

Lead Isotope Studies in the Aegean (The British Academy Project)

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Summary. In 1988 the British Academy awarded one of three special grants for collaborative research in the humanities for a project, initiated from Oxford, entitled “Science and archaeology: Bronze Age trade in the Mediterranean”. Within this project stress is laid on the collaboration of archaeologists with archaeological scientists in an attempt to study ancient ‘trade’ by welding together more traditional approaches (textual evidence, typology, distribution of artefacts, tomb paintings, underwater archaeology, etc.) with scientific studies of the provenance of ceramics and metals and of the organic contents of vessels. The work involves colleagues in Cambridge, Athens, London, Manchester and Oxford. A major aspect of Bronze Age trade was probably that in metals such as copper, silver, lead and gold. Though it has long been a goal for archaeological science to find a way to establish the ore sources that supplied the metal for particular archaeological objects, it has been only in the past ten years that success has come through the application of comparative lead isotope analyses of ores and artefacts. The contribution of these and other archaeometallurgical methods to the aims of the British Academy project are outlined in this paper.

1. The British Academy Project

In the context of new developments in archaeological science our topic has double relevance. It was our intention that both the organisation of the British Academy Group Research Project and the subject of provenancing metals using lead isotopes should engender new developments in archaeologi-

cal science. The British Academy Project has the title "Science and Archaeology: Bronze Age Trade in the Aegean and Adjacent Areas". It was conceived by the present authors, who formed a team involving archaeologists at Oxford, Cambridge, and the British School of Archaeology at Athens together with archaeological scientists at Oxford, Manchester, London and the Fitch Laboratory, Athens (see, for the personnel and a discussion of the aims of the project, Knapp 1989; Knapp and Cherry 1990). Exciting developments in new fields of archaeological research have often not been fully exploited because of the diversity of disciplines involved. We hope to make a major advance by integrating a variety of research strengths in a comprehensive study of an archaeological problem.

There have of course been a number of occasions before this when archaeologists and scientists have planned together collaborative work on a single archaeological site. In such ventures archaeological science (for instance, geophysical prospection, dating, environmental studies, etc.) often plays a rather subservient role, as a handmaiden to archaeology, producing data for interpretation by others or, worse, relegation to appendices of little apparent relevance in a site report. We hope to go beyond this, in demonstrating the validity of archaeological science as an academic discipline in its own right, just as within the Earth Sciences isotope geochemistry or geophysics have advanced the overall subject in ways not available to more traditional geological approaches. We selected a broad archaeological theme, depending on material from a host of sites, which we hope to advance by fusing together the methods of archaeology and archaeological science to achieve a result impossible for either alone. That theme is Trade in the Bronze Age Aegean. Both disciplines need to be brought together to establish the character and the details of such trade or exchange. The archaeological approach from excavation, field survey, typology, the decipherment of ancient texts and the study of Egyptian tomb paintings is no less necessary than the techniques of the natural sciences. Archaeological science can perhaps contribute most directly in the attempt to identify non-local materials, and to isolate or eliminate likely sources for these materials, be they ceramics, metals or organic residues. If we can make any progress with that, we can then try to build models of ancient trade and try to discover its importance to Bronze Age societies and its possible impact on their development. To give proper consideration to historical reconstruction and cultural issues, scientific analyses and statistical techniques must be incorporated in a research design that fosters basic quantitative research and builds interlocking arguments amongst data, analysis and interpretation.

One aspect of Bronze Age trade is that in metals, perhaps especially in the copper and tin which comprise the metallic alloy which names this period, but also in gold, silver and lead. There can be no doubt that metals were of

considerable importance in the Bronze Age of the Eastern Mediterranean, on the one hand as prestige objects which took part in the move to increasing social stratification such as the gold hilts of swords and the bronze daggers elaborately inlaid with gold, silver and niello, and the elaborate gold jewellery, all from the Mycenaean Shaft Graves (Iakovides 1974, 274–280; Hood 1978, 176–186, 190–207). No less important was the use of bronze for domestic utensils (Hood 1978, 170–172), in the provision of tools (Renfrew 1972, Plate 17) which transformed carpentry and ship-building and in weapons (Renfrew 1972, Plate 22; Iakovides 1982) and armour (Iakovides 1974, 282–283; Warren 1975, 128) which revolutionised war. Most of this paper discusses new developments in using comparative lead isotopic analyses of artefacts and metal ores to trace the ore sources which supplied the metals for this trade.

2. Lead isotopes

Let us now turn to the main focus of this paper: advances in the use of variations in the isotopic composition of lead for the provenancing of metals. It was just over 21 years ago that Brill (1970) reviewed progress in the beginnings of the archaeological use of lead isotopes in a lecture delivered in London in another symposium organised jointly by the Royal Society and the British Academy and published in the *Philosophical Transactions of the Royal Society*. We hope to show that we have progressed a little since then. My own (N.H.G.) introduction to the subject sprang out of collaborative work in cosmochemistry with the Max-Planck-Institut für Kernphysik in Heidelberg, when the then Director, Wolfgang Gentner, suggested joint work on the sources of silver for the Archaic and Classical Greek coinage (Gale *et al.* 1980). Subsequently the enthusiasm of Colin Renfrew steered us towards research in the Bronze Age Aegean.

3. Provenancing metals

The attempt to trace ancient metal objects back to the ore deposits/mines which supplied the metal from which they were made has a long history (e.g., Desch 1928, Marechal 1966, Junghans *et al.* 1968). Most of that history is of a succession of ill-fated efforts to establish trace and minor element patterns of the chemical composition of metal objects which might group sets of artefacts, and might be related to similar patterns of chemical compositions in ore deposits. Coles (1982, 287–291) has expressed the deep disappointment of archaeologists with the confusion introduced into Bronze Age studies by the misleading and false interpretation of trace element analyses of metals.

The failure of such an approach is well documented and was to be expected from the beginning (Catling 1964, 12; Gale and Stos-Gale 1982). First, ore deposits are not homogeneous in chemical composition, even on the small scale (e.g., Thompson 1958; Chernykh 1966; Griffiths *et al.* 1972; Bowman *et al.* 1975; Constantinou 1982). Second, the process of smelting metal from an ore involves the addition of fluxing minerals to separate, as slag, the unwanted gangue minerals in the ore from the desired metal. The fluxes introduce a chemical pattern different from that in the ore, the partitioning of elements between slag and metal varies from element to element and depends also on furnace temperatures and atmospheres (e.g., Tylecote *et al.* 1977), whilst some elements are relatively volatile and may in part be lost in furnace fumes. Subsequent processes like fire refining or cupellation can cause further changes in chemical composition. Third, chemically representative samples are not simple to obtain from relatively impure ancient metals which are themselves inhomogeneous in chemical composition (e.g., Slatér and Charles 1970), whilst corrosion can alter their original composition.

These disadvantages are not shared by comparisons of the isotopic composition of lead in an ore deposit with that of lead in an artefact made of metal from that ore. This is true whether we are talking of objects made of lead itself, or of silver, copper, bronze, iron or zinc containing traces of lead deriving from that in the original ore deposit. On the one hand the isotopic composition of lead in an ore deposit varies (with rare exceptions) by quite a small amount, less than about $\pm 0.3\%$. On another, the isotopic composition of lead in metal obtained from ore by extractive metallurgy is identical with that in the ore, and is constant throughout a metal artefact. The isotopic composition remains unaltered by subsequent purification of the metal, by the processes of making objects from the metal, or by subsequent corrosion of the objects (Barnes *et al.* 1978).

All this would be of no avail unless the isotopic composition of Pb were different in different ore deposits. Fortunately this is so. This is because three of the terrestrial isotopes of lead are partly radiogenic in origin, being derived from the radioactive decay of $^{238}\text{U}\{^{206}\text{Pb}\}$, $^{235}\text{U}\{^{207}\text{Pb}\}$ and $^{232}\text{Th}\{^{208}\text{Pb}\}$. Since mass spectrometers measure isotope ratios, we usually report analyses in terms of such ratios. These lead isotopic ratios record the time integrated record of the U/Pb and Th/Pb ratios of the sources in which the lead developed. In most cases ore deposits of lead/silver, of copper, of gold, of tin, of iron, of zinc, etc., contain vanishingly small amounts of uranium or thorium, so that after ore formation the isotopic composition of the lead which they contain will not change. (For more detailed discussions of the basic aspects of lead isotope geochemistry and systematics see Gale and Mussett 1973; Köppel and Grünenfelder 1979; Faure 1986; Gulson 1986).

The lead isotopic composition frozen into such an ore deposit which also

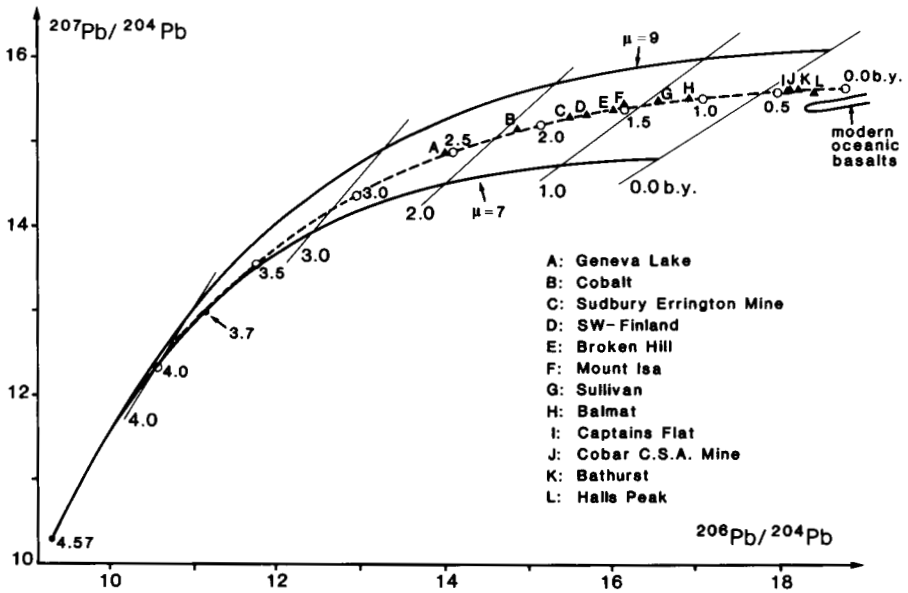


Figure 1. Lead isotope ratios for conformable ore deposits which appear approximately to fit a single-stage growth curve having μ ($=^{238}\text{U}/^{204}\text{Pb}$ at the present day) of about 8. The data come from a compilation by Kanasewich (1968). The numerals on the curve are dates B.P. in billions (10^9) of years.

had a relatively straightforward geological history will, to first order, reflect the geological age of that ore deposit. Such well behaved ore deposits are typified by the large, strata bound, conformable (stratiform, see Stanton and Russell 1959) ore deposits for which the Holmes-Houtermans single-stage evolutionary model was originally formulated (Holmes 1946). Figure 1 shows how the isotopic compositions of such ores lie on a curve according to the age of the deposit; nowadays the evolution of the isotopic composition of such ores is described by slightly more complicated models due to Stacey and Kramers (1975) and Cumming and Richards (1975). Fine structure deviations from such simple models will result for most ore bodies, since in practice the lead which they contain has had a complicated history. This may have involved residence in the Earth's mantle, a period of crustal residence accompanied possibly by metamorphism, erosion and sedimentation, until the lead and other metals were finally extracted during a volcanic cycle, or by the action of hot brines, and concentrated into an ore deposit. (See, for example, Barnes (1989) for an elementary account of modern ideas about ore deposition). Relatively rare deposits may reflect extreme isotopic anomalies not related to their apparent age. Figure 2 shows how such extremely anomalous, rare, ore deposits, as Ivigtut, Joplin or Idaho, plot completely away from the

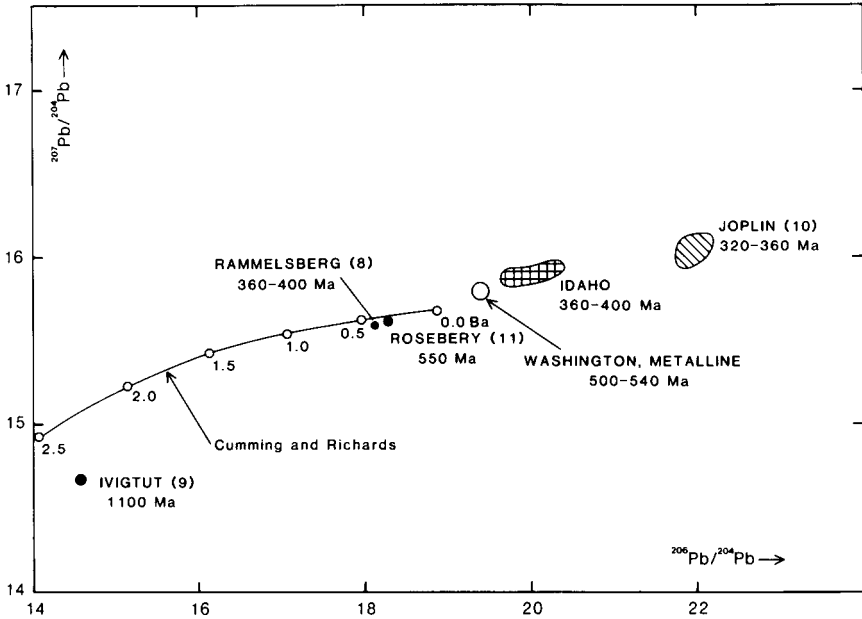


Figure 2. Lead isotope ratios for some ore deposits which are grossly anomalous (Ivigtut, Metalline, Idaho, Joplin) and for others (e.g., Rammelsberg) which lie (more typically) only a little away from the curve predicted by the model of terrestrial lead evolution devised by Cumming and Richards (1975) to describe the conformable ore deposits.

curve describing the conformable deposits; the more usual situation is exemplified by the Rammelsberg ore deposit, which plots just a little off the Cumming/Richards model curve. The usual situation, where the isotopic composition of lead in an artefact is related to first order to the age of emplacement of the ore deposit from which its metal came, is a useful advantage of lead isotope provenancing, since it can guide one in seeking possible ore deposits for which detailed lead isotope measurements should be made. There is of course no similar feature or advantage for chemical analyses.

