–1565) (total: 20, control: 8)
 - (1) of seven objects, all fell into group 2a
 - (2) of 20 objects, 18 showed trends J, K, L, while one showed trends J, K, O and the other trend K
 - (3) of seven objects, all fell into group 2a, and six showed trends J, K, L and the other trend K
- Group VII* Later Nayaka and Maratha period (c. AD 1565–1800) (total: 12, control: 7)
 - (1) one object matched ratios for Dariba ores in western India
 - (2) of 11 objects, eight showed trends M, N, O, three showed two of these trends, one showed trend N
 - (3) combined lead isotope and trace element analysis could not be attempted on any