

### COMMENTS . . . III

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There is no doubt that lead isotope analysis (LIA) generally provides a much better link between ores and metal artefacts than trace element patterns, because the lead isotopic composition is not changed by chemical reactions on the way from ore to metal. Although there are some complexities associated with copper-based alloys (Muhly 1983), their relationship to copper ores is rather straightforward, if one can be reasonably certain that the lead entered the copper metal as an impurity from the ores.

A second important advantage of LIA as compared to chemical analysis is the observation that many ore deposits show rather small variations in isotope ratios of lead (Wagner *et al.* 1989) but not in trace element concentrations. Therefore, there are generally less stringent requirements concerning the number of samples, their size, composition, and location to characterize an ore deposit. Within a relatively short time after the introduction of LIA into archaeology (Brill and Wampler 1965; Grögler *et al.* 1966) a sizeable data base of lead isotope ratios of ores from the Mediterranean therefore has been accumulated which can be used for comparison with archaeological metal objects. This certainly merits a review such as presented by Sayre *et al.* of the present status of lead isotope research concerning major ore deposits in the eastern Mediterranean. However, their article gives the impression that what is needed most is the accumulation of more lead isotope data per ore deposit and a better, that is narrower, definition of source characteristics using multivariate statistical techniques. Allegedly this will eventually lead to better discrimination and a greater probability of positive significance when the lead isotope ratios of an artefact are found to be compatible with an ore source.

There are reasons to believe that the opposite is true. It is highly unlikely that any ore deposit has a unique isotopic signature which would distinguish it from all other possible sources. Already there are many cases of overlap between two or more archaeologically

feasible ore sources. More data will only aggravate this situation as has been noted before (Pernicka *et al.* 1990): 'We are in the very ungratifying situation that more measurements lead to more ambiguity!' The samples discussed by Sayre *et al.* derive from a geographical region that stretches from the Aegean to Iran, comprising at least a dozen major copper deposits, about twice as many major lead and mixed sulphide deposits, and innumerable smaller ore occurrences of variable size. It is obvious that there are bound to be isotopic overlaps so that their differentiation based on LIA alone will be impossible even with the most sophisticated statistical techniques.

Besides these basic limitations it would be desirable to reach consensus or at least consistent treatment of a number of details in the interpretation of LIA.

#### ACCURACY OF LEAD ISOTOPE MEASUREMENTS

Lead isotope ratios can be measured with a precision of the order of 0.05% (one standard deviation) on a routine basis. Although it is a well-known fact in analytical chemistry that an interlaboratory comparison generally yields larger standard deviations than the precision determined in each laboratory this does not appear to be true for LIA. This was demonstrated in practice when a series of objects from Troia was unintentionally analysed at Mainz and Oxford at the same time (Pernicka *et al.* 1984; Stos-Gale *et al.* 1984). For 23 out of 25 metal artefacts the agreement was better than 0.1% for the  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{208}\text{Pb}/^{206}\text{Pb}$  ratios and better than 0.15% for the  $^{204}\text{Pb}/^{206}\text{Pb}$  ratio (Begemann *et al.* 1989). Therefore, error bars of 0.1% in plots of lead isotope *ratios* appear to be realistic.

Sayre *et al.* suggest the use of relative isotope abundances as an alternative to the commonly used isotope abundance *ratios*. The authors fail, however, to demonstrate what the advantages might be. On the contrary, it follows from the law of error propagation that the error associated with isotopic *abundances* of lead in a single sample must be larger than those of abundance *ratios*, because at least three ratio measurements are necessary for the calculation of abundances.

It has therefore to be considered as most unfortunate that Sayre *et al.* (fig. 4, p. 84) chose to define the Laurion source field in an abundance plot of Pb-204 versus Pb-207. This makes it impossible to directly compare this field with earlier interpretations of the same data without tedious calculations, and this without any hint what, in the opinion of the authors, the merit of this different approach might be. Things are made even more complicated in that the authors define the source field of Cyprus in yet different variables so that it cannot even be compared with the other diagrams in the same article. Such a practice will surely reduce the number of scholars who would care for a discussion of LIA or would even pay attention. To quote Chairman Brown (EOS 1991, 72, p. 105): 'I would ask all scientists and engineers to give up something that may be very precious to them: the right to be utterly incomprehensible, to speak in rarified jargon, discernible to no more than a select group of peers.' Incidentally, all diagrams of lead isotope ratios in Sayre *et al.* are not labelled with equidistant tick marks. This may be due to rounding by the computer but it is disturbing, if one wants to check the data, and it certainly does not increase the faith of the uninitiated in those calculations.

## NUMBER OF SPECIMENS THAT CHARACTERIZE AN ORE DEPOSIT

Based only on statistical arguments and totally neglecting the geological evidence, it has been suggested that at least 20 specimens are necessary to define an ore body properly (Reedy and Reedy 1988). This cannot be accepted without further qualification. In geochemistry zoning or compositional variation is known to occur over a very wide length scale ranging from less than a millimetre to hundreds of kilometres. For the purpose of provenance studies, it would be ideal if only regional or districtwise zoning would occur with ores of base metals, that is if one could distinguish 'Anatolian' from 'Iranian' ores or at least 'Cypriot' from 'Central Taurus' ores. For example, it indeed emerges that there are probably three lead isotope provinces discernible in Anatolia (Pernicka *et al.* 1990). Consequently, individual ore deposits within those provinces are likely to overlap considerably in their lead isotope characteristics and other means of discrimination like chemical analyses and field observations may be needed in addition to LIA, if one really aims at identifying a single ore body as a source. Nevertheless, it is certainly worthwhile to analyse as many individual deposits and outcrops as possible within a region in order to obtain as much information on its isotopic variation as possible. It would, however, be absurd to demand 20 analyses from each outcrop if already one analysis can testify to the conformity with other regional ores and if this ties in with the regional metallogeny. Such a situation for instance was encountered in the Troad.