One possible problem which would affect any technique for the provenancing of metals is that of possible melting down for re-use of diverse artefacts made from metal derived from a variety of ore sources. This has been considered in detail (Gale and Stos-Gale 1982, 1985; Gale *et al.* 1985; Stos-Gale *et al.* 1986; Pernicka 1989) and Pernicka *et al.* (1984) have devised a technique which combines lead isotope and trace element analyses in such a way as to prove whether mixtures exist or not. Use of this, together with archaeological arguments and the fact that we do not observe the smeared out lead isotope compositions for artefacts which would characterise mixing,

shows that it is not a problem for the Bronze Age Mediterranean. Possible problems with the addition of tin or the practice of liquation in the Bronze Age have also been discounted (Gale and Stos-Gale 1982, 1985; Stos-Gale *et al.* 1984; Pernicka *et al.* 1990).

4. Methodology and techniques of measurement

One of the most important developments of this provenancing method has been its use for a wide ranging, but well defined, archaeological research programme in the Mediterranean world. This has necessarily meant that the lead isotope compositions have had to be determined for the many copper and lead-silver deposits in this region. In assigning the provenance of, say, a copper object to a copper ore deposit on the basis of a match between their lead isotope compositions it is vital to discover whether the lead isotopic compositions of the relevant ore deposits can be distinguished, the one from the other. One important factor which is decisive for this is the accuracy with which lead isotope ratios can be determined. Since the whole range of lead isotope compositions yet found in ore deposits is only about 8%, and most fall in a region covering a range of only about 1.7%, it is clearly necessary to be able to determine lead isotope ratios with an accuracy of less than or equal to about $\pm 0.1\%$.

This was not possible until 1969. Before that the methods used for isotopic analysis of lead by thermal ionisation mass spectrometry (TIMS) involved the use of 1 microgram or more of lead in the nitrate or sulphide form, and reproducibility was no better than $\pm 1\%$. In 1968 Catanzaro *et al.* published a triple Pt filament TIMS technique which allowed lead isotope ratios to be measured to about $\pm 0.05\%$ at the 95% confidence level, but the technique required 1000 microgram samples, too high for most archaeological applications. This did however allow the United States National Bureau of Standards (NBS, now NIST) to prepare three international lead isotope reference standards, so that henceforward all laboratories could place their lead isotope analyses on an absolute basis. In 1969 Cameron *et al.* published a technique using a silica gel emitter which allowed of the order of 300 nanograms of lead to be isotopically analysed. [In fact Akishin *et al.* (1957) in Russia had published a similar technique a decade earlier, and Amov (1968) in Bulgaria also anticipated the American work, but these developments took a long time to be widely known in the west]. It was however not until 1973 that Barnes *et al.* at NBS in the USA, and independently Arden and Gale (1974) in Oxford, published low blank (blanks of about 1 nanogram) chemical separative techniques for lead and a silica gel emitter technique which allowed 100 nanogram samples of lead to be

measured to $\pm 0.1\%$ (at 95% confidence level). Since almost all the work in this field published by Brill involved analyses made at NBS, all of Brill's analyses published prior to 1974 are of an accuracy too low to be of any use. All Oxford work in lead isotope analysis applied to archaeology postdates 1976 and has the requisite modern precision and is moreover on an absolute basis by reference to the NBS lead isotope standards. An exciting new development has been the use of an internal standard ^{202}Pb — ^{205}Pb tracer against which to correct fractionation in the mass spectrometer, which for all lead isotope ratios reduces the absolute 2 sigma error to $\pm 0.01\%$ (Todd *et al.* 1984) but, the tracer being very expensive, this has not yet been used in archaeological applications.

5. Interpretation

Provenancing using lead isotope analyses is simple in principle, involving just comparative measurements of lead isotopic compositions of artefacts and ore deposits and the search for matches of composition, but there are many traps for the unwary. (For an extended review of the use of lead isotopes in provenance studies see Gale 1991).

The method of comparison and interpretation introduced and used by Brill is illustrated in Figure 3, adapted from one of his papers (Brill *et al.* 1974). The diagram is constructed from data obtained for some 400 specimens of various archaeological materials and ores from all over the ancient world. For analyses of new artefacts matches were sought with the regions of isotopic composition shown on the diagram, labelled L, X, E, S, etc. This approach, perhaps understandable in the early days when few isotopic analyses were available, in fact displays almost all the faults of interpretation which spring first to mind. First, the so-called lead isotope fields depend on both ores and artefacts from all over the ancient world and are labelled merely according to the geographical origin of most of the ores or artefacts within a given field; these are clearly not coherent groups and their use also involves some circularity of argument. Secondly there is mostly only an analysis of one ore from each ore deposit, so the extent of the field of isotopic composition for a given ore deposit remains unknown. Thirdly the ore analyses are almost solely of lead ores from deposits which do not contain copper, tin, etc, yet the artefacts include, for instance, bronzes which cannot possibly have been derived from such ore deposits. Fourthly the artefacts include glasses, glazes, pigments, bronzes, lead metal, debased silver from the Bronze Age to Roman times from all over the ancient world, amongst all of which one should hardly be seeking groupings with such gay abandon. Fifthly the fields are too catholic in their embrace; for instance that labelled

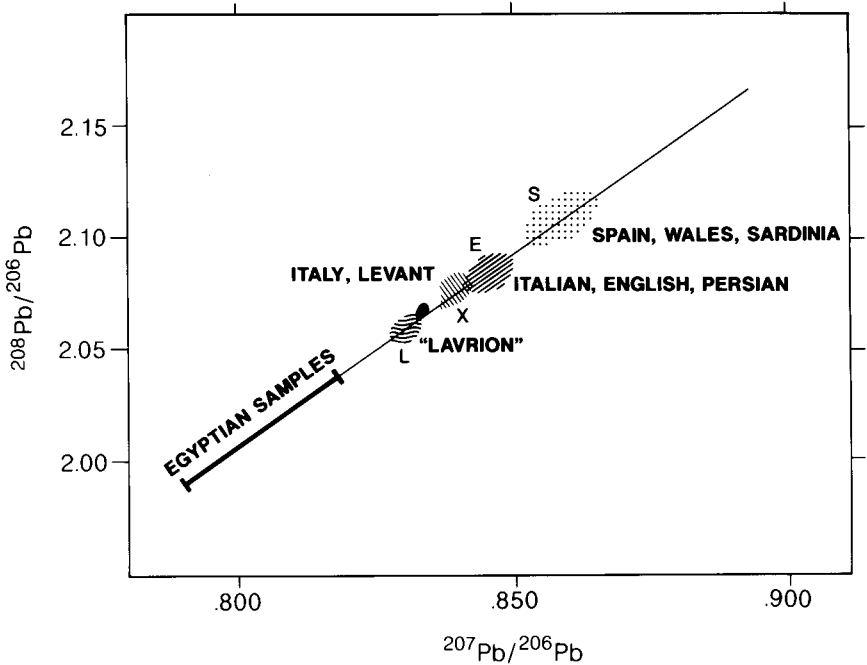


Figure 3. The lead isotope "fields" used by Brill and his colleagues (1974; 1988) to attempt to interpret lead isotope analyses in an archaeological context.

E includes ores and/or artefacts from Italy, England and Persia. Even the field labelled L for Lavrion includes ores and objects having nothing to do with that famous ore deposit in Attica upon which the wealth of ancient Athens depended. Gale (1980, 175, Figure 8) has shown by comparison how restricted is the true Lavrion field, constructed from our analyses of Lavrion ores only. And so on.

Though this approach was perhaps natural in the early days it seems unfortunate for it still to be in use (Brill 1988); progress cannot be achieved in this way. Progress can come only from the laborious construction of lead isotope fields for ore deposits in the region of interest and by the comparison of like with like: copper artefacts with copper deposits, lead/silver artefacts with lead/silver ore deposits, etc. Fieldwork is therefore essential, not only to collect ores from known geological horizons for isotopic analysis but also to establish the exact character of the mineral assemblages in particular ore deposits, and to differentiate between ore deposits and mere mineral occurrences which can never have been used at any period.

Fieldwork is also necessary to look for any traces of associated mining activities and to explore mines and slag heaps for ceramics or charcoal to date

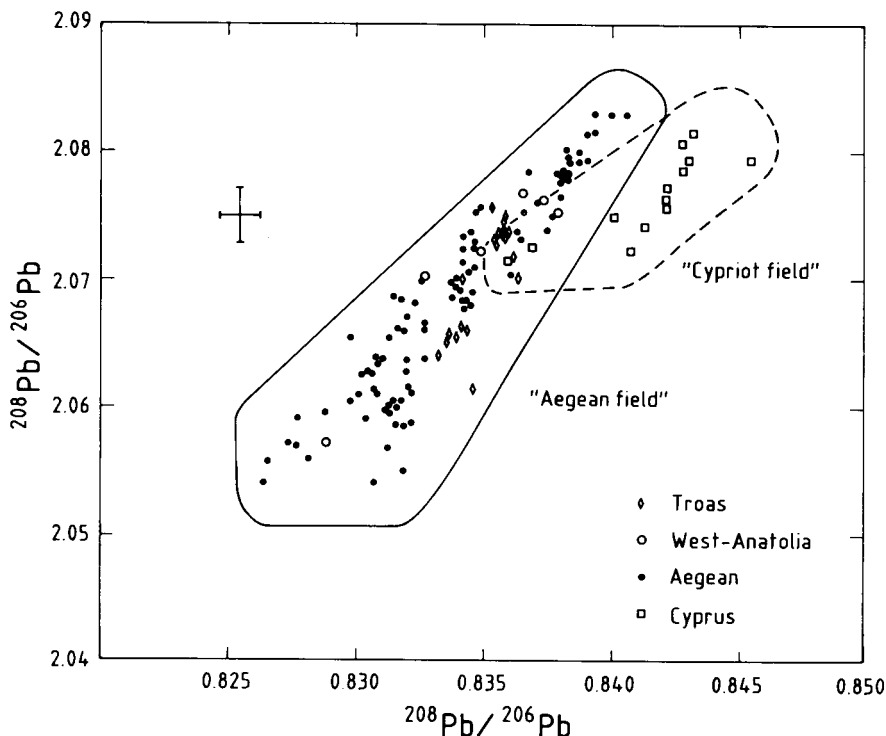


Figure 4. The so-called portmanteau "Aegean field" for lead isotope analyses of Aegean ores posited by Pernicka *et al.* (1984).

the activities. One will be immeasurably more confident about lead isotope evidence that a third millennium copper alloy dagger from Crete was made from copper from the Cycladic island of Kythnos against the background of the dating to that period of the copper slag heap on that island, by ceramic and accelerator C-14 dating (Gale and Stos-Gale 1989b; Stos-Gale 1989).

The vital importance of fieldwork in providing a proper basis for lead isotope studies is recognised by the Turkish/American team led by Aslihan Yener and by the German team also engaged in metal provenancing, and one cannot praise too highly the excellent fieldwork in Anatolia led by Gunther Wagner. Nevertheless we would not go along with the pessimism expressed by his colleagues (Pernicka *et al.* 1990, 282–283) that the lead isotope composition of lead in ore deposits is usually not unique, so that an assignment is not possible of an artefact to a single ore deposit. Their pessimism springs from constructing diagrams of what they call an Aegean lead isotope field (see Figure 4, adapted from Abbildung 28 of Pernicka *et al.* 1984), with which they try to compare analyses of artefacts made of lead, silver, and copper

alloys. At first sight it does seem that ore deposit is overlapping with ore deposit and that not much discrimination between them is possible.

This is a key point for provenancing using lead isotopic analyses; one must be able to discriminate between ore sources if one is to be able to trace a metal artefact to its ore source. We believe that diagrams like that reproduced in Figure 4 have some problems. First, it plots together on one diagram data from lead/silver deposits *and* copper deposits. Second, it depends in fact predominantly on isotopic analyses of Pyrite-Blende-Galena (PBG) ore deposits which contain no copper recoverable in ancient times, so that it is incorrect to compare copper alloy objects with most of it except for those parts representative of Cyprus or Lavrion. Third, we know that lead was not exploited in the Bronze Age Aegean except where it contained silver above a certain lower limit (Gale and Stos-Gale 1981a; Gale *et al.* 1984; Pernicka 1989); this diagram does not distinguish between the ore deposits which have silver above or below that limit. Fourth, the individual ore deposits are not distinguished so that possible structure in this diagram is ignored. Fifth, it displays only two out of the three Pb isotope ratios (a usual feature of papers from Heidelberg/Mainz), so that possible extra discrimination available by using all the data is missed. We have emphasised in many papers (e.g., Stos-Gale *et al.* 1984; Gale and Stos-Gale 1985; Gale and Stos-Gale 1989b) the necessity of using all three lead isotope ratios in order to obtain both maximum discrimination between ore sources and maximum confidence in assigning a metal artefact to an ore source, but it is only recently that others have begun to take up this point (Reedy and Reedy 1988; Sayre *et al.* 1990; Yener *et al.* 1991).

A first look at the isotopic evidence for some of the copper ore deposits in the Aegean (see Figure 5) brings out some of these points. First, the structure is evident, which is obscured if all the lead deposits are plotted also on the same diagram. Second, the extra discrimination afforded by use of all the data is illustrated, for example, by the overlap between the Kythnos and Cypriot fields in the upper diagram and the complete separation of these ore deposits in the lower diagram. Separation of two ore deposits in *one* lead isotopic diagram is clearly sufficient proof that their isotopic 'fingerprints' can be distinguished. Each of the two lead isotopic diagrams used in this paper is of course a particular section through the three-dimensional cloud of data points with coordinates $^{208}\text{Pb}/^{206}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, $^{206}\text{Pb}/^{204}\text{Pb}$.

