

Lead isotopes and archaeometallurgy

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This special issue presents several studies applying lead isotope analysis (LIA) to address archaeological questions in the field of ancient mining and metallurgy. They were presented during an international workshop, held in Fribourg (Switzerland), June 19–20, 2008, organised in collaboration between the Geosciences Department of the University of Fribourg and the Laboratory of Prehistoric Archaeology and Human Peopling (Department of Anthropology and Ecology) of the University of Geneva with the financial support of the Swiss National Science Foundation. Dr. F. Cattin and Dr. B. Guénette-Beck were in charge of the scientific organization. More than 30 scientists from Belgium, France, Germany, Italy and Switzerland attended

the sessions with the seven key lectures dedicated to different aspects of actual research in the field of LIA.

Over the last few years, several studies of archaeological artefacts using lead isotope analysis have shown a renewed interest in this technique. The workshop aimed to bring together experienced senior researchers and young scientists active in the field to promote contacts and discussions. The idea was to address research questions on four general points:

1. Analytical considerations: possibilities, advantages and shortcomings of various methods like TIMS, ICP-MS and LA-ICP-MS

Since the first application of Pb isotopic data in archaeology, its acquisition has relied on different analytical methods. From this concern, it is legitimate to ask if one lead isotope analysis is equivalent to another lead isotope analysis. First of all, thermal ionisation mass spectrometry (TIMS) was the only method used in Pb isotope measurement until the introduction of high precision multi-collector inductively coupled plasma mass spectrometer, as detailed in the LIA historical perspective of Stos-Gale and Gale. The more recent development of mass spectrometry with laser ablation reduced the sample size and therefore widened new perspectives of applications, with the opportunity to investigate small or precious artefacts and micro-scale sampling in a heterogeneous matrix. The methodological paper of Villa discusses the protocols of all these techniques, with a focus on the one processed at the Laboratory of Isotope Geology at the University of Berne (Switzerland). What comes out of this contribution is that accuracy should concentrate our full attention when acquiring new data and that precision, whatever high or not, may allow distinction between what needs to be differentiated regarding specific archaeological questions.

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2. LIA in regard with the materials studied, namely what are the specificities of applying LIA to metals such as lead/silver, copper and iron, to natural ores as well as to metallurgy-related traces of Pb in sediments?

From the beginning, the determination of the provenance of metal artefacts, based on comparison between metallic products and natural ores, has been one of the main questions addressed by LIA and a major expectancy from archaeologists. The technique was first applied to lead and silver ores and metals and then to copper and only very recently to iron. Despite significant success, it became clear that several pitfalls do exist and that LIA interpretation must be made carefully with a good background for the mineral resources, the history of their exploitation and the technology involved.

Regarding the chaîne opératoire of silver, Guénette-Beck et al. highlights the problem of re-melted silver in the case of coin minting. Actually, the control of the silver content in coins requires its purification by cupellation, leading to lead contamination. Lead artefact studies based on lead isotope analysis have also a specificity in the interpretation of the data. As described in Bode et al., when the mixing of two sources of lead is ascertained by a mixing line in binary diagrams, the mixture ratio can be calculated.

Within the chaîne opératoire of copper, the lead isotope composition may be a concern in the case of alloying. Actually, the signature would no longer reflect that of copper, but an intermediate solution of lead issued from the copper and the admixed lead. It is generally considered that lead in tin to produce bronze does not significantly affect the lead signature of the copper.

Moving on to the analytical methods, as described by Degryse et al., LIA of iron artefacts requires a huge amount of sample material (up to several milligrams). This is far more than for lead, silver and copper sampling, with the exception of native copper, for which lead content is generally inferior to 1 ppm. The complementary use of Sr isotopes by Degryse et al., for which TIMS measurements are more precise than Pb isotopes, is a very interesting way to narrow down the possible sources. In the future, this approach that holds simultaneously elements coming from various elements might be pursued, extended to other isotope systems and applied to different metals. At the same time, the influence of the process on these other elements deserves to be understood.

3. LIA with regard to archaeology, to what kind of archaeological questions can LIA offer new elements for our understanding of the past? How can geo-scientists aid in solving archaeological problems and what are the expectations of the archaeologists in this respect?

Archaeology seeks to understand human populations of the past. Nowadays, there is a panel of related disciplines that can be applied to resolve archaeological problems. At present, the use of LIA remains most often restricted to specific fields in the framework of research programs. It has been fieldwork, however, that has been the most productive in terms of archaeological finds and publications. Despite this, we note a scarcity of site monographs that include lead isotope analysis as part of the interpretation of metallic data. Perhaps the lack of knowledge or understanding regarding the potential of the method and the lack of publications in general archaeological journals are to be taken into account. In our opinion, lead isotope analysis should not be restricted to academic research or specific topics as ingots or coins. Indeed, the interpretation of collected data can provide fieldwork archaeology with valuable information. As shown by Cattin et al., this method improves understanding of a settlement and its cultural background within the final Neolithic of western Switzerland. Guénette-Beck et al. relate the exploitation history of Ag/Pb ore bodies for the production of silver objects in the metallogenic region of the Valais region, Switzerland, from the Late Iron Age to the Late Mediaeval Period. Degryse et al. contribute new elements related to iron trade routes in Sagalassos (Turkey) from the early Roman period to the early Byzantine period. Bode et al. question the motives of the conquest of Germania by the Roman Empire. They seek to demonstrate Roman-related lead mining east of the Rhine river and then to quantify its economic importance.

4. Methods of establishing databases and their practical use

The paper of Stos-Gale and Gale discusses the databases in LIA. The long developments on methodology and history of data acquisition that it contains highlight the heterogeneity of the data within only the Oxford laboratory. In that sense, the wish to merge into a unique database all available Pb isotope data for ores and artefacts—and related information on the analytical process, the precision and accuracy, the site coordinates, the geology of the ore body, the mining evidences, the artefacts period of usage, etc.—cannot be accomplished without a time-consuming, difficult and multidisciplinary approach. Nevertheless, to give a basis for future research in LIA, a non-exhaustive collection of published papers with lead isotope data for ores and artefacts is presented in the “Appendix”. It remains an important concern to make the huge amount of existing data available for all.

Even if this special issue presents only selected LIA approaches, this volume, through the diversity of the papers, already shows a heterogeneous panel in the acquisition and the interpretation of the Pb isotope data.

From these approaches, we conclude that a lead isotope analysis cannot be separated from those technical parameters, since they aimed at answering specific questions with the best appropriate tools for it. In the future, hopefully, the development of analytical methods will help improve protocols towards a less and less destructive Pb isotope acquisition. Simultaneously, analysts, archaeologists and archaeometrists should keep working together on new and complementary ways of research in the quest for material provenances.

As a final remark, we would like to thank the Swiss National Science Foundation for their financial support regarding the organisation of the workshop and for the two doctoral grants that made our research possible.

Appendix

This appendix presents a non-exhaustive collection of published papers with lead isotope data for Cu, Pb/Ag and Fe ores, as much as copper, silver, lead and iron artefacts. The scope region is Europe and the Mediterranean World. It has been constructed based on Guénette-Beck (2005) and Cattin (2008) and significantly increased by M. Bode's, P. Degryse's, N. Gale's and S. Stos' suggestions.

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