Unfortunately regional zoning is often overlain by districtwise zoning or zoning even within handpieces so that it can happen that the variation within a single ore deposit encompasses the total variation encountered in different regions, as exemplified by the ancient mining site of Feinan in Jordan (Hauptmann *et al.* 1992). Depending on the size of the ore body, 20 samples may not suffice for adequate characterization, especially if they are not carefully selected, taking into account the chemical and mineralogical composition of the specimens and their location within the ore body. Since there is no way to predict the isotopic variance of a deposit, it is dangerous to exclude deviant samples as outliers from a 'source field'. They may merely reflect incomplete sampling. Therefore, the Heidelberg/Mainz group prefers to look for individually matching ore specimens for individual metal objects when discussing the provenance of artefacts (e.g. Pernicka *et al.* 1990). Such a sample-by-sample comparison is certainly the most straightforward technique and requires the least assumptions and pre-selections.

## MATERIALS TO DEFINE A SOURCE FIELD

If isotopically overlapping ore samples derive from different regions then a choice has to be made as to which of the potential ore sources can be considered to be more reasonable than others for the artefact in question. The most obvious constraint is that it should be possible to produce from the ore the same metal as found in the artefact. Many copper ores can thus be excluded as viable sources for lead and silver objects because they simply do not contain these metals in significant concentrations. This is true, for example, for the mining districts of Cyprus, Ergani Maden, Timna, Feinan, and Kythnos. Therefore, it does not make any sense at all to relate, even with a question mark, silver from such diverse sites and periods as Troia II, Mycenae, and late third millennium BC Tello in Mesopotamia with copper ores from Kythnos (Sayre *et al.* p. 92).

Conversely, it is generally not unreasonable to relate copper and bronze objects to lead ores because they frequently occur in so-called mixed sulphide deposits which may contain variable amounts of copper ores. This is the basis of the claim by the Oxford group that Laurion may have supplied some copper besides lead and silver (Gale and Stos-Gale 1982). It is, however, highly questionable that Laurion, predominantly a lead-zinc deposit, was indeed a major copper source for Late Bronze Age Crete as has also been suggested by the same authors.

When a mine is exhausted it is frequently difficult to find good ore samples and one has to settle for low-grade ores. Nevertheless, in order to be of any relevance the analysed samples should at least contain minor amounts of those minerals that contain the metal searched for. The Taurus 1B field as defined by Yener *et al.* (1991), for instance, comprises three 'cassiterite' samples (AON 399, 463, 466) with <0.028% tin (pure cassiterite contains 79% tin), no detectable copper, and lead concentrations below 0.01%. In addition, it includes a slag sample (ASN 459) which contains neither copper nor lead in detectable concentrations. It is not clear what kind of metal objects such ore and slag samples are to be compared with.

In this respect a few words are in order on the significance of slag samples. In principle, slags are the ideal materials for characterizing a production centre, because they average over a much larger sample of ore than what is usually used for lead isotope measurements. In addition, they include all other sources of lead that might have entered the metal, such as fluxes, host rocks, and fuels, and, if they can be dated, they reflect the isotopic composition of the furnace charge that was actually used to produce metal in ancient times. In contradistinction to Sayre *et al.* who prefer to exclude as source materials slags which do not match in isotope ratios the ores from the same mining site, we maintain that slags provide lead isotope characteristics which are even more relevant for the provenance of artefacts than the ore field (Wagner *et al.* 1986).

#### DEFINITION OF A SOURCE AREA

The most valuable aspect of LIA is that certain ore deposits or, for that matter, mineralized regions can be excluded as sources of metal objects if they differ significantly in their lead isotope ratios. If a region has been as well sampled as the Troad or the Aegean it is definitely possible to define an 'Aegean' (Pernicka *et al.* 1984) or even 'Anatolian' (Pernicka *et al.* 1990) ore field. Whether it is reasonable to define such a field would appear to depend on the archaeological problem under investigation. As an example, when it was investigated (Pernicka *et al.* 1984) whether the Early Bronze Age metallurgy of the Troad, and especially the use of tin bronze, was an indigenous development, the lead isotope ratios of many objects, especially from the Troia II period, turned out to be incompatible with Aegean ores which immediately argued against the hypothesis of an indigenous development of tin bronze in the Aegean. The confidence with which this can be concluded depends, of course, on the ratio of analysed to known mineralizations within the Aegean.

For the purpose of most provenance studies, the Troad, the island of Cyprus or the central Taurus region can certainly be considered as potential source areas. Any archaeologist would only be interested to know if, say, the raw material of a silver object found somewhere in the eastern Mediterranean derives from the central Taurus region, the Troad, Chalkidike, or Laurion. It is of secondary interest at most which statistically resolvable subgroup within those mining areas could be identified. Therefore, it would be most logical to draw an

encompassing line around all data from those regions even if not all areas within the fields are equally densely populated at present. The approach adopted by Sayre *et al.* to circumvent the problem of overlap by narrow definition of individual outcrops of ore within a mining region is unyielding for principle reasons and would only lead to fruitless discussions on the selection of outliers.

In summary, LIA can and will continue to contribute a great deal to our knowledge of the trade in early metals but it would certainly be appreciated if, as in thermoluminescence or radiocarbon dating, some general rules as to how to report and interpret lead isotope data could be established. Otherwise it will be impossible, even for interested scholars, to follow the discussion and to comprehend the conclusions.

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