6. Separation of lead isotope fields for ore deposits

Discrimination between two ore deposits having similar lead isotope compositions is vital to metal provenancing; it depends not only on the precision

COPPER ORES IN THE AEGEAN ANATOLIA AND SINAI

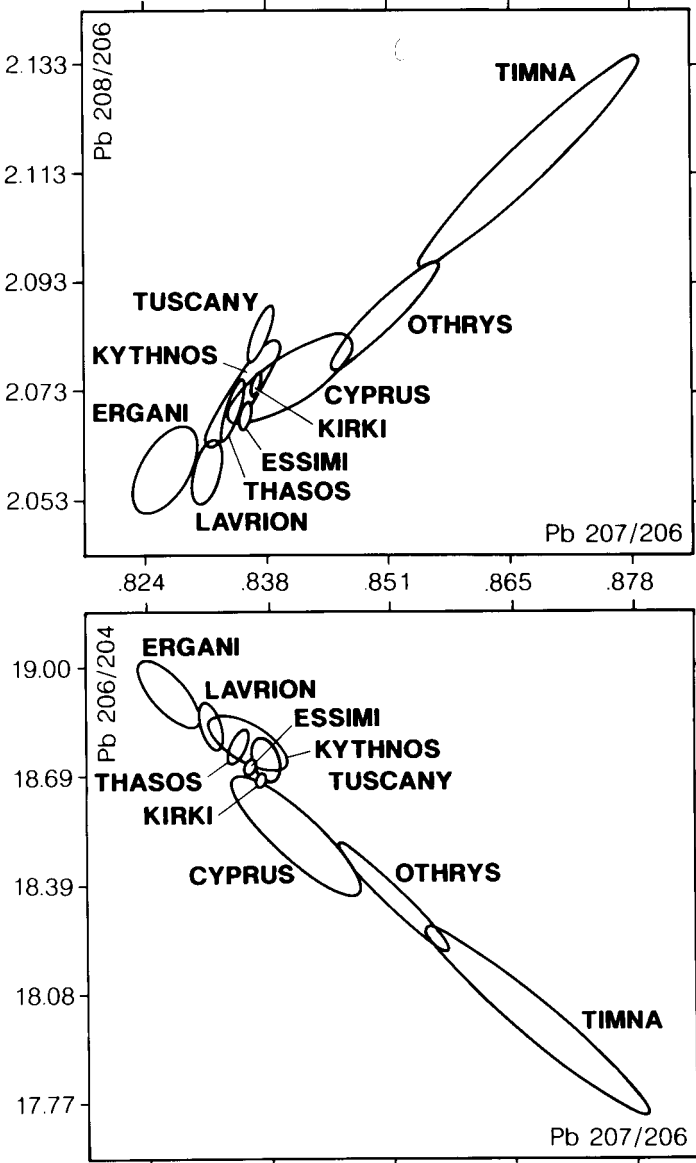


Figure 5. The two available ordinary bivariate lead isotope fields for some Mediterranean copper ore deposits.

and accuracy of the isotopic analyses, but also on the proper definition of the range of isotopic compositions found in each ore deposit. Some ore deposits seem to have, for samples collected from various positions within the deposit, lead isotopic compositions which are constant within the experimental uncertainty of $\pm 0.1\%$ [such as Balya in Turkey (Begemann *et al.* 1989) and Bleiberg in Austria (Köppel and Köstelka 1976)]. These are the exception rather than the rule, and it is always necessary to make a number of different analyses for different samples through the deposit in order to establish its isotopic field. Until we began work in this field in Oxford it had been very rare for more than one or two lead isotope measurements to be made for a single ore deposit. Without at least ten to twenty analyses for a deposit the range of compositions characteristic of that deposit (its lead isotope field) remains poorly known, and the degree of its possible overlap with nearby fields cannot be assessed with any confidence. This point was emphasised by Gale and Stos-Gale (1982), Stos-Gale *et al.* (1986), Gale (1986), Gale and Stos-Gale (1989a) and was re-emphasised by Reedy and Reedy (1988).

It is in this vital question of possible overlap of ore deposit lead isotope fields and of data analysis that important recent advances have been made. We illustrate this in terms of the available copper deposits in the Aegean, the locations of which are shown in Figure 6, and some in the Central and Eastern Mediterranean, such as those in Tuscany (Figure 7) for some of which there is disputed evidence that they may have been exploited at least by Etruscan times (see, for instance, Camporeale 1989). We shall also consider important copper deposits (see Figure 8) in Timna (Rothenberg 1988), in Cyprus (Muhly *et al.* 1982), in Ergani Maden (Griffitts *et al.* 1972), in the Taurus Mountains (Yener *et al.* 1989, 1991) and in the Troas and its hinterland (Pernicka *et al.* 1984; Sayre *et al.* 1990).

First a digression about how we construct, from lead isotope measurements of a number of different ore samples from a particular ore deposit, the lead isotope field of that deposit. Our initial approach (e.g., Stos-Gale *et al.* 1986) was to plot the data for the three isotope ratios involved on two diagrams and to draw an envelope around the error bars of the outlying data, as shown in Figure 9 (for one of the two possible diagrams) for ores and litharge from the Cycladic island of Siphnos, which has direct and indirect evidence for Bronze Age lead-silver mining (Gale and Stos-Gale 1981a, 195–203).

This was not very sophisticated, and we have now written a computer graphics programme (PICTURE) which, for the data points for each ore deposit, plots these diagrams together with ellipses drawn around the data for each ore deposit and calculated at a chosen statistical confidence level. This method depends on the assumption that the lead isotope ratio data for a given ore deposit are distributed according to a tri-variate normal distribu-

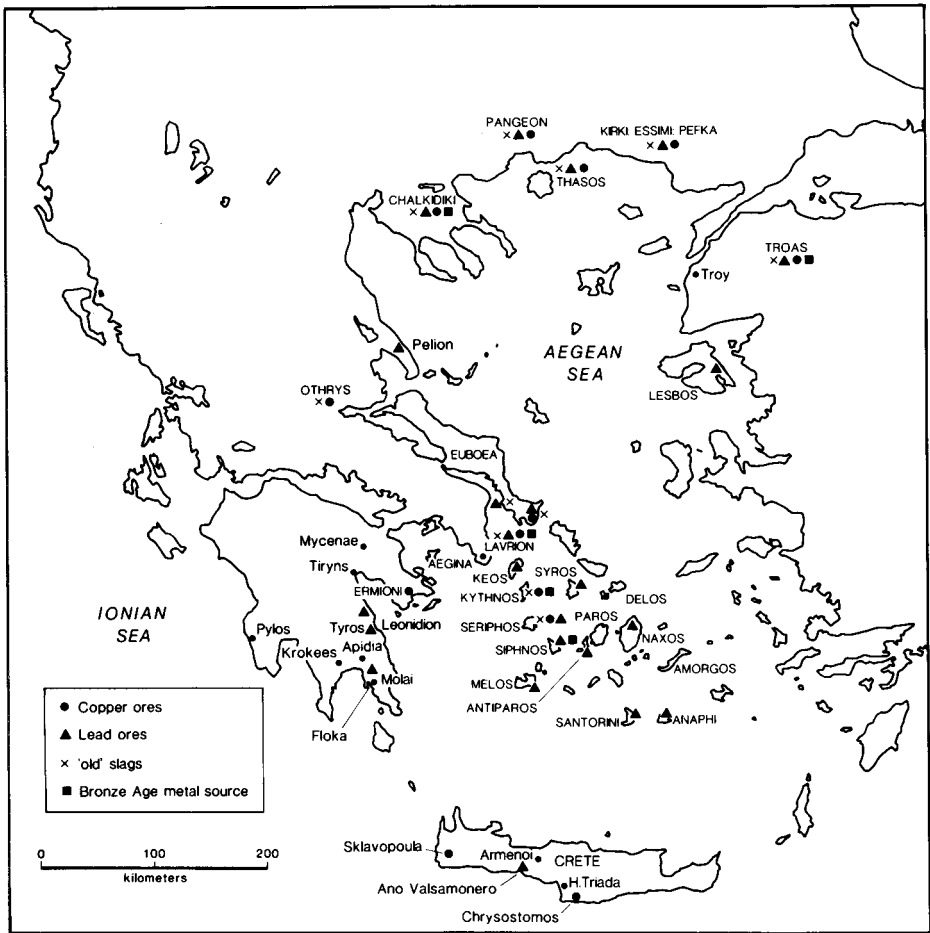


Figure 6. Locations of the principal copper and lead deposits around the Aegean.

tion function. We stress again that to define properly the field of each ore deposit at least 20 *geologically well-selected* ore samples are needed from each deposit, which again emphasises the need for fieldwork. For fewer samples something can be done with the use of F-tests and Hotelling's T^2 tests, but the true extent of the fields and proper assessment of overlapping will remain uncertain. Another approach to plotting bivariate lead isotope ratio diagrams, with ellipses drawn at preselected confidence levels (with or without the use of Hotelling's T^2 distribution for small samples), is to use the programme RAPLOT within the Brookhaven multivariate statistical software (Sayre 1975), now available for IBM PC compatible desktop computers.



Figure 7. Locations of the important copper and lead deposits in Tuscany.

We digress a little to consider briefly why we use lead isotope ratios rather than, as advocated by Reedy and Reedy (1988), the fractional atomic abundances (fractional compositions) of the four isotopes of lead [^{204}Pb , ^{206}Pb , ^{207}Pb , ^{208}Pb], which they claim to be less correlated than the ratios (one must in fact choose only three of these fractional abundances; they are not completely independent, their sum being unity). In fact their analysis shows rather marginal differences between the correlations for atomic abundances *versus* ratios, but there is a more fundamental reason for choosing ratios. All mass spectrometers used for accurate isotopic work in geochemistry or archaeology produce their primary data as ratios, for technical reasons partly connected with the techniques used for correcting for fractionation during an analysis (e.g., Dodson 1963); *all lead isotope data in the literature has been produced as ratios*. So far the vast majority of lead isotope data produced for

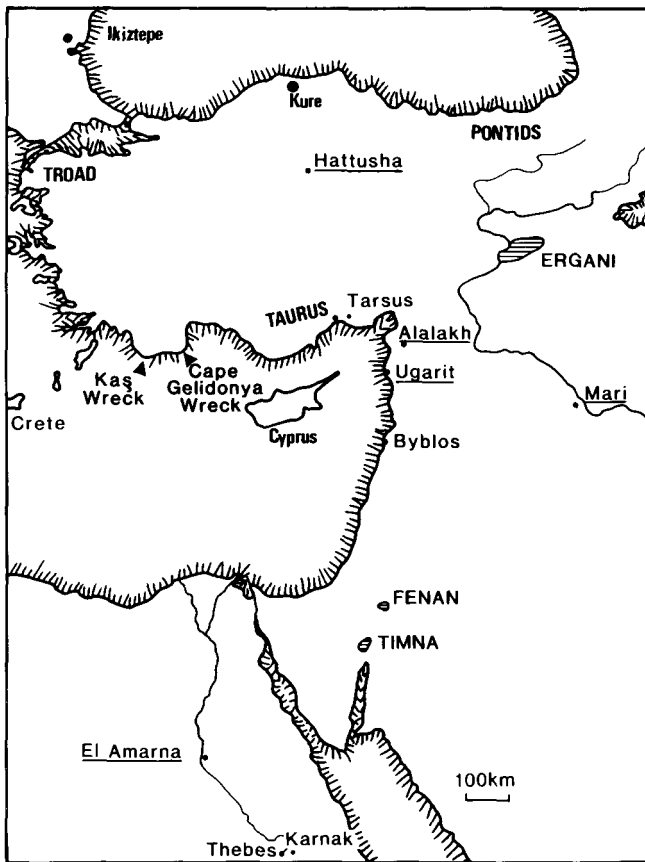


Figure 8. Approximate locations of important copper deposits in Timna, Fenan, Cyprus, Ergani Maden, the Taurus Mountains and the Troas. The locations are also shown of the Bronze Age shipwrecks of Cape Gelidonya and Ulu Burun (Kas) excavated by Bass (1967, 1986).

archaeology has been obtained using single collector mass spectrometers where the ion beams for the different isotopes were switched rapidly and sequentially into the collector; unavoidable instability in the thermal ionisation process forced the data to be obtained as ratios of ion beam currents, with interpolation procedures to take account as far as possible of the time varying ion currents (Webster 1960; Dodson 1963; Wasserburg *et al.* 1969; Wasserburg 1987, 134–138). Modern mass spectrometers often have software which allows the data to be printed out as fractional atomic abundances, but this data is secondary, having been computed from the directly measured ratio data. The atomic abundance data is therefore, due to propagation of errors, always less accurate than the primary ratio data; the primary ratio data is therefore the first choice for multivariate statistical analysis, other

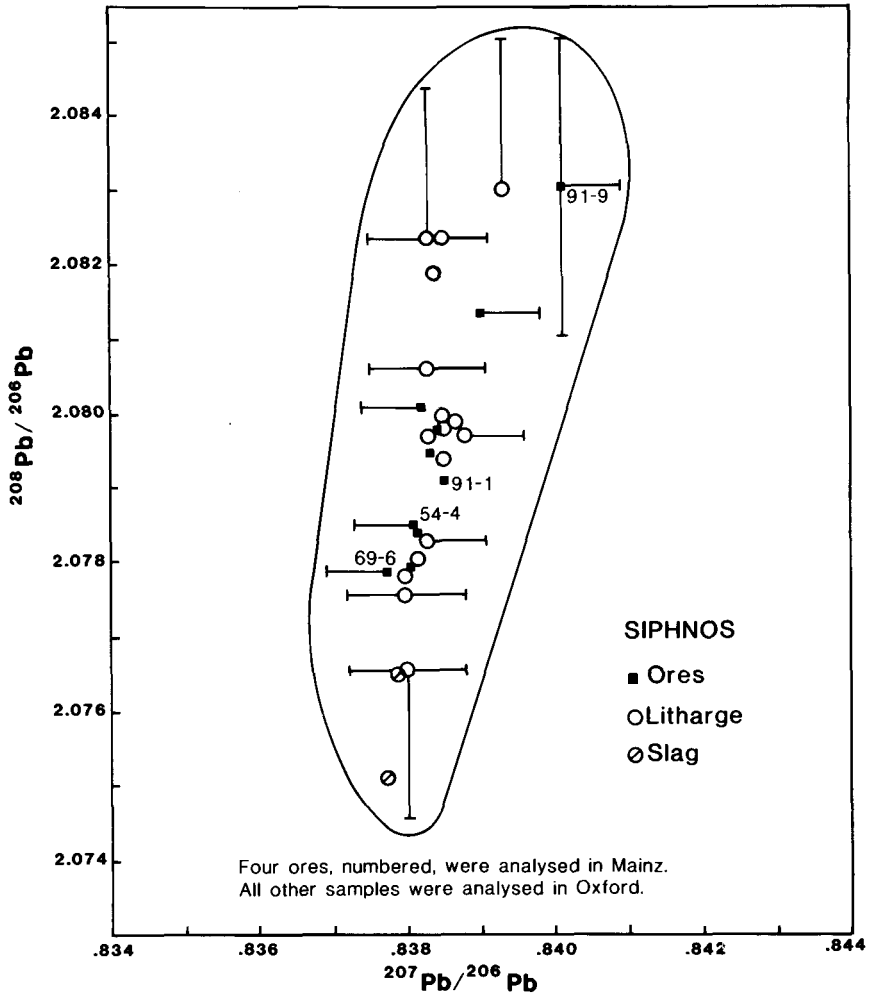


Figure 9. One of the two ordinary bivariate lead isotope fields for the lead-silver ores from the Cycladic island of Siphnos.

things being equal. In practice it seems to make little difference whether one uses the ratios we select, another choice of ratios, or the atomic abundances. We have empirically investigated the effect of such alternative choices of input data for the BMDP programme 7M (discriminant analysis) and the Brookhaven programmes RAPLOT, CONDIST, ADCORR and ADSEARCH; we find no differences, within computational error, between

the computed probabilities etc., which result from the different choices of input data type. Sayre (pers. comm.) reports the same conclusion.

Figure 5 shows 90% confidence level fields (a confidence level used also by Sayre and his colleagues in two-dimensional plots of lead isotope data, e.g., Yener *et al.* 1991), produced using the programme PICTURE, for some relevant copper ore deposits, including most of those in the Aegean and the Central and Eastern Mediterranean. It covers nearly the same range of isotopic compositions as the large Aegean "field" (see Figure 4) drawn by the Mainz/Heidelberg team, but it includes only copper deposits and reveals the structure within the larger "field" and the possibility of assigning individual artefacts to individual ore sources not shown by their approach. It also shows that the overlaps in the upper diagram (of Figure 5) between Kythnos and Cyprus, and Essimi with Cyprus, vanish in the lower diagram. The large extent of the isotopic field for Timna is due to the presence of uranium in some of the copper ores there, so that evolution away from the isotopic composition at deposition has continued until the present day.

The necessity of using all three isotope ratios is clear in attempts to unscramble the apparent overlapping of ore deposit lead isotope fields in the lower third of Figure 5. We also need to have a method of assigning statistical probabilities of the lead isotope data for one ore deposit falling into the isotopic field of another deposit, and for estimating the probability that the isotopic composition of an artefact assigns it to one ore deposit rather than another. We clearly need to combine all three isotopic ratios together in some form of multivariate statistical analysis. Since we know in advance that each geologically well defined ore deposit constitutes a group for statistical purposes, we can use the method of stepwise discriminant function analysis, for which the BMDP programme 7M is appropriate. (We used this approach first in Gale 1986). Figure 10 shows the result of using this approach to investigate the possible overlap between the lead isotope data for copper deposits on Kythnos and Cyprus, and the possible overlap of each with the fields for Essimi and Kirki in the Rhodope Mountains (Greece, see Figure 6); both the scatterplot illustrated and the numerical data generated by BMDP 7M give good separations of these copper ore deposits. [In subsequent discussion we shall present just the scatterplots of the first two Canonical variables, but in each case where we refer to good separation being obtained between two ore deposit lead isotope fields it should be understood that the numerical probabilities of all the samples from one field belonging to a nearby field are less than 5%. The enclosing lines drawn around samples from the same ore deposit are merely to guide the eye; they have no statistical significance.]

Figures 11 and 12 show how other apparent possible overlaps between copper ores in Tuscany, Kythnos and Cyprus, and between the very large

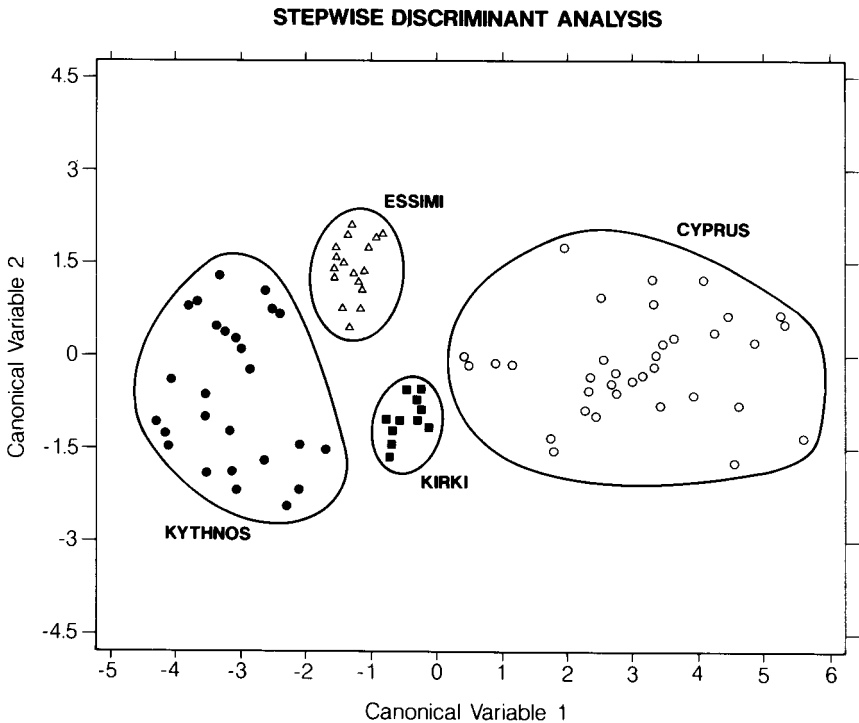


Figure 10. Stepwise trivariate discriminant function analysis used to show that the lead isotope data for the copper ore deposits of Cyprus, Kythnos, Essimi and Kirki can be resolved from each other. For brevity only the scatterplot of the canonical discriminant functions are shown. The outlines have no statistical significance, and are drawn merely to guide the eye.

Anatolian copper mine of Ergani Maden in Turkey and Lavrion in Attica are easily resolved by discriminant analysis involving all three lead isotope ratios.

We cannot afford to leave out of account the copper deposits in Turkey in any investigation of copper sources which may have played a part in the Bronze Age metals trade, especially for sites in the North Eastern Aegean. Here however we meet a dilemma which has been pointed up also by the Smithsonian Laboratory (Sayre *et al.* 1990). Although rather thorough field-work has been carried out in Turkey by the team led by Gunther Wagner, in which over a five year period they have visited and briefly described in excess of 150 ore and slag deposits (Wagner *et al.* 1989 and references therein), there are currently only about 80 lead isotope analyses published for these deposits, many of which are lead rather than copper deposits. Four of the deposits account for 29 of the analyses, so that most deposits have one, or at the most two, analyses, and many remain completely unanalysed. In common with the Smithsonian Laboratory we find that there are too few analyses of the

STEPWISE DISCRIMINANT ANALYSIS

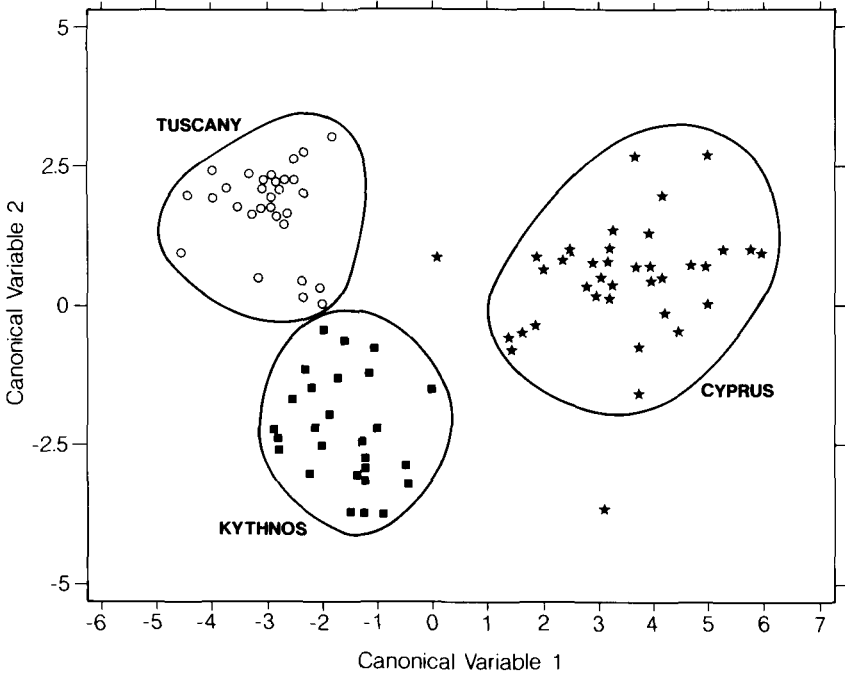


Figure 11. Discriminant function (trivariate) analysis used to resolve lead isotope data for the copper deposits of Tuscany, Kythnos and Cyprus.

Anatolian Black Sea sources, and from many other parts of Anatolia, for meaningful statistical characterisation. We have already dealt with the Ergani Maden mine; in addition one has from the German work enough data to characterise the Keban copper ore source at the headwaters of the Tigris and Euphrates rivers, the Kure mine on the Black Sea coast NNW of Ankara, and two groups of mines in the Troad region of Northwest Anatolia designated Troad 1 and Troad 2 in Figure 13, which shows the data in one of the two available ordinary lead isotope diagrams. We should mention here that Troad 1 embraces the deposits and slags designated by the German team as TG13A, TG14, TG15, TG16A, TG17A, TG142H, TG144C, TG144D, TG145A, TG146A, TG147A, TG150, TG152; Troad2 is made up of TG19, TG128B, TG130A, TG133B, TG133E, TG134A, TG137A, TG140A, TG143B. It should be emphasised that Troad1 and Troad2 are formed only by associating together two groups of ore and slag samples which have similar lead isotope ratios; there seems to be too little geological information to provide a sure geological basis to form these two "fields". Each group consists of over 8 different deposits, mostly with one analysis each, linearly

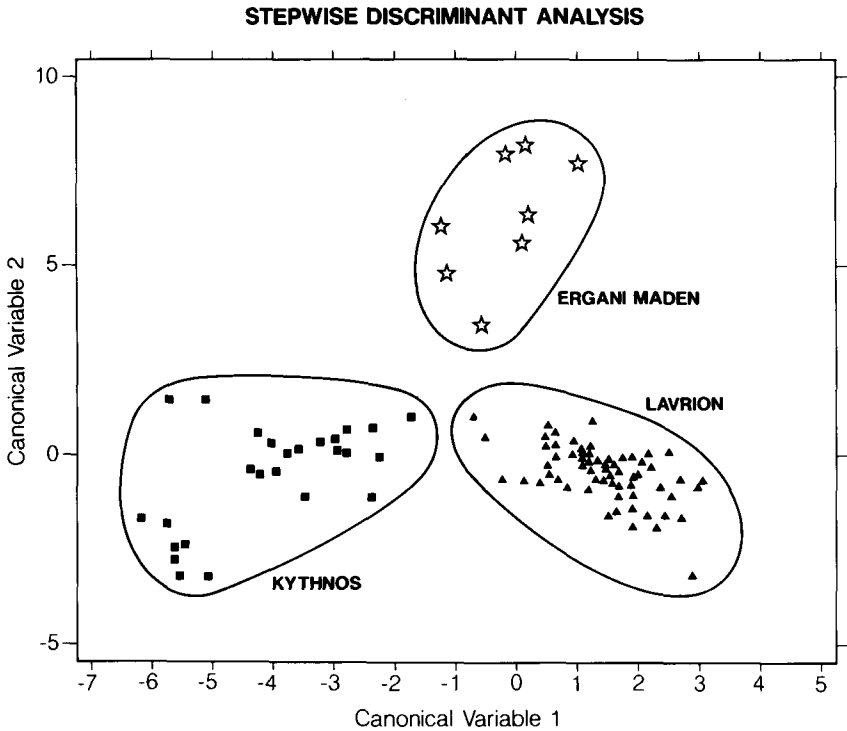


Figure 12. Discriminant function (trivariate) analysis used to resolve lead isotope data for the copper deposits of Ergani Maden in Anatolia, Lavrion in Attica and Kythnos.

distributed over a distance of about 100km. We shall accept these two Troad "fields" provisionally for the present, but only as a matter of convenience for the immediately succeeding discussion.

Figure 13 shows that we do not need to use statistics to see that there is no problem of overlap as far as the Keban and Kure mines are concerned, but at first sight there is an apparent problem with overlap between the Kythnos field and the Troad1 and Troad2 fields. Discriminant analysis (Figure 14) shows good separation of the Troad2 field, and rather good separation of the Troad1 field from the Kythnian field, with just a few Kythnos data points straying into the Troad1 field.

The Smithsonian Laboratory has analysed a number of possible copper ores from the Taurus Mountains (Yener *et al.* 1991). Figure 15 shows a partial overlap of the Taurus 2A field with Cyprus in one conventional bivariate diagram which is resolved in the other, whilst Figure 16 shows that complete separation is obtained by resorting to trivariate stepwise discriminant analysis.

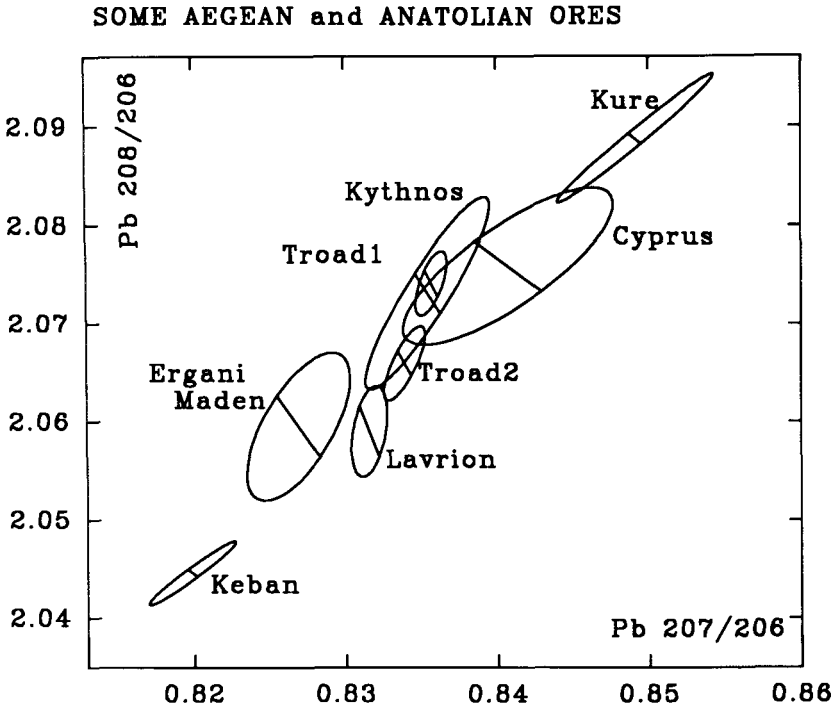


Figure 13. One of the two available ordinary bivariate lead isotope diagrams for copper ores, including the Anatolian deposits of Keban and Kure and two groupings of deposits in the Troad.

There is no space for overmuch repetition of such case studies here; suffice it to say that we have encountered only one serious overlap of lead isotope composition for copper ores in the Aegean, Central and Eastern Mediterranean. That is for the island of Thasos (locality given in Figure 17) which overlaps with Kythnos. Discriminant analysis does not completely resolve this overlap. This must be set in context, by noting that copper ores are insignificant in amount on Thasos and that the thermoluminescence date obtained for a copper slag heap there at Makrirarchi is 260 ± 160 BC (Pernicka and Wagner 1988, 229). Even more persuasive evidence that we do not have to worry about Thasos as a significant source of copper in the Bronze Age are our lead isotope analyses of a series of Late Bronze Age bronze objects, mainly knives, from Dr. Koukouli-Chrysanthaki's (1982 and references therein) excavations on Thasos, shown in Figure 18. Statistical analysis is not needed to see that not a single artefact is consistent with having been made from Thasian copper ores, whose lead isotopic fields are shown by the ellipses. Indeed the wide range of isotopic composition of the Thasian bronzes shows that the Thasians were, in the Late Bronze Age, using a wide

DISCRIMINANT ANALYSIS OF ORES FROM KYTHNOS, TROAD1, TROAD2.

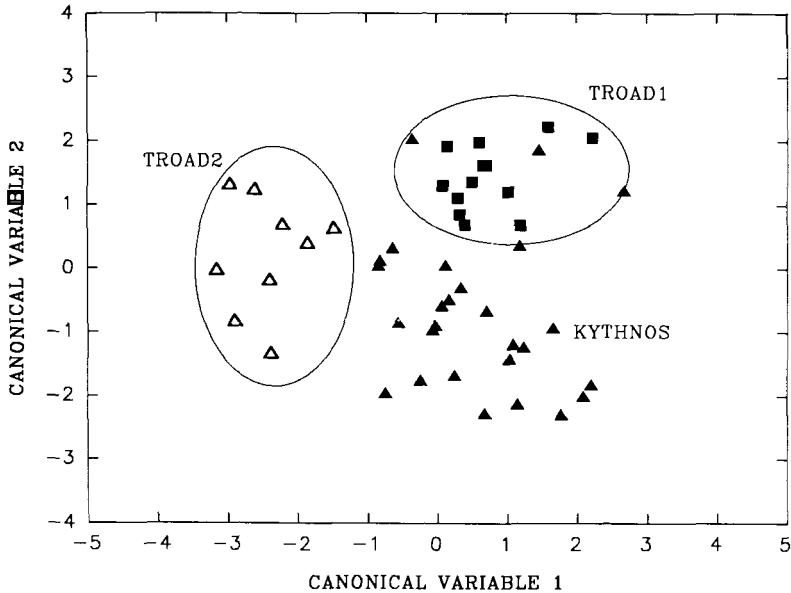


Figure 14. Discriminant function (trivariate) analysis showing good separation of the Troad1 and Troad2 "fields" from the Kythnian copper ores and slags.

variety of copper sources, which they would not be likely to have done if copper were easily available on their own island and they had the knowledge to exploit it. This sort of evidence, that in the period under review the local people did not exploit their own meagre resources and that therefore the resource in question cannot have been used abroad in that period, seems to us admissible extra evidence in the rare cases where partial overlap of ore source lead isotope compositions prevents more direct conclusions.

7. Oxhide ingots

We stress again that in the Bronze Age Mediterranean which forms the focus of our British Academy Project, overlapping ore fields are very rare. We now examine briefly the conclusions which lead isotope analyses allow us to draw about an important part of the copper trade in the Bronze Age Mediterranean. This took the form of the transport of copper ingots weighing on average about 29 kilograms and of a characteristic shape. They are called oxhide ingots by reason of a fancied resemblance of their shape to the flayed skin of an ox. The most comprehensive archaeological accounts of these

AEGEAN, TUSCAN AND ANATOLIAN ORES

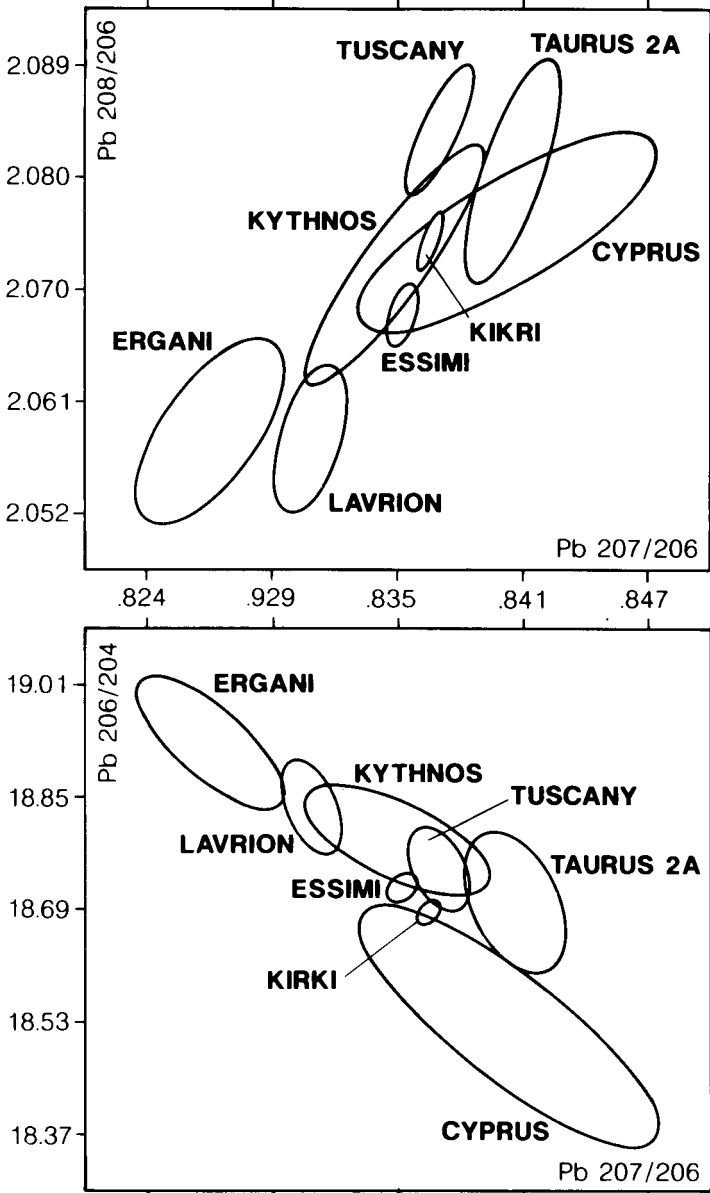


Figure 15. The two available ordinary bivariate lead isotope diagrams showing, amongst other things, a partial overlap between the Taurus2A field and the Cypriot field.

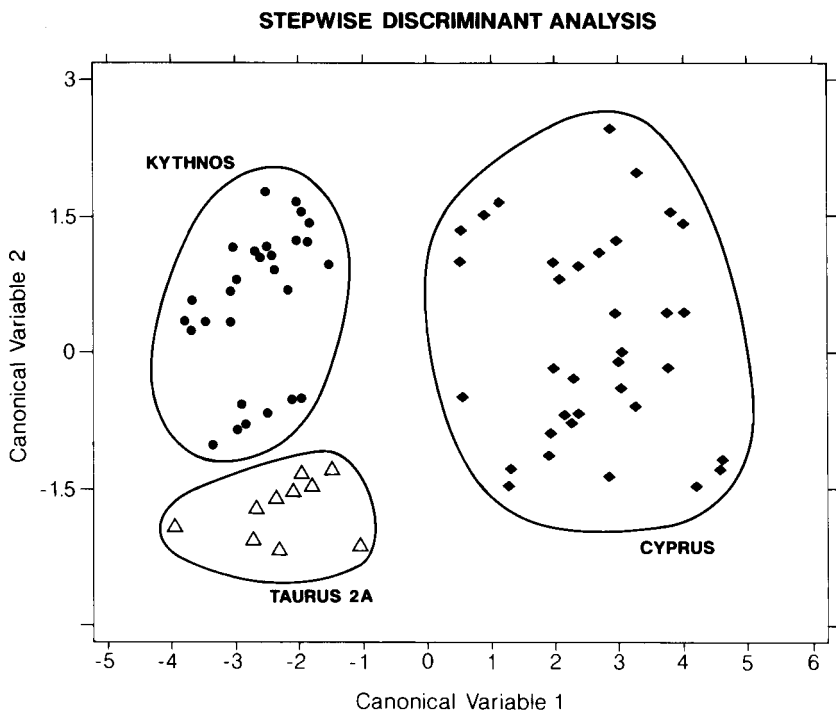


Figure 16. Trivariate stepwise discriminant function analysis produces complete separation of the lead isotope data for Cypriot and Taurus2A copper ores.

ingots are given by Buchholz (1959) and Bass (1967), both of whom provide many illustrations of typical examples.

Figure 19 is a map of the distribution of find spots of oxhide ingots around the Mediterranean. For the finds on land, which often consist only of fragments, it is difficult to estimate the number of whole ingots represented, but it is about 130, with about 20 on Cyprus, 37 on Crete, roughly 50 on Sardinia, 22 in Greece (chiefly Mycenae and Kyme) and one each in Sicily and Lipari. Two Bronze Age shipwrecks have been excavated off the coast of Turkey by Bass at Cape Gelidonya (Bass 1967) and Ulu Burun (Kaş; Bass 1986); the localities are shown in Figure 8. There were 38 ingots on the Cape Gelidonya wreck and more than 200 on the Ulu Burun wreck, so that about 65 per cent of all known ingots have been found on only two shipwrecks.

That indicates the scale of the trade in copper in the oxhide ingot form alone (or at least that *intended*; how many ships foundered with their cargoes, how many voyages were successful?), with at least 6 metric tonnes of copper as oxhide ingots on the Ulu Burun ship alone. The distribution of find spots on land suggests that these ingots were almost exclusively transported by

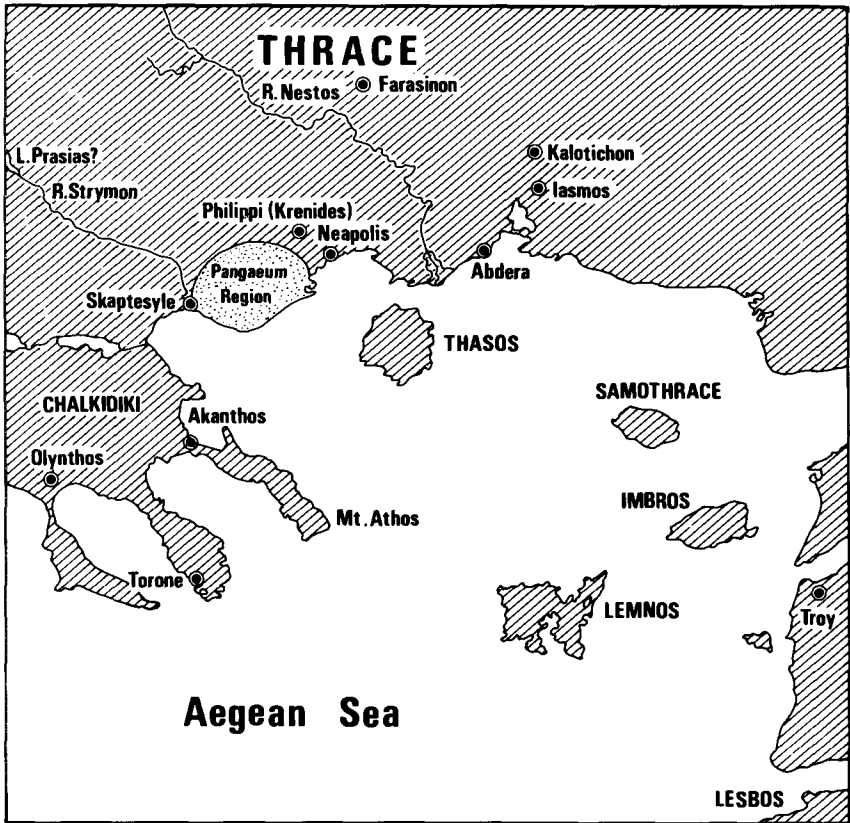


Figure 17. The locality of the island of Thasos in relation to its hinterland on the mainland.

ship. They have been found overwhelmingly either on islands or at mainland sites not far from the sea; sites which in the main have other evidence for international trade.

Before our work there was no evidence about the origin of these ingots, though the common view was that they represented trade in copper from Cyprus, based chiefly on the idea that Cyprus was a copper rich island with evidence for copper metallurgy and on which a few oxhide ingots had been found. Our first lead isotope analyses proved that the 13th century BC oxhide ingots found on Cyprus were indeed consistent with having been made from Cypriot copper ores (Gale and Stos-Gale 1986). Figure 20 gives the evidence for that as it appears in a discriminant analysis scatter plot, showing that lead isotopic analyses of Cypriot copper oxhide ingots fall inside the region defined by Cypriot copper ores. [We shall use such discriminant analysis scatter plots throughout the rest of this section; it is implicit that the BMDP

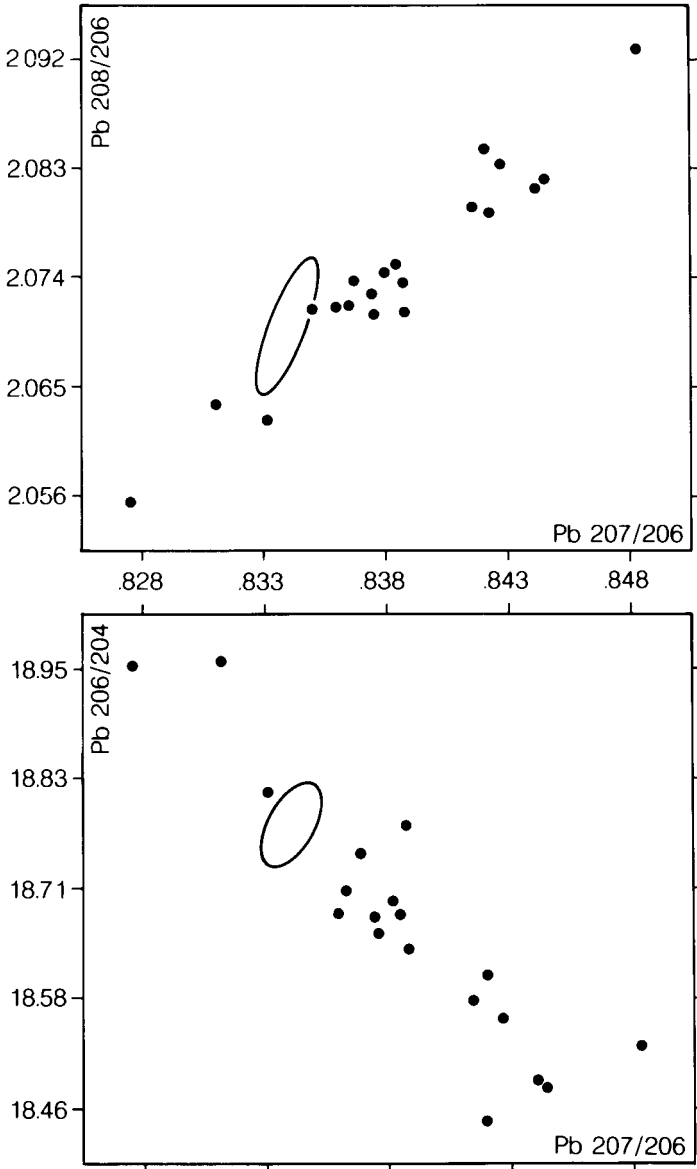
LBA THASOS BRONZES IN RELATION TO THASOS ORE FIELD

Figure 18. The two ordinary bivariate lead isotope diagrams for Late Bronze Age copper alloy artefacts excavated in Thasos by Dr Koukouli-Chrysanthaki, showing that none fall inside the lead isotope field (elliptical outline) for Thasian ores and copper slags.

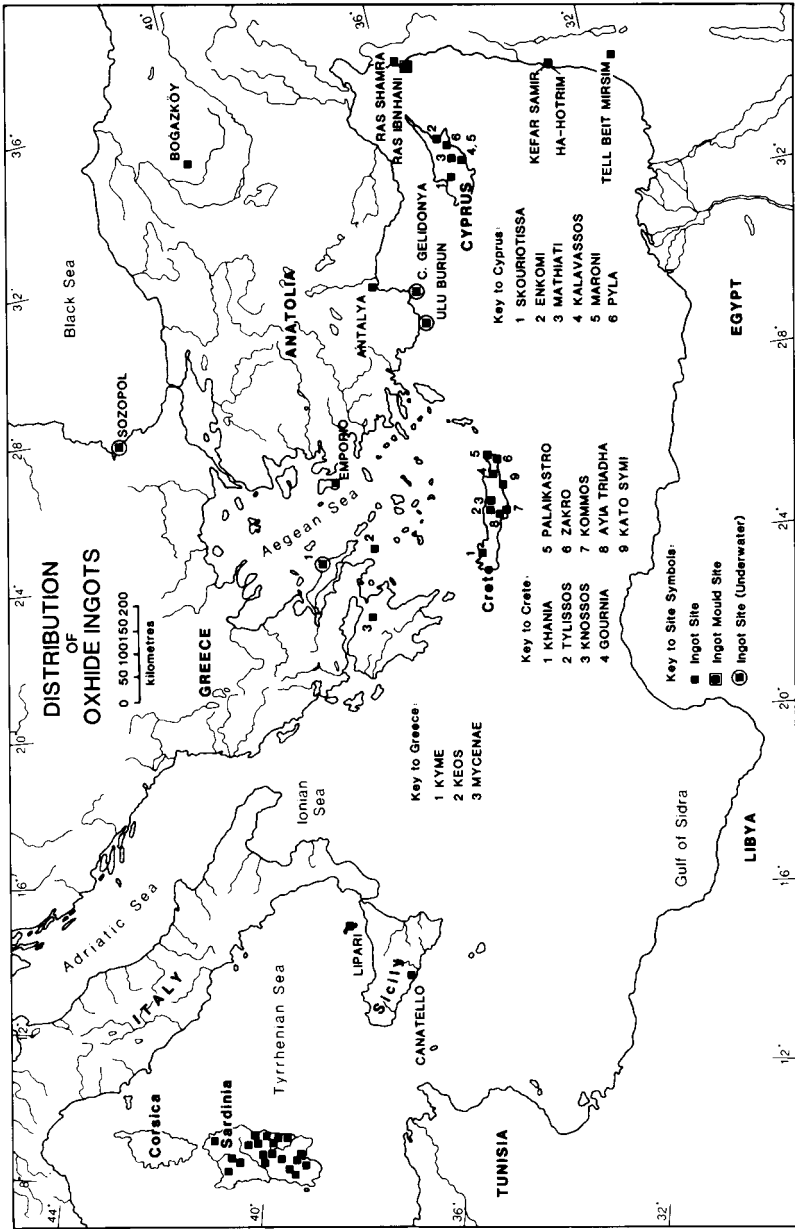


Figure 19. Sketch map showing the distribution of find spots of copper oxhide ingots around the Mediterranean.

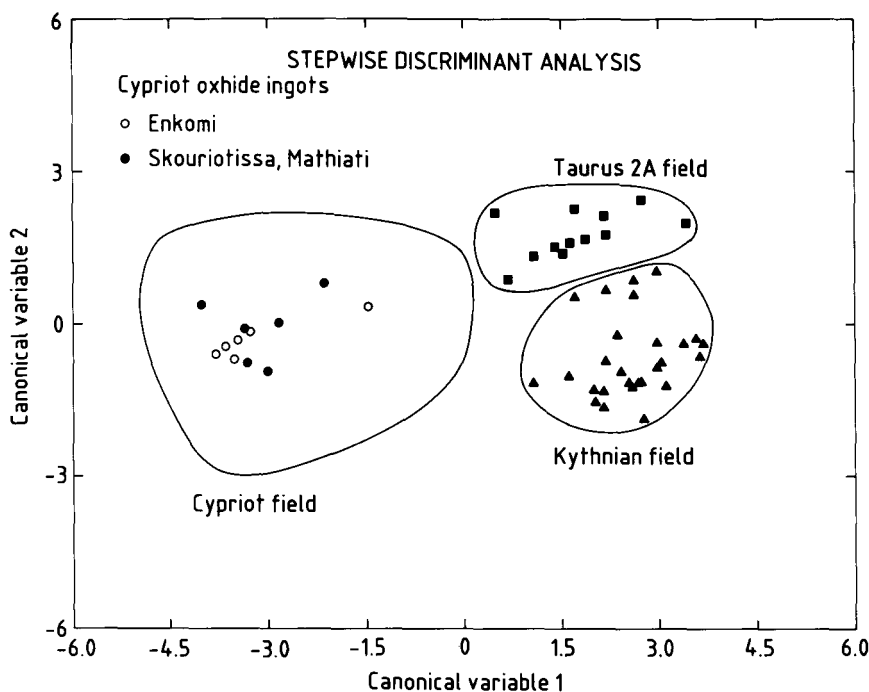


Figure 20. Trivariate discriminant function analysis showing that the lead isotope analyses for the copper oxhide ingots found on Cyprus group with the lead isotope analyses of Cypriot ores.

7M calculated probabilities of the ingots belonging to the field discussed are greater than 70% and mostly greater than 90%]. We also discovered another trace element characteristic of copper extracted from Cypriot copper ores in the Late Bronze Age; it has Au and Ag values falling in a restricted field (Stos-Gale *et al.* 1986, Figure 8; Gale 1989, Figure 29.11). Figure 21 shows that the Cypriot copper oxhide ingots also fall into this Au-Ag field. We have both a lead isotope and a Au/Ag fingerprint for copper produced from Cypriot copper ores in the Late Bronze Age; fingerprints which we can use to trace the movement overseas of Cypriot copper

A surprise was in store when we came to examine the 16th Century BC oxhide ingots found at Hagia Triadha and Tylissos on Crete. Figure 22 shows an ordinary lead isotope diagram for these ingots in relation to the Cypriot lead isotope field; we do not need statistics to see that these were not made of Cypriot copper. Provided that the two sources of copper clearly involved are not isotopically anomalous we can deduce from the Cumming and Richards (1975) model that these ingots were predominantly made of copper coming from a Precambrian ore source unknown in the Aegean but certainly known in Iran or Afghanistan. This may not be so far fetched if we recall that

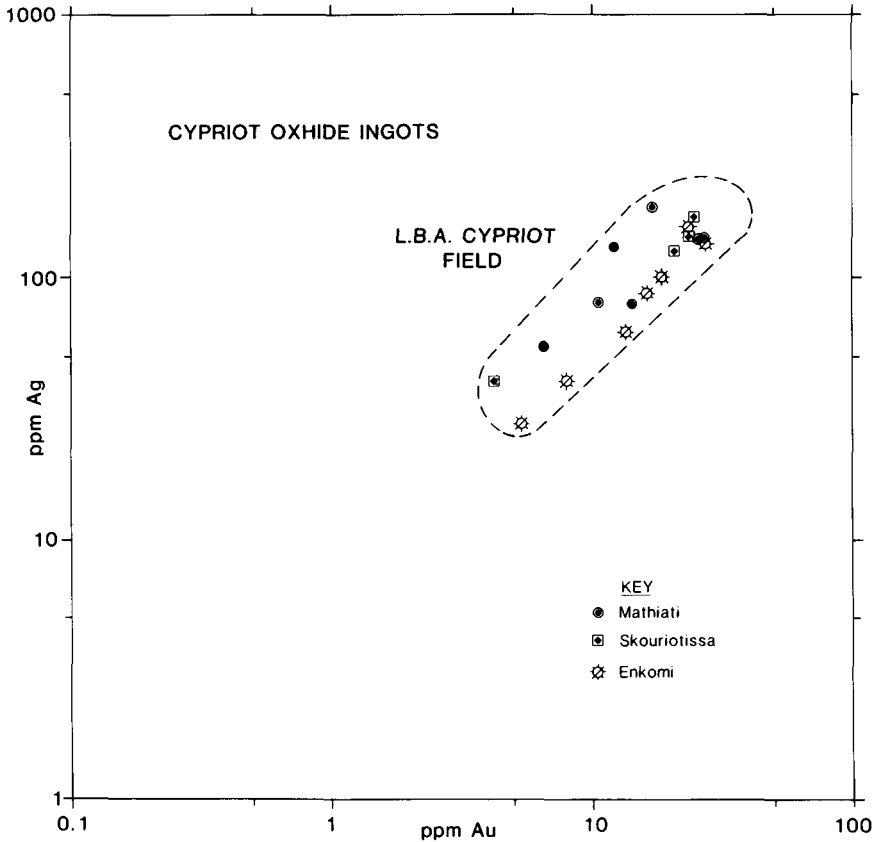


Figure 21. Gold and silver analyses of copper oxhide ingots found on Cyprus, showing that they fall within the field defined by analyses of copper alloys excavated from Late Cypriot archaeological sites.

the Minoan and Mycenaean source of lapis lazuli is thought to have been in Afghanistan (Hermann 1968) and that part of the tin trade to Crete came through Mari (Dossin 1970), possibly also from tin sources in Afghanistan (Shareq and Abdullah 1977; Cleuziou and Berthoud 1982). These are the earliest oxhide ingots known and, since they are not made of Cypriot copper, suggest strongly that the start of the Mediterranean copper trade in this form had nothing to do with Cyprus.

At Mycenae a complete copper oxhide ingot (illustrated by Iakovides 1974, 297) was excavated by Tsountas; twelve fragments of oxhide ingots (illustrated in Gale 1989, Figure 29.15) were excavated by Wace (1953) as part of the Poros Wall Hoard. Figure 23 shows that these 13th Century BC

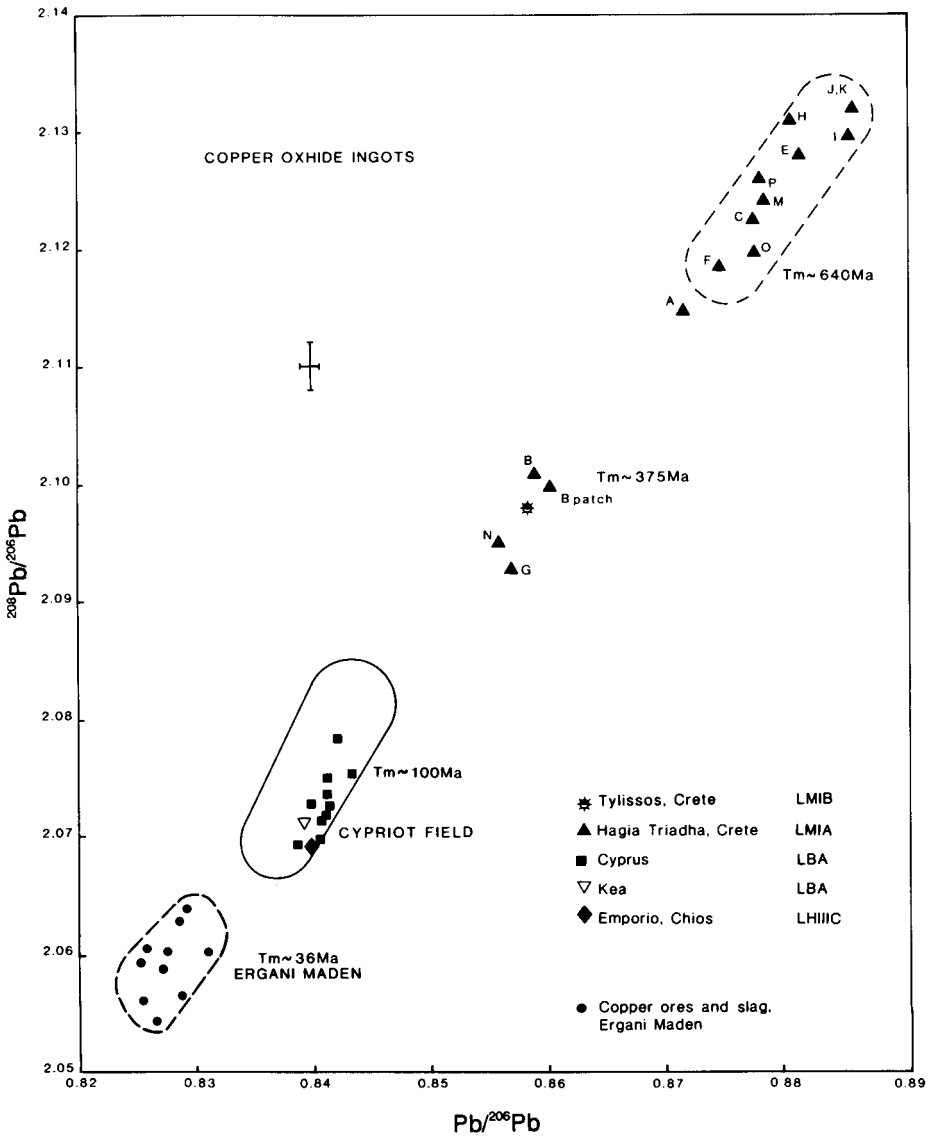


Figure 22. An ordinary bivariate lead isotope diagram showing that the 16th. century BC copper oxhde ingots excavated at Hagia Triadha and Tylissos on Crete fall far outside the Cypriot field.

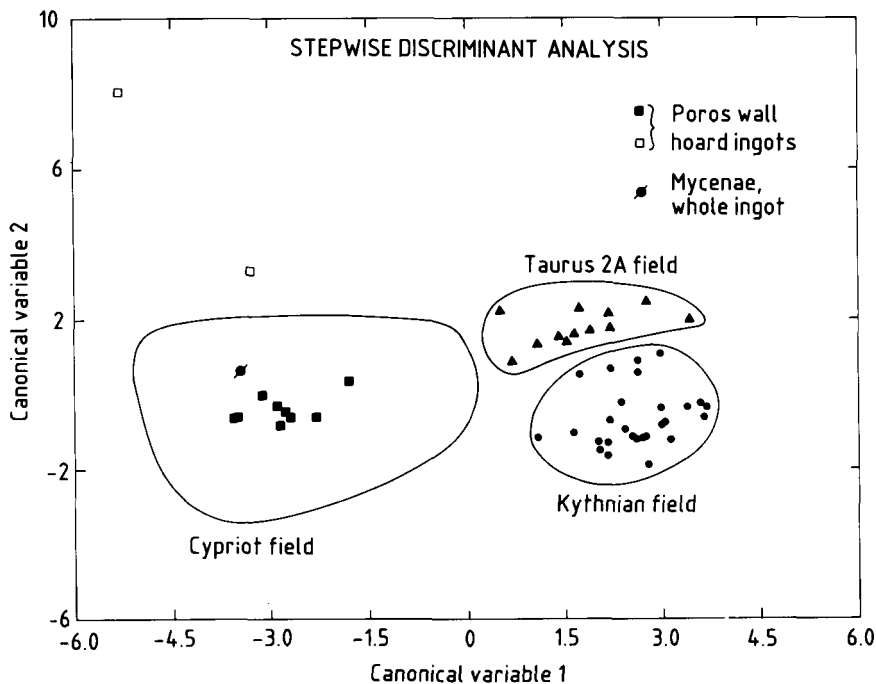


Figure 23. Trivariate discriminant function analysis of lead isotope analyses of the copper oxhide ingot and fragments excavated at Mycenae, showing that they group with copper ores from Cyprus.

oxhide ingot fragments are made of Cypriot copper, so that there was then at least some trade bringing Cypriot copper to Mycenae.

What of the copper oxhide ingots on the two Bronze Age shipwrecks off the coast of Turkey? Figure 24 shows that all 15 analysed ingots from the 13th century BC Cape Gelidonya ship were clearly made from Cypriot copper. Figure 25 shows that Cyprus also supplied the copper for the 4 analysed oxhide ingots from the Late 14th Century BC Ulu Burun ship, though this ship was carrying bun ingots which are not all of Cypriot copper. We can at least be sure that some Cypriot copper was being carried through the Mediterranean in the Late Bronze Age, though to establish quantitatively the overall picture many more ingots from these ships should be analysed.

Turning to Sardinia, there has been a modern tendency to ascribe the large numbers of oxhide copper ingot fragments found there to indigenous manufacture from Sardinian ore deposits, though the earliest discussion of these ingots saw them clearly as an import from the Eastern Mediterranean (Pigorini 1904). Modern scholars, thinking of the copper deposits on Sardinia, have tended to reject an Eastern origin for these oxhide ingots

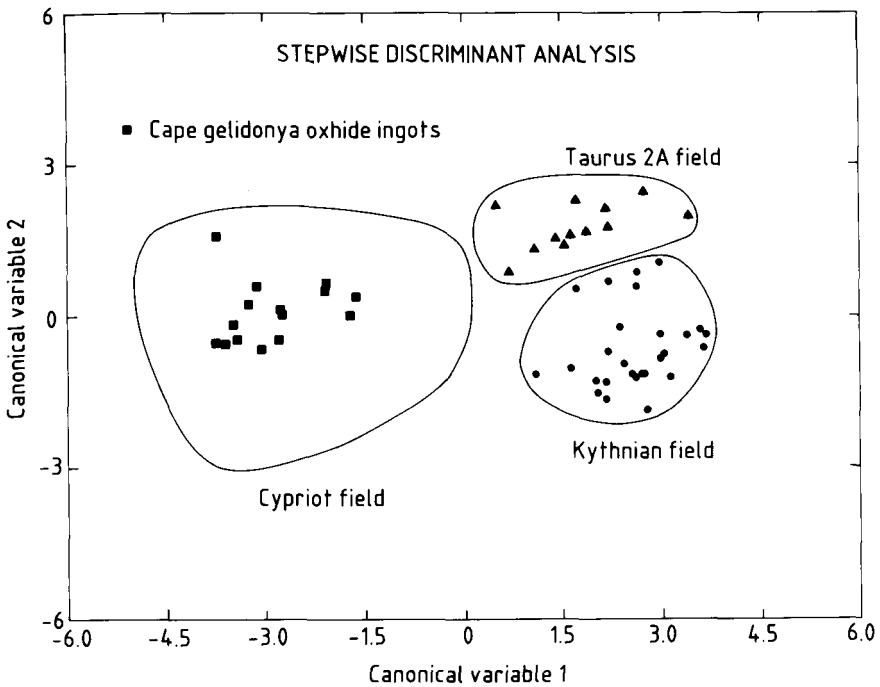


Figure 24. Trivariate discriminant function analysis of lead isotope analyses of copper oxide ingots from the 13th century BC shipwreck excavated by Bass at Cape Gelidonya off the coast of Turkey.

found on Sardinia as an absurd example of “coals to Newcastle”, though the modern example of cheap imports of coal from Poland to Britain should perhaps have given them pause in putting forward so simplistic an argument. Economic considerations aside, there is also the example from the ancient world that the Romans carefully distinguished copper from different ore deposits as of different quality (Pliny, NH.XXXIV.II).

In fact Sardinia is not noted for its reserves of copper, but rather for its deposits of lead. For example, the total production of copper minerals from Sardinia in the period 1851–1948 was 75,000 tonnes, compared with 3,147,000 tonnes of lead minerals in the same period (Sarda 1949, 59). That the reserves of copper in Cyprus are overwhelmingly larger is shown by the fact that in Cyprus one mine (Mavrovouni) exported 131,093 tons of copper concentrates in the year 1938 alone (Bear 1963, 58). Moreover there are only about six small copper mines of any account at all in Sardinia, whilst in Cyprus there are more than 30 cupriferous ore bodies, each vastly larger than any in Sardinia. The probability of ancient discovery of copper in Cyprus is vastly larger than in Sardinia.

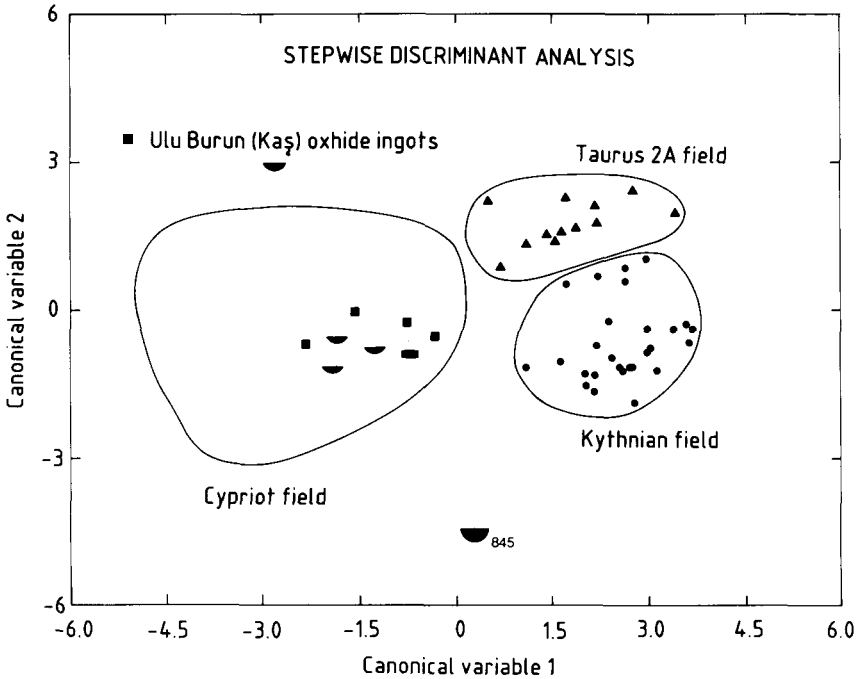


Figure 25. Trivariate discriminant function statistical analysis of lead isotope analyses of copper oxhide and bun ingots from the late 14th. century BC shipwreck excavated by Bass at Ulu Burun off the coast of Turkey.

Our comparative lead isotope analyses of Sardinian copper ore deposits and oxhide ingots found on Sardinia do not support the idea that the ingots were made of Sardinian copper; there is no match (Gale and Stos-Gale 1987). Figure 26 shows that there is a very clear match between the oxhide ingots found on Sardinia and Cypriot copper ores. Moreover Figure 27 shows that the oxhide ingots found on Sardinia display also the range of Au/Ag contents characteristic of Late Bronze Age Cypriot copper. Significantly, analyses depicted in the same figure of characteristic Sardinian Nuragic bronzes [from Santa Maria in Paulis, (Macnamara *et al.* 1984)] both have Au/Ag analyses lying outside the Cypriot field *and* have lead isotope characteristics consistent with their manufacture from Sardinian copper ores (Gale and Stos-Gale 1987, Figure 7.15). Present evidence suggests that we should see the Sardinian ingots as the successful Western extreme of the trade in Cypriot copper exemplified by the ill-fated Cape Gelidonya and Ulu Burun ships.

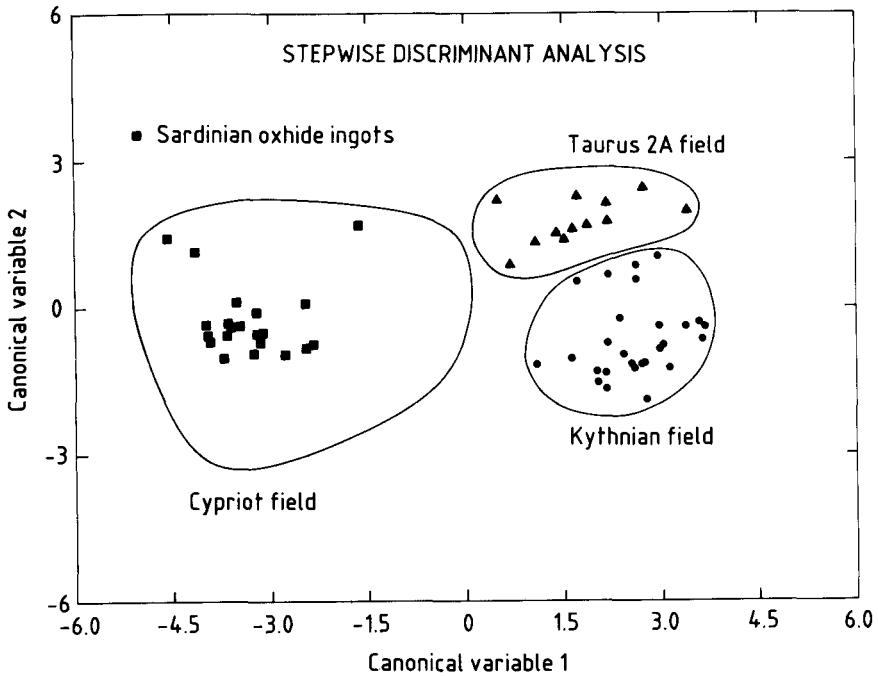


Figure 26. Trivariate discriminant function analysis of lead isotope analyses of copper oxhide ingots found on Sardinia, showing that they group with Cypriot copper ores.

8. The British Academy Project

We conclude by summarising very briefly something about what, within the purview of the British Academy Project, we have discovered about the metals trade in lead, silver, and copper in the Bronze Age Mediterranean by the application of the approaches sketched out in this paper. Figure 28 shows in histogram form the sources of lead and silver found to have been used at different periods of the Bronze Age in the Aegean. Very briefly, the chief sources of lead, silver and the cupellation by-product litharge in the Early Bronze Age were the Cycladic island of Siphnos (Gale 1980) and the mainland mines at Lavrion in Attica (Gale and Stos-Gale 1981a). In the Late Bronze Age Lavrion becomes the dominant source, with an indication that a very little lead and silver may have been coming from the deposits in the Halkidiki (Stos-Gale and MacDonald 1991). A major finding has been the Late Bronze Age trade in lead (and silver?) through the Cycladic islands from Lavrion to Thera and Crete (Gale and Stos-Gale 1981b; Stos-Gale and Gale 1982; Stos-Gale and Gale 1984; Gale *et al.* 1984; Stos-Gale 1985).

Figure 29 is a similar histogram for the sources of copper. It is based

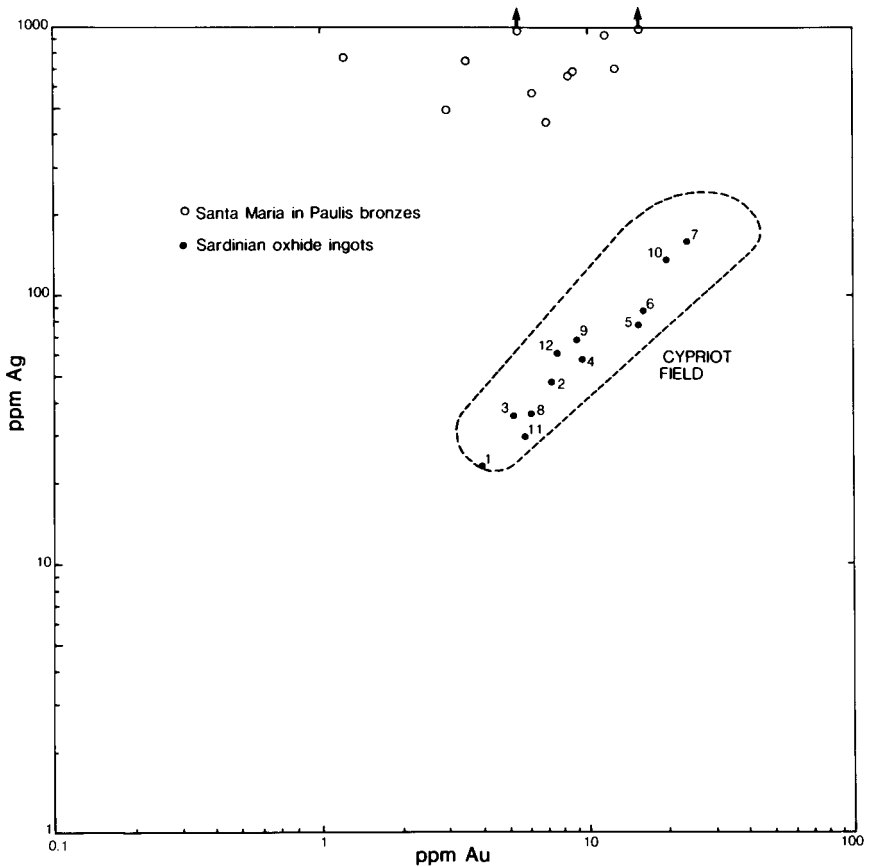
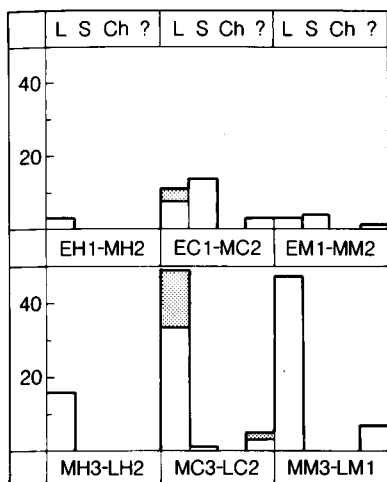


Figure 27. Gold and silver analyses of copper oxhide ingots found on Sardinia, showing that they fall within the field established by analyses of Late Bronze age copper alloy objects excavated in Cyprus.

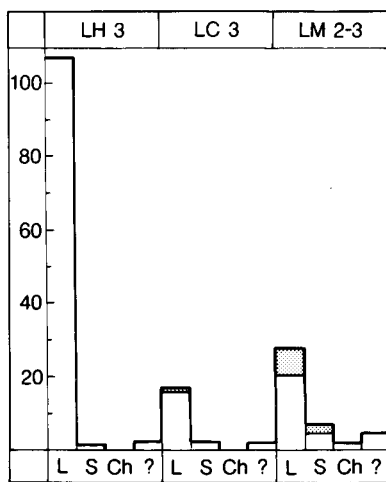
chiefly on the analysis of copper and copper alloy artefacts, rather than the raw material represented by the copper oxhide ingots. It suggests that for copper in the Cyclades and Crete the dominant Early Bronze Age source seems to have been the Cycladic island of Kythnos (Stos-Gale 1988), with a little copper coming even then from Lavrion and other sources. By the Late Bronze Age the dominant source of copper for the Mycenaeans and the Minoans becomes Lavrion in Attica, with a little copper from Cyprus and other sources. When we examine the source of the copper most frequently used for making objects we find that Cyprus does not have the dominance in the Mycenaean and Minoan worlds which examination of the copper oxhide ingots might suggest, a fact in itself which raises interesting archaeological

(a)



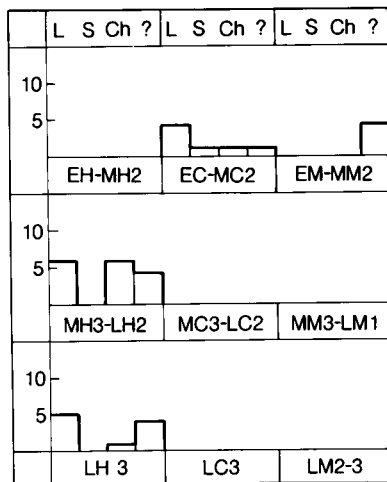
EB-LB2 LEAD SOURCES
(excluding LM2. ▨ = PbO)

(b)



LB 3 LEAD SOURCES
(including LM2. ▨ = PbO)

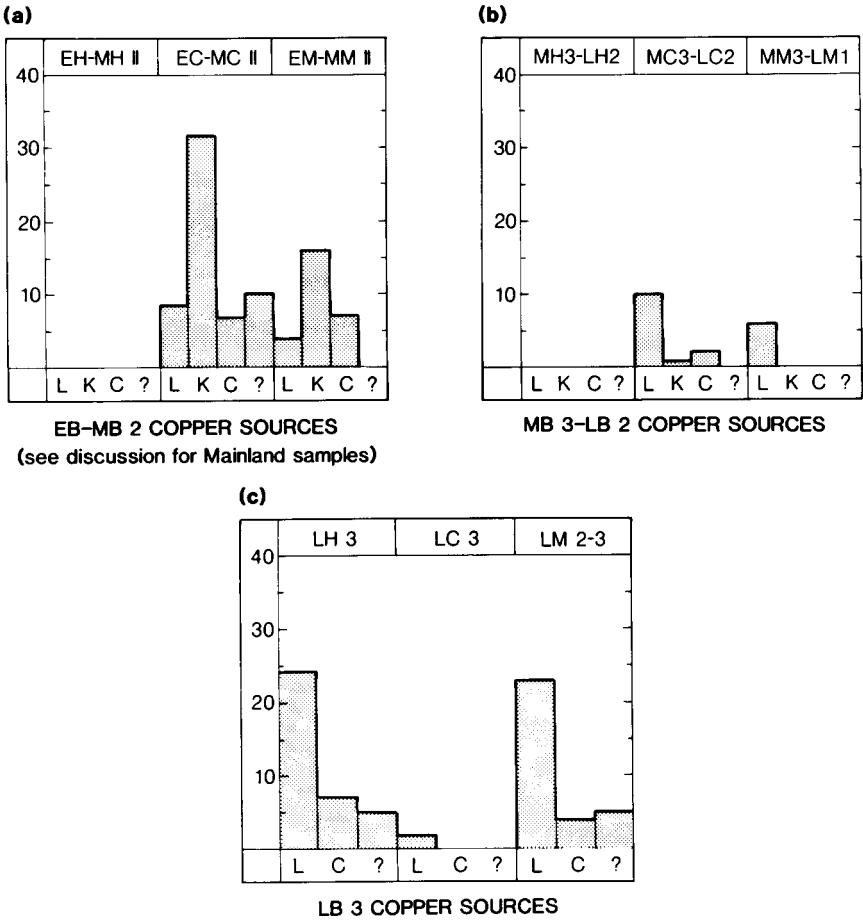
(c)



BA SILVER SOURCES

(L=Lavrion, S=Siphnos, C=?Chalkidiki, ?=Unknown)

Figure 28. A histogram summarising present knowledge of the sources of lead and silver used at various periods in the Aegean.



(L = Lavrion, K = Kythnos, C = Cyprus, ? = Unknown)

Figure 29. A histogram summarising present knowledge of the sources of copper used at various periods in the Bronze Age in the Aegean.

questions. We have of course to admit that many more artefacts need to be analysed to substantiate this view, and to trace through time the evolution of the metals trade in the Aegean. But, in conclusion, we think that it can be claimed that there have been advances in the use of lead isotopes in solving the otherwise intractable problem of provenancing metals and, through their use, in advancing the aims of the British Academy Project. These advances could not have been made without the careful development of the lead isotopic analytical approach to provenancing which has taken place since the

last Royal Society/British Academy symposium on archaeological science in 1969.

The important advances in archaeological lead isotope analyses which have taken place since the beginning of this subject are:

- i) the recognition of the need for close definition of the archaeological problem whose solution is sought,
- ii) the recognition of the importance of field work for examination of ore deposits and metallurgical remains,
- iii) the mineralogical examination of possible ore sources, and the comparison of copper alloy artefacts not with lead/silver deposits but only with ore deposits which could anciently have yielded copper metal, etc.,
- iv) the necessity of taking into account ore geology and isotope geochemistry together with the processes of extractive metallurgy,
- v) ever more accurate mass spectroscopy and low blank chemical separative techniques, and
- vi) more sophisticated data analysis and interpretation.

We hope that we have demonstrated that developments iii), v) and vi) are vital for differentiating ore deposits whose lead isotope compositions are close together, without which it is indeed not possible to assign artefacts to a single ore source.

9. Isotope archaeology

The use of lead isotopic analyses for provenancing metals is but one aspect of a broader subject. The technique of mass spectrometry can now advance archaeological science to such an extent that we believe that we can now speak of a new subject of isotope archaeology, just as about 30 years ago there began the then new subject of isotope geochemistry. Isotope geochemistry and geochronology brought about a revolution in the study of the Earth Sciences. Studies of variations in the isotopic composition of Pb, Sr, Nd, Hf, Ar etc. on the one hand allowed the construction of an absolute chronology (not hitherto available) stretching back for 4,500 million years. In another direction the isotope geochemistry of these and other elements (including Mg, Cr, Ag, Xe, Pu) allowed fundamental questions of petrogenesis and the mode of formation of ore deposits, the origin of the elements and the origin and evolution of the Solar System, etc, to be attacked (Wasserburg 1987; Allegre 1987). We believe that similar studies may have a similar impact upon archaeological science.

Table 1. Mass Spectrometry In Archaeological Science

-
- i) Gas Source Mass Spectrometry:
- (a) C, N isotopic compositions for identification of burned or cooked food residues and for the construction of palaeodiets by the analysis of bone collagen: both can be used also to assess modern animal diets.
 - (b) C, N for tracing ivory back to source (with TIMS for Sr and Pb).
 - (c) S (with Sr) isotopes for provenancing objects made of Gypsum; S isotopes (ores/metal) to aid the provenancing of copper alloy objects.
 - (d) C, O, (with TIMS for Sr and Pb), for provenancing marble.
- ii) Accelerator Mass Spectroscopy:
- (a) Small sample C-14 dating in the range up to 50,000 years BP.
- iii) Thermal Ionisation Solid Source Mass Spectroscopy:
- (a) Pb isotopic compositions of metal ores and metal objects to trace the ore sources from which the metal was smelted to make metal objects; applicable principally to objects made of lead, silver, copper alloys, but also to some iron, tin and zinc objects.
 - (b) Pb isotopic compositions for provenancing (authenticating) glasses, glazes and pigments.
 - (c) Sr isotopic compositions for provenancing obsidian and gypsum.
 - (d) Sr (with Pb, Nd) isotopes (with C, N) for tracing ivory back to source.
 - (e) Cu isotopes may allow direct tracing of copper back to ore source.
 - (f) A combination of Sr, Nd, (Ca, Pb) isotopes may allow better provenancing of ceramics and marble.
 - (g) Mass spectrometric U, Th disequilibrium dating allows certain materials such as corals, speleothems (bones?; molluscs??) to be dated with a precision of 1% in the range 100 years to 400,000 years.
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In applications both to geology and to archaeology there is a broad trichotomy, based on the type of mass spectrometer which has to be used, into gas mass spectroscopy, accelerator mass spectroscopy and thermal ionisation mass spectroscopy. To illustrate something of the scope of this approach Table 1 gives some examples of the applications to archaeological science. They embrace the provenancing of metals (this paper and Gale and Stos-Gale 1985), obsidian (Gale 1981), marble (Duke 1988; Roos *et al.* 1988), gypsum (Gale *et al.* 1988), glass (Brill *et al.* 1979), ivory and bone (van der Merwe *et al.* 1990; Vogel *et al.* 1990), pigments (Keisch and Callahan 1976), etc., dating using C-14 (Purser *et al.* 1982) and the uranium, thorium disequilibrium method (Lawrence Edwards *et al.* 1987), identification of food residues (DeNiro and Epstein 1981; Hastorf and DeNiro 1985), the reconstruction of prehistoric diets (Schoeninger and DeNiro 1984; Walker and DeNiro 1986, van der Merwe 1982; Sealy *et al.* 1991), etc.

The importance to archaeological science of mass spectrometry and

isotope archaeology is perhaps reflected in the papers presented to this discussion meeting, in that no less than 4 out of a total of 15 papers are involved with various aspects of this subject. The importance, and range of application, of isotope archaeology within modern archaeological science has led us to set up at Oxford, with the help of the Science and Engineering Research Council and the University, a new laboratory for research into this subject on a broad front.